

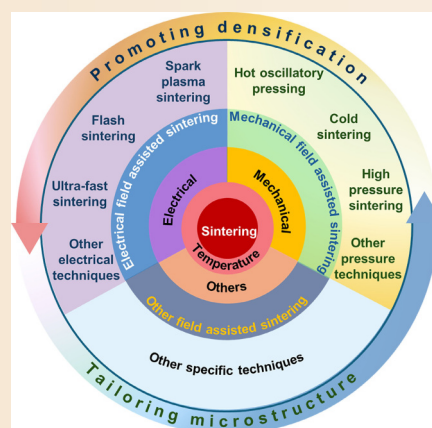
Field-assisted sintering: Overview of thermo-electro-mechanical coupling effects

Dianguang Liu¹, Gang Shao^{2,✉}, Xiaoqian Tai³, Xinyi Li³, Sang-Chae Jeon^{4,✉}, Jing Guo^{3,✉}, Mengting Lin⁵, Wei Ji^{5,✉}, Yan Xiong^{6,✉}, Sha Chen⁶, Fei Zuo^{7,✉}, Salvatore Grasso⁸, Hua-Tay Lin⁷, Xilin Wang^{9,✉}, Xinghua Su^{10,✉}, Ke Ren¹¹, Yifei Liu⁹, Jinling Liu^{12,✉}

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ABSTRACT: Field-assisted sintering technology has revolutionized material processing by integrating temperature, mechanical, electrical, and magnetic fields to achieve unprecedented densification efficiency and microstructural control. Recent advances in techniques such as hot oscillatory pressing, cold sintering, high/ultra-high pressure sintering, spark plasma sintering, ultrafast high-temperature sintering, and flash sintering have enabled the fabrication of previously unattainable materials, including ultrafine-grained ceramics, nanostructured composites, and functionally graded materials. These materials possess exceptional performances under extreme conditions, expanding applications in aerospace, electronics, energy, and biomedicine. However, the rapid development of these methods has exposed limitations in conventional sintering theory, particularly in describing mass transport and interface evolution under multi-physics coupling. This review systematically examines representative field-assisted sintering technologies and discusses their principles, equipment configurations, and application cases. By analyzing current challenges and opportunities, we aim to bridge fundamental understanding with industrial implementation, providing insights for the design and fabrication of next-generation high-performance materials.

KEYWORDS: hot oscillatory pressing; cold sintering; high pressure and ultra-high pressure sintering; spark plasma sintering; ultrafast high-temperature sintering; flash sintering



1 Introduction

The sintering of advanced ceramics faces three core challenges: high energy consumption, instability of microstructure, and the simultaneous optimization of performance limits. Conventional solid-state sintering relies on long-term high-temperature treatment, which not only consumes a large amount of energy but also easily induces abnormal grain growth, resulting in the

deterioration of the material's mechanical properties. At the same time, the thermal, electrical, and mechanical properties of the material often mutually restrict each other, making it difficult to simultaneously break through their theoretical limits. The emergence of multi-field coupling sintering technology provides a disruptive physical approach to solve these deep-seated contradictions. These technologies introduce external energy fields such as mechanical field, electric field, and magnetic field in a

¹School of Integrated Circuits Science and Engineering, Southwest Jiaotong University, Chengdu 611756, China. ²School of Materials Science and Engineering, Zhengzhou University, Zhengzhou 450001, China. ³State Key Laboratory for Mechanical Behavior of Materials & School of Materials Science and Engineering, Xi'an Jiaotong University, Xi'an 710049, China. ⁴School of Materials Science and Engineering, Changwon National University, Changwon 51140, Republic of Korea. ⁵State Key Laboratory of Advanced Technology for Materials Synthesis and Processing, Wuhan University of Technology, Wuhan 430070, China. ⁶School of Materials & Chemical Engineering, Hubei Province Key Laboratory of Green Materials for Light Industry, Hubei University of Technology, Wuhan 430068, China. ⁷School of Electromechanical Engineering, Guangdong University of Technology, Guangzhou 510006, China. ⁸School of Engineering and Materials Science, Queen Mary University of London, London E1 4NS, UK. ⁹Engineering Laboratory of Power Equipment Reliability in Complicated Coastal Environments, Tsinghua Shenzhen International Graduate School, Tsinghua University, Shenzhen 518055, China. ¹⁰School of Materials Science and Engineering, Chang'an University, Xi'an 710061, China. ¹¹Institute of Advanced Structure Technology, Beijing Institute of Technology, Beijing 100081, China. ¹²School of Mechanics and Aerospace Engineering, Southwest Jiaotong University, Chengdu 611756, China.

✉ Corresponding authors. E-mail: G. Shao, gang_shao@zzu.edu.cn; S. Jeon, scjeon@changwon.ac.kr; J. Guo, jingguo19@xjtu.edu.cn; W. Ji, jiwei@whut.edu.cn; Y. Xiong, xiongyan1980@hotmail.com; F. Zuo, zuofei@gdut.edu.cn; X. Wang, wang.xilin@sz.tsinghua.edu.cn; X. Su, suxinghua@chd.edu.cn; J. Liu, liujinling@swjtu.edu.cn

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coordinated manner, fundamentally changing the material transfer mechanism dominated solely by thermal diffusion. For example, oscillatory pressure promotes densification, while its alternating shear stress can effectively inhibit grain growth and strengthen grain boundaries, thereby obtaining extreme performance materials [1]; electric field-assisted sintering (such as flash sintering (FS)) can achieve complete densification within seconds and under conditions far below the conventional temperature, reducing energy consumption by more than 80% [2]. These technologies achieve precise intervention in sintering dynamics and thermodynamics through the spatiotemporal coordination of multiple physical fields, making it possible to prepare ceramic materials with ultra-high thermal conductivity, ultra-high strength, and excellent functional properties at lower temperatures and shorter time, thus breaking the inherent shackles of energy consumption, microstructure, and performance in conventional processes.

1.1 Basic concept

Sintering is a material processing technique that involves heating powder or compacted green bodies to temperatures below the melting point of their primary constituents to achieve densification [3]. The fundamental principle relies on thermally driven mass transport mechanisms—including surface diffusion, grain boundary diffusion, and volume diffusion—which promote neck formation between particles, leading to reduced porosity and enhanced mechanical strength through microstructural evolution. This process entails complex physicochemical changes such as solid-state reactions, grain growth, and pore transformation, making it a critical step in powder metallurgy, ceramic

manufacturing, and post-processing of additively manufactured components.

Modern sintering technology has advanced from conventional pressureless sintering to multi-parameter coupled processes that enable precise control over temperature, mechanical fields, electric fields, magnetic fields, and atmospheric conditions. These advanced techniques facilitate the production of nanostructured materials (e.g., with grain sizes < 100 nm) and provide essential means for achieving tailored properties such as graded porosity and controlled crystallographic texture. The performance of sintered products is highly dependent on the synergy between process parameters—such as temperature, time, and pressure—and the characteristics of the raw materials, making sintering a decisive factor in determining the final density, microstructure, and mechanical properties of the material.

1.2 Development history

The historical evolution of sintering technology can be divided into four key stages [3–5]:

1.2.1 Ancient empirical stage (Prehistory–18th Century)

Sintering technology dates back to the ancient empirical stage (prehistory–18th century), primarily represented by pottery firing. Artisans relied entirely on experience to control the sintering process—for example, the firing temperature of Neolithic painted pottery ranged between approximately 800 and 1000 °C. Sintering at this stage mainly achieved physical bonding between particles through high temperatures, lacking systematic scientific guidance. This experience-based sintering process laid the foundation for later developments and reflected early human exploration and understanding of material processing techniques.

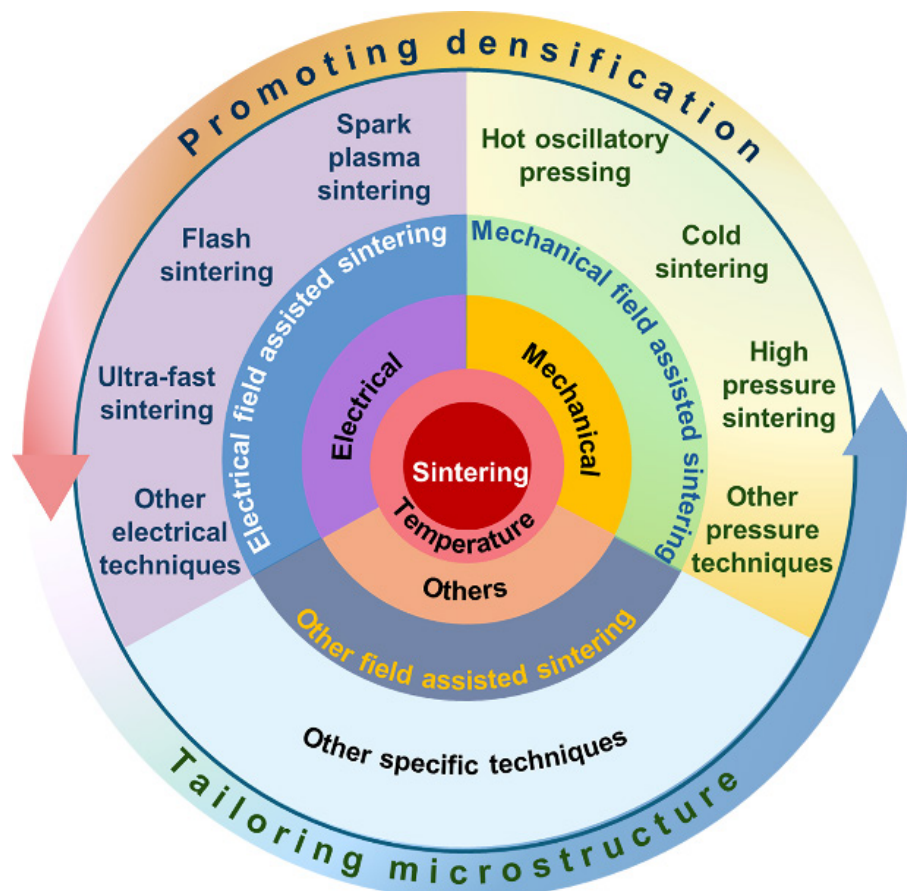


Fig. 1 Schematic diagram of field-assisted sintering.

1.2.2 Scientific enlightenment stage (19th Century–1940s)

During the scientific enlightenment stage (19th century to the 1940s), research on sintering mechanisms began to systematize with the rise of powder metallurgy. In 1909, Coolidge achieved the first industrial-scale sintering of tungsten powder [6], marking the transition of sintering from an empirical practice to a science. In 1945, Frenkel established the first viscous flow sintering model [7], providing a theoretical basis for understanding sintering processes. Subsequently, in 1949, Kuczynski's diffusion-controlled neck growth equation further refined the theoretical system of sintering kinetics [8]. These groundbreaking advances established a solid scientific foundation for modern sintering technology.

1.2.3 Technological innovation stage (1950s–1990s)

During the technological innovation stage (1950s–1990s), sintering technology advanced through both theoretical refinement and process innovation. In 1961, Coble's grain boundary diffusion model laid the theoretical foundation for modern sintering kinetics [9]; in the 1970s, the emergence of microwave sintering technology introduced a new, highly efficient sintering process [10]; in the 1990s, spark plasma sintering units from Japan became commercially available [11]. These major breakthroughs not only advanced the theoretical framework of sintering but also drove innovations in sintering technology, establishing a solid foundation for modern material preparation techniques.

1.2.4 Intelligent sintering stage (21st century–present)

Since the 21st century, sintering technology has entered an intelligent sintering stage. Driven by interdisciplinary integration, a series of groundbreaking technologies have emerged: Nanosintering enables precise control of grain sizes below 100 nm [12]; field-assisted sintering techniques (such as flash sintering [13] and ultrafast sintering [14]) significantly improve sintering efficiency through coupled external fields; digital intelligent sintering uses AI algorithms to optimize process parameters [15], advancing sintering toward intelligent operation. Together, these innovations are ushering sintering technology into a new era of precision, high efficiency, and intelligence.

1.3 Fundamental knowledge of sintering

1.3.1 Solid-state and viscous sintering

During solid-state sintering, mass transport can occur through at least six distinct paths, which determine the sintering mechanisms. In practice, under any given sintering condition, more than one mechanism may be active, and the presence of multiple mechanisms complicates the analysis of sintering rates and the identification of dominant sintering mechanisms. The most significant consequence introduced by grain boundaries is the growth of grains and pores during sintering, a process commonly referred to as coarsening. Coarsening provides an alternative pathway for reducing the free energy of the powder system, thereby diminishing the driving force for densification.

Compared to solid-state sintering, the analysis of viscous sintering appears relatively straightforward in principle. Matter transport occurs through a viscous flow mechanism, although the specific pathways of this flow are not explicitly defined. Instead, the equations describing matter transport are derived from the energy balance concept proposed by Frenkel [7], wherein the rate of energy dissipation due to viscous flow is equated to the rate of energy gain from surface area reduction. Models based on this

energy balance concept have generally proven successful in describing the sintering kinetics of amorphous materials.

(I) Driving force for sintering

Like all other irreversible processes, sintering is accompanied by a reduction in the system's free energy. The factors responsible for this free energy decrease are generally referred to as the driving forces for sintering. Three potential driving forces exist: the curvature of the particle surfaces, an externally applied pressure, and a chemical reaction.

(i) Surface curvature

In the absence of external stress and chemical reactions, surface curvature becomes the driving force for powder sintering. To understand why, consider analyzing one mole of powder composed of spherical particles with radius a . The number of particles in this powder is (Eq. (1)):

$$N = \frac{3M}{4\pi a^3 \rho} = \frac{3V_m}{4\pi a^3} \quad (1)$$

where ρ denotes the particle density (assuming the particles are pore-free); M is the molecular weight; V_m is the molar volume. The total surface area of the particle system is (Eq. (2)):

$$S_A = 4\pi a^2 N = \frac{3V_m}{a} \quad (2)$$

If γ_{sv} is the specific surface energy of the particles (i.e., the surface energy per unit area), then the surface free energy associated with this system of particles is (Eq. (3)):

$$E_s = \frac{3\gamma_{sv} V_m}{a} \quad (3)$$

where E_s represents the reduction in surface free energy if one mole of particles were to form a fully dense body, and it provides the kinetic basis for sintering. Assuming $\gamma_{sv} = 1 \text{ J/m}^2$, $a = 1 \text{ }\mu\text{m}$, and $V_m = 25 \times 10^{-6} \text{ m}^3$, then $E_s = 75 \text{ J/mol}$.

(ii) Applied pressure

In the absence of chemical reactions, when pressure is applied, the external pressure typically provides the dominant contribution to the driving force during most stages of the heating process (as in hot pressing and hot isostatic pressing). Although surface curvature also contributes to the driving force, its contribution is generally much smaller than that provided by the external pressure in most practical situations. The external pressure performs work on the particle system, and for one mole of particles, this work can be approximated as Eq. (4):

$$W = p_a V_m \quad (4)$$

where p_a represents the applied pressure and V_m denotes the molar volume. W signifies the driving force for densification resulting from the application of external pressure. For $p_a = 30 \text{ MPa}$ (a typical value of applied stress in hot pressing) and $V_m = 25 \times 10^{-6} \text{ m}^3$, $W = 750 \text{ J}$.

(iii) Chemical reaction

Theoretically, if a chemical reaction can enhance the densification process during sintering, it can provide a driving force for sintering. The associated change in free energy can be expressed by Eq. (5):

$$\Delta G^0 = -RT \ln K_{eq} \quad (5)$$

where R is the gas constant (8.3 J/mol); T is the absolute temperature; K_{eq} is the equilibrium constant of the reaction. Assuming $T = 1000 \text{ K}$ and $K_{eq} = 10$, then $\Delta G^0 \approx 20,000 \text{ J/mol}$. This energy reduction is significantly greater than the driving

force generated by the applied stress. In practice, chemical reactions are rarely intentionally utilized to drive densification in advanced ceramics, as the simultaneous occurrence of chemical reactions and sintering makes microstructural control extremely challenging.

(2) Stages of sintering

Sintering is generally considered to progress through three consecutive stages, designated (1) the initial stage, (2) the intermediate stage, and (3) the final stage. In some sintering analyses, an additional stage (stage 0) is considered, describing the instantaneous contact between particles where they first coalesce due to elastic deformation-induced reduction in interfacial surface energy [16]. Each stage represents a time interval or density range during which the microstructure is considered relatively well defined and stable. For polycrystalline materials, Fig. 2 illustrates the idealized geometries proposed by Coble [9], which serve as representative models for the three stages.

(i) Initial stage

The initial stage is primarily characterized by rapid neck growth between particles through diffusion, vapor transport, plastic flow, or viscous flow. During this stage, significant differences in surface curvature are eliminated, and shrinkage (or densification) occurs concurrently with neck growth as a result of densification mechanisms. For powder systems composed of spherical particles,

the initial stage can be described as the transition between Figs. 2(a) and 2(b). This stage is generally considered to persist until the neck radius reaches approximately 0.4–0.5 times the particle radius. For powder systems with an initial density of 0.5–0.6 times the theoretical density, this corresponds to a linear shrinkage of 3%–5% or an increase in density to approximately 0.65 times the theoretical density when densification mechanisms dominate.

(ii) Intermediate stage

The intermediate stage commences when the pore morphology reaches an equilibrium state governed by surface tension and interfacial tension while maintaining pore connectivity. In sintering models, this microstructure is typically idealized as a sponge-like network of interconnected pore channels along grain edges, as illustrated in Fig. 2(c). Densification is assumed to proceed primarily through the reduction of pore cross-sectional areas. Eventually, these continuous pore channels become unstable and fragment, leaving isolated pores—this transition marks the beginning of the final stage. The intermediate stage generally encompasses the majority of the sintering duration and is considered complete when the density reaches approximately 0.9 of the theoretical value.

(iii) Final stage

In one of the simplest descriptions, the final stage commences when the pores pinch off and become isolated at the grain corners, as depicted in the idealized structure of Fig. 2(d). Within this simple description, the pores are assumed to shrink continuously and may disappear entirely.

Some of the main parameters associated with the three idealized stages of sintering [17] are summarized in Table 1, and examples of the microstructures (planar section) of real powder compacts in the initial, intermediate, and final stages are shown in Fig. 2.

Table 1 summarizes the key parameters involved in the three idealized sintering stages, and Fig. 3 presents schematic diagram of powder compacts during the initial, intermediate, and final stages.

(3) Mechanisms of pressureless sintering

The sintering of polycrystalline materials is achieved through diffusional transport of matter along specific paths, which determine the sintering mechanisms. Matter is transported from regions of higher chemical potential (referred to as matter sources) to regions of lower chemical potential (referred to as matter sinks). In polycrystalline materials, there exist at least six distinct sintering mechanisms, as illustrated in Fig. 4, which depicts a system of two sintering particles. All these mechanisms contribute to interparticle bonding and neck growth, thereby increasing the strength of the powder compact during sintering. However, only certain mechanisms lead to shrinkage or densification, and a distinction is typically made between densifying and nondensifying mechanisms. Surface diffusion, lattice diffusion from the particle surface to the neck, and vapor transport (mechanisms 1, 2, and 3) promote neck growth without densification and are thus classified as nondensifying mechanisms. Grain boundary diffusion and lattice diffusion from the grain

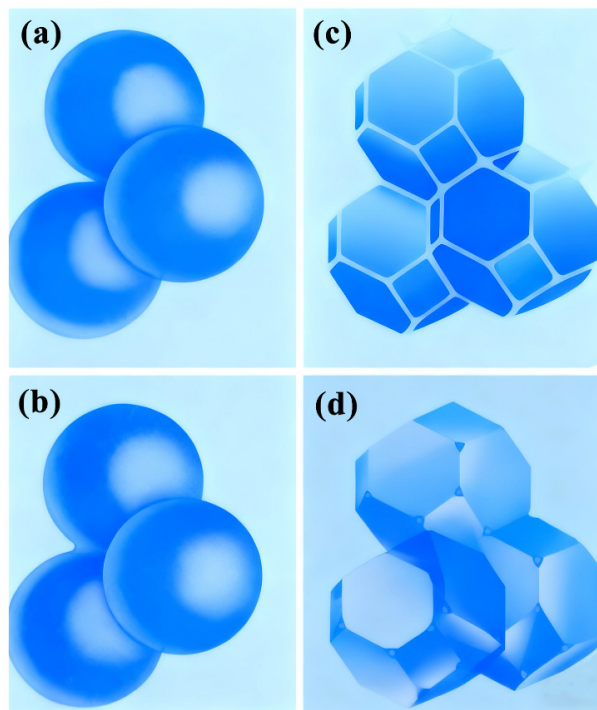


Fig. 2 Idealized models of three stages in the sintering process: (a) initial stage, (b) near the end of the initial stage, (c) intermediate stage, (d) final stage. Reproduced with permission from Ref. [9], © The American Institute of Physics 1961.

Table 1 Typical parameters at different stages of solid-phase sintering

Stage	Typical microstructure	Relative density	Typical theoretical model
Initial	Quick neck growth at particle contacts	< 0.65	Two equal-sized spheres touching each other
Intermediate	Steady-state pore morphology featuring interconnected porosity	0.65–0.90	Tetrahedron with edge-aligned cylindrical pores of equal radius
Final	Steady-state pore morphology featuring disconnected porosity	> 0.90	Tetrahedron with equal-sized spherical pores positioned at the corner

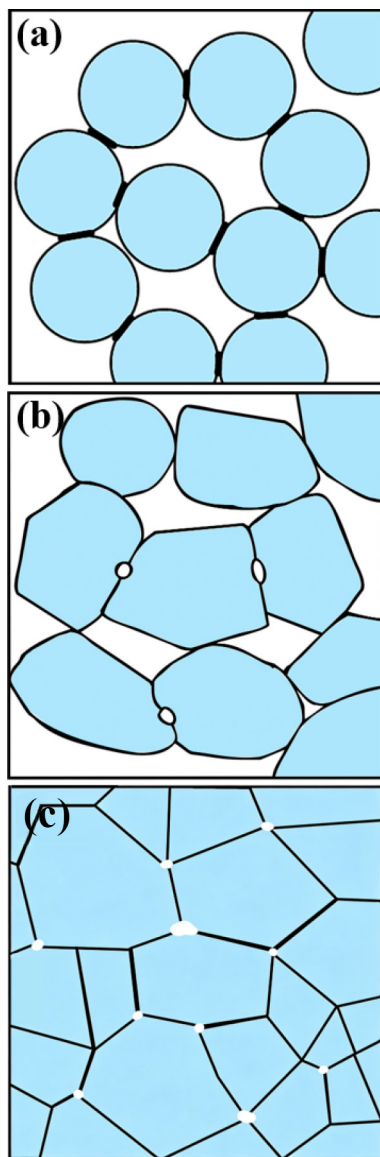


Fig. 3 Schematic diagrams corresponding to (a) initial stage, (b) intermediate stage, and (c) final stage of sintering.

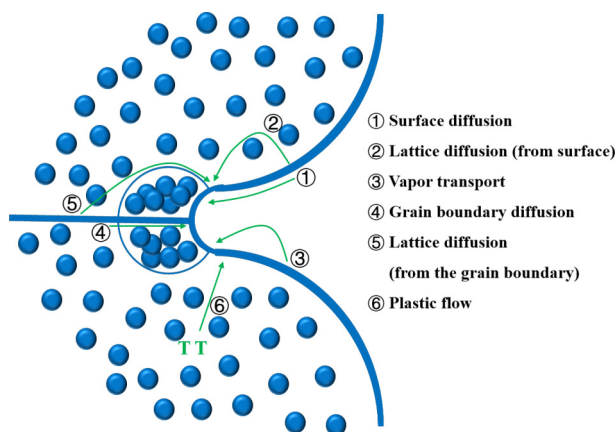


Fig. 4 Six different mass transfer pathways in sintering: (1) surface diffusion, (2) lattice diffusion from the surface, (3) vapor transport, (4) grain boundary diffusion, (5) lattice diffusion from the grain boundary, and (6) plastic flow.

boundary to the pore (mechanisms 4 and 5) represent the most important densification mechanisms in polycrystalline ceramics. Diffusion from the grain boundary toward the pore facilitates both neck formation and the densification process. Plastic flow via dislocation motion (mechanism 6) also contributes to neck formation and densification, although this phenomenon is more common in the sintering of metal powders. Nondensifying mechanisms cannot be simply disregarded, as their occurrence reduces the curvature of the neck surface (i.e., the driving force for sintering), thereby diminishing the rate of densifying mechanisms.

For amorphous materials (glasses), sintering proceeds through viscous flow induced by particle deformation, leading to neck growth and densification. Figure 5 exemplifies the sintering schematic diagram of two glass spheres via viscous flow. The pathways of material transport are not distinctly defined. The geometric changes accompanying viscous flow are relatively complex, requiring strict simplifying assumptions when formulating the material transport equations. For the sintering of spheres, Fig. 6(a) illustrates two possible flow fields for viscous sintering [18]. While the pattern shown on the left may be expected in actual systems, simulation results (Fig. 6(b)) align well with the flow field on the right. Table 2 summarizes the sintering mechanisms in polycrystalline and amorphous solids [17].

(4) Mechanisms of sintering for hot pressing

The sintering mechanisms discussed previously remain active during hot pressing, although nondensifying mechanisms can generally be neglected, as they are not enhanced by the applied stress, whereas densifying mechanisms are significantly intensified. Due to this selective enhancement of densifying mechanisms over nondensifying ones, hot pressing provides an effective approach for identifying the dominant densification mechanisms. However, the applied stress may also activate additional mechanisms and introduce certain complexities, making it essential to characterize the mechanisms that ultimately govern the process.

During the initial stage, particle rearrangement contributes to powder densification, although it is difficult to analyze. In the intermediate and final stages, grain boundary sliding becomes essential to accommodate shape changes governed by diffusion-controlled processes. As shown in Fig. 7, a representative unit of the system (e.g., four idealized grains with a hexagonal shape) must be capable of simulating the overall shape evolution of the powder compact. Since the diameter of the hot-pressing die is fixed and powder compression occurs primarily along the loading direction, the grains undergo flattening. This shape change must be accommodated by grain boundary sliding. It is important to note that grain boundary sliding and diffusional mass transport are not independent mechanisms. They occur sequentially, meaning that the slower process controls the overall densification rate. In summary, the dominant mechanisms typically active during hot pressing include lattice diffusion, grain boundary diffusion, plastic deformation via dislocation motion, viscous flow, particle rearrangement, and grain boundary sliding.

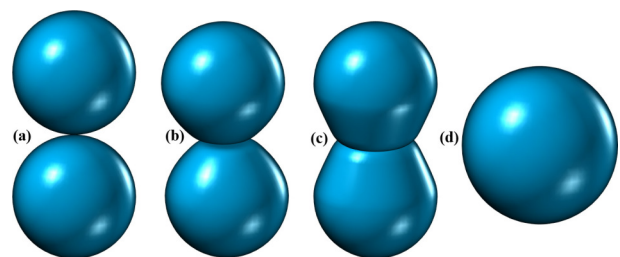


Fig. 5 The sintering schematic diagrams of two glass spheres via viscous flow: (a) initial point contact, (b) neck growth (short time), (c) neck growth (long time), (d) final form.

When the applied pressure is significantly greater than the driving force generated by curvature (which corresponds to a common practical situation), the densification rate during hot pressing can be expressed as Eq. (6) [17]:

$$\frac{1}{\rho} \frac{d\rho}{dt} = \frac{HD\phi^n}{G^m k T} P_a^n \quad (6)$$

where H is a numerical constant; D is the diffusion coefficient of the rate-determining species; ϕ is the stress intensification factor; G is the grain size; k is the Boltzmann constant; T is the absolute temperature; m and n correspond respectively to the grain size exponent and the stress exponent. Table 3 provides the values of m and n , along with the appropriate diffusion coefficients corresponding to various mechanisms.

Due to the enhanced densification mechanisms and suppressed grain growth, hot pressing provides an effective approach for investigating densification mechanisms. According to Eq. (6) and Table 3, plotting the densification rate (at constant density) against P_a allows determination of the exponent n and thereby identification of the dominant densification mechanism. For

commonly used hot-pressing pressures (typically 10–50 MPa), data from numerous ceramic materials indicate $n \approx 1$, suggesting that diffusion mechanisms govern densification [20,21]. This finding is consistent with the strong bonding in ceramics (limiting dislocation motion) and the prevalent use of fine powders (favoring diffusion mechanisms). The dominant densification mechanism may vary with applied pressure, temperature, and grain size. For instance, finer powders and lower temperatures tend to favor grain boundary diffusion over lattice diffusion due to the larger grain size exponent ($m = 3$) and generally lower activation energy compared to lattice diffusion ($m = 2$). However, for some ceramic materials, higher observed n values suggest the involvement of dislocation mechanisms [22].

(5) General features of grain growth

(i) Grain growth and coarsening

The term “grain growth” describes the increase in grain size within a single-phase solid or the growth of matrix grains in a solid containing second-phase particles. At sufficiently high temperatures, grain growth occurs in both dense and porous polycrystalline solids. For the conservation of matter, the total volume of individual grains must remain constant; therefore, an increase in average grain size is necessarily accompanied by the disappearance of some grains, typically smaller grains. In porous solids, both grains and pores generally increase in size, while the

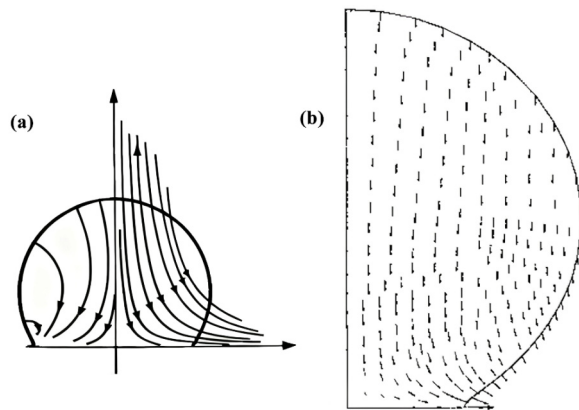


Fig. 6 (a) Schematic diagrams of two possible flow fields for viscous sintering of spheres. Reproduced with permission from Ref. [18], © D. Reidel Publishing Company 1969. (b) Simulations via finite element analysis indicate that the flow field on the right is more realistic. Reproduced with permission from Ref. [19], © the American Ceramic Society 1990.

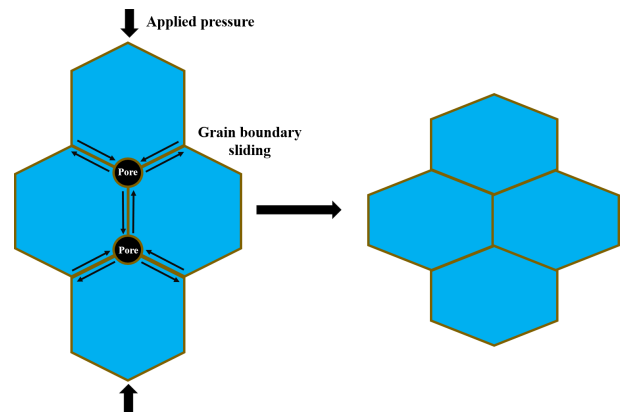


Fig. 7 Schematic diagrams illustrating the morphological evolution of grains during hot pressing.

Table 2 Mechanisms of sintering in polycrystalline and amorphous materials

Type of material	Mechanism	Source of matter	Sink of matter	Densifying or nondensifying
Polycrystalline	Surface diffusion	Surface	Neck	Nondensifying
	Lattice diffusion	Surface	Neck	Nondensifying
	Vapor transport	Surface	Neck	Nondensifying
	Grain boundary diffusion	Grain boundary	Neck	Densifying
	Lattice diffusion	Grain boundary	Neck	Densifying
	Plastic flow	Dislocations	Neck	Densifying
Amorphous	Viscous flow	Unspecified	Unspecified	Densifying

Table 3 Mechanisms of hot pressing and the related exponents and diffusion coefficients

Mechanism	Grain size exponent	Stress exponent	Diffusion coefficient
Lattice diffusion	2	1	Lattice diffusion coefficient
Grain boundary diffusion	3	1	Grain boundary diffusion coefficient
Plastic deformation	0	≥ 3	Lattice diffusion coefficient
Viscous flow	0	1	—
Grain boundary sliding	1	1 or 2	lattice diffusion coefficient or grain boundary diffusion coefficient
Particle rearrangement	—	—	—

number of pores decreases. Due to the extensive interactions between grains and pores, the microstructural evolution is considerably more complex than in dense solids. Often, the term “coarsening” is used to describe the process combining grain growth and pore growth.

(ii) Occurrence of grain growth

In the widely accepted schematic representation, grain boundaries are depicted as disordered regions between two crystalline domains (grains), as illustrated in Fig. 8. High-resolution TEM reveals that the thickness of the grain boundary regions ranges from 0.5 to 1 nm. Grain growth occurs when atoms (or ions) diffuse from one side of the boundary to new positions on the other side over distances smaller than an interatomic spacing. This process leads to the growth of one grain at the expense of another. Atoms move more readily from the “convex” side to the “concave” side of the boundary than in the reverse direction because the chemical potential of atoms on the convex side is higher than that on the concave side. The net flux resulting from this asymmetry causes the boundary to migrate toward its center of curvature (Fig. 9).

(iii) Driving force for grain growth

Atoms located at grain boundaries possess higher energy than those within the grain interior, consequently imparting a specific energy to the grain boundary, typically ranging from 0.2 to 1.0 J/m². The driving force for grain growth arises from the reduction in free energy associated with the decrease in total grain boundary area.

(iv) Normal and abnormal grain growth

Grain growth in ceramics is typically categorized into two types: (1) normal grain growth and (2) abnormal grain growth, sometimes referred to as exaggerated grain growth, discontinuous grain growth, or, in the context of metals, secondary recrystallization. In normal grain growth, the average grain size increases, but the sizes and shapes of the grains remain within a relatively narrow range. As a result, the grain size distribution at a later time is quite similar to that at an earlier time (Fig. 10(a)). This characteristic is often described as the grain size distribution being scaled or self-similar, meaning its shape remains invariant over time. In abnormal grain growth, certain larger grains form and grow at a faster rate than the surrounding matrix of smaller grains. The grain size distribution may change significantly, resulting in a bimodal distribution (Fig. 10(b)). Consequently, the time invariance of the distribution is lost in this case. Eventually, these larger grains impinge upon one another and may revert to a normal size distribution.

The terms “normal grain growth” and “abnormal grain growth” are also used to describe grain growth in porous ceramics, but with consideration of the interactions between pores and grains. Thus, normal grain growth in porous ceramics is characterized

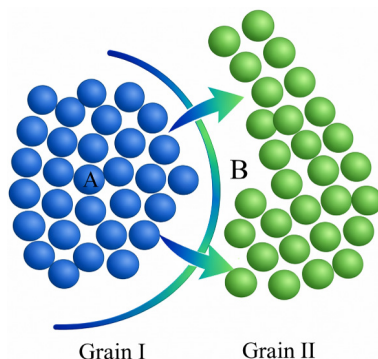


Fig. 8 Classical schematic diagram of a grain boundary.

not only by the features seen in dense materials but also by the presence of pores located at grain boundaries. When grain boundaries break away from pores, leaving them trapped within grains, this typically indicates the occurrence of abnormal grain growth. Although ceramic microstructures often exhibit nearly equiaxed grain shapes, there are cases where normal or abnormal grain growth proceeds anisotropically, resulting in elongated or plate-like grain morphologies. In such instances, this type of growth is referred to as anisotropic grain growth.

1.3.2 Liquid-phase sintering

Liquid phase sintering is particularly effective for ceramic materials such as Si₃N₄ and SiC that have highly covalent bonding and are difficult to densify through solid-state sintering. Moreover, this process is also crucial when the cost of solid-state sintering is too high or when a very high manufacturing temperature is needed. Typically, the purpose of liquid phase sintering is to enhance the densification rate, accelerate grain growth, or produce specific grain boundary properties. The distribution of the liquid phase formed during sintering and the resulting solid phases after cooling are crucial for achieving the desired properties of the sintered material. Usually, the amount of liquid formed during sintering is small, generally not exceeding a few volume percent (vol%), which makes precise control of the liquid composition difficult. In some systems, such as alumina, the amount of liquid phase may be so minimal that it is difficult to detect. Consequently, many studies originally thought to involve solid-

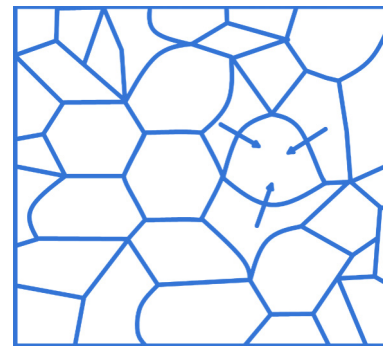


Fig. 9 Migration of the grain boundary toward its curvature center.

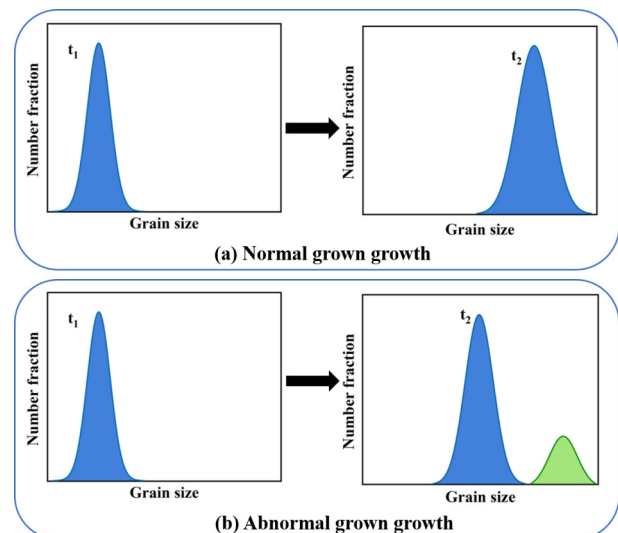


Fig. 10 Microstructure with an initially unimodal grain size distribution: (a) normal grain growth, (b) abnormal grain growth.

state sintering actually involved liquid silicate phases, as later confirmed by careful high-resolution transmission electron microscopy examination.

(1) Driving force for densification-capillary force

Compared to solid-state sintering, the presence of a liquid phase enhances densification through two primary mechanisms: (1) enabling more efficient rearrangement of particulate solids and (2) facilitating matter transport within the liquid. Figure 11 presents a schematic diagram of an idealized model for liquid-phase sintering. In liquid-phase sintering, if the liquid wets and spreads to cover the solid surfaces, the particles become separated by liquid bridges. Interparticle friction is significantly reduced, allowing the particles to rearrange more readily under the compressive capillary stress exerted by the liquid. In solid-state sintering via mechanisms such as grain boundary diffusion, a key parameter controlling the diffusion rate is the product of the grain boundary diffusion coefficient D_{gb} and the grain boundary thickness δ_{gb} . In liquid-phase sintering, the corresponding parameter is the product of the solute diffusion coefficient D_L in the liquid and the liquid bridge thickness δ_L . Since δ_L is typically many times larger than δ_{gb} , and diffusion in liquids is considerably faster than that in solids, the liquid provides a highly efficient pathway for material transport.

Assuming the liquid wets and covers the solid surfaces, the solid-gas interface in the particle system will disappear, and pores will form within the liquid. The reduction in the liquid-gas interfacial area provides the driving force for system shrinkage (densification). For a spherical pore of radius r within the liquid, the pressure difference across the curved interface is given by the Young-Laplace equation (Eq. (7)):

$$\Delta p = -\frac{2\gamma_{lv}}{r} \quad (7)$$

where γ_{lv} is the specific surface energy of the liquid-vapor interface. The pressure within the liquid is lower than that inside the pore, generating a compressive capillary stress on the particles. This compressive stress induced by the liquid is equivalent to subjecting the system to an external hydrostatic pressure, with its magnitude given by Eq. (7). Assuming $\gamma_{lv} \approx 1 \text{ J/m}^2$ and $r \approx 0.5 \mu\text{m}$, then $\Delta p \approx 4 \text{ MPa}$. This pressure level can provide a considerable driving force for sintering.

(2) Stages of liquid-phase sintering

In most liquid-phase sintering systems, the chemical reactions between the particulate solid and the liquid are relatively weak, making the interfacial energy a decisive factor in determining the sintering rate. Under such conditions, as illustrated in Fig. 12, liquid-phase sintering is generally considered to proceed through a sequence of key stages [23,24]:

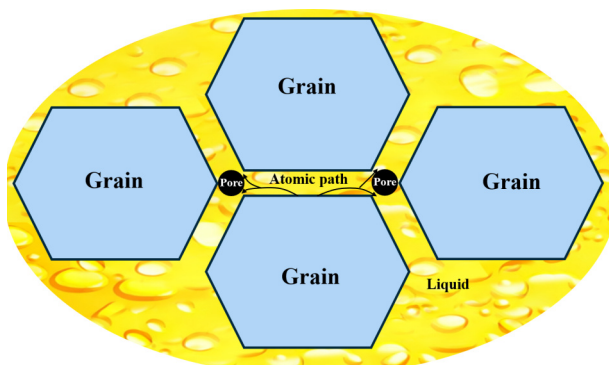


Fig. 11 Schematic diagram of an idealized model for liquid-phase sintering.

(i) Stage 1: rearrangement and liquid redistribution

As the green body temperature increases, solid-state sintering may occur prior to liquid formation, leading to significant densification in some systems. Assuming good wettability between the liquid and the particulate solid, the capillary forces exerted by the liquid on the particles will induce further densification. When the solid dissolves into the liquid, the particles rapidly rearrange to form a higher packing density, releasing liquid to fill the interparticle pores. Capillary stresses cause the liquid to redistribute among particles and into small pores, resulting in additional rearrangement. Due to their higher solubility in the liquid, the contact points between agglomerates dissolve, while fragments undergo rearrangement. Throughout this process, the dissolution of sharp edges smoothens the particle surfaces, reducing the interfacial area and facilitating system rearrangement. Initially, the rearrangement process proceeds rapidly, but as densification progresses, the viscosity of the system increases, leading to a continuous decrease in the densification rate.

(ii) Stage 2: solution precipitation

As densification from rearrangement slows, factors dependent on the solid solubility in the liquid and the diffusion rate through the liquid begin to dominate, initiating the second stage known as "solution precipitation". The solid dissolves at solid-liquid interfaces where the chemical potential is high, diffuses through the liquid, and precipitates at other particle locations where the chemical potential is lower. One type of dissolution site occurs in the wetted contact regions between particles, where the chemical potential is elevated due to capillary stresses from the liquid or externally applied stresses. Precipitation takes place away from the contact regions. In systems with a distribution of particle sizes, matter can also be transferred from smaller particles to larger ones via diffusion through the liquid, a process known as Ostwald ripening. The net result is microstructural coarsening. Densification via the solution-precipitation mechanism is accompanied by changes in grain shape. When the amount of liquid is fairly large, the grains typically become rounded. However, with a small amount of liquid, the grains develop smooth surfaces and adopt polyhedral shapes to achieve more efficient packing, a process referred to as grain shape accommodation.

(iii) Stage 3: Ostwald ripening

The final stage of liquid phase sintering is governed by the

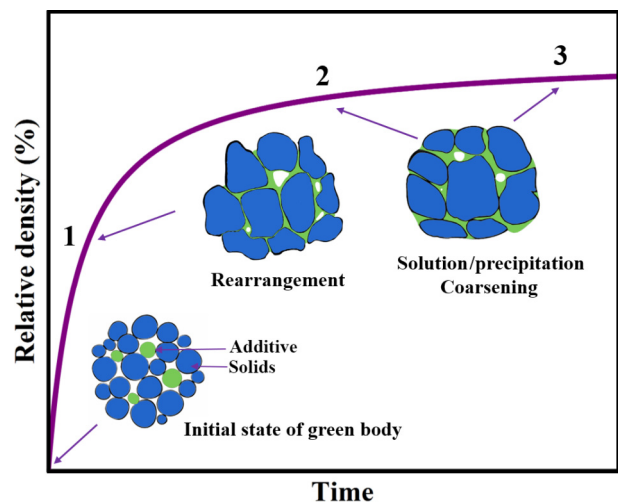


Fig. 12 Schematic diagram of the evolution of a green body during liquid-phase sintering.

densification of the solid particle skeleton network. This process proceeds slowly due to the large diffusion distances in the coarsened structure and the rigid framework of contacting solid particles. Ostwald ripening dominates during this final stage; if trapped gases are present in the residual pores, they can cause pore growth, leading to swelling of the structure. Coarsening is accompanied by grain shape accommodation, which enables the grains to pack more efficiently. Liquid may be released from more efficiently packed regions and may flow into isolated pores, thereby contributing to densification.

The stages of liquid phase sintering are illustrated in Fig. 12. The extent to which each stage contributes to densification depends on the volume fraction of the liquid phase, leading to multiple variations in this conceptual diagram. When the volume fraction of the liquid phase is high, full densification can be achieved through the rearrangement stage alone. On the other hand, in systems with low liquid content—a common situation in many practical cases—the solid skeleton inhibits densification, making solution precipitation and final-stage sintering necessary for further densification. It should be noted that in the classic three-stage theory of liquid-phase sintering, there are usually no clear and absolute time nodes between the three stages. It is more accurate to view these three stages as a continuous evolution process that partially overlaps and is dominated by different densification mechanisms.

(3) Hot pressing with a liquid phase

When a liquid phase is present, densification can also be combined with hot pressing. The chemical potential of atoms at the contact surfaces increases with applied stress, thereby enhancing matter transport from the contact regions to the pores and resulting in an accelerated densification rate. Bowen *et al.* [25] employed liquid-phase hot pressing to densify Si_3N_4 powder with MgO as a liquid-forming additive. MgO reacts with the SiO_2 -rich layer on the surface of the Si_3N_4 powder, generating a MgSiO_3 - SiO_2 eutectic liquid at approximately 1550 °C. The measured densification kinetics were consistent with a solution-precipitation mechanism, wherein diffusion through the liquid phase is the rate-controlling step. This process can be described by an equation analogous to Coble's equation for grain boundary diffusion-controlled solid-state sintering, provided that (1) the grain boundary thickness δ_{gb} is replaced by the liquid layer thickness δ_{l} ; (2) the grain boundary diffusion coefficient D_{gb} is replaced by the solute diffusion coefficient in the liquid D_{l} , and (3) the solid-vapor interfacial energy γ_{sv} is replaced by the liquid-vapor interfacial energy γ_{lv} . When the applied pressure p_a is much greater than the capillary pressure due to the liquid meniscus, the densification rate can be expressed as Eq. (8) [17]:

$$\frac{1}{\rho} \frac{d\rho}{dt} = \frac{AD_{\text{l}}\delta_{\text{l}}\Omega}{G^3kT} p_a \phi \quad (8)$$

where A is a geometric constant (with a value of 47.5 for the intermediate stage of sintering); Ω is the atomic volume of the rate-controlling species; G is the grain size; k is Boltzmann constant; T is the absolute temperature; ϕ is the stress intensification factor.

1.4 Characterization of new techniques

Modern sintering technology has entered a new stage characterized by intelligentization and multi-field coupling, exhibiting the following core features.

1.4.1 Multi-field synergistic control

Modern sintering has achieved breakthroughs in multi-field synergistic control. By innovatively integrating mechanical fields (e.g., hot oscillatory pressing), electric fields (e.g., flash sintering),

and magnetic fields (e.g., microwave sintering), sintering efficiency has been significantly enhanced. This multi-field coupling technology not only substantially reduces sintering temperatures (e.g., flash sintering can lower temperatures by 200–400 °C) but also shortens conventional sintering cycles from hours to seconds (e.g., ultrafast high-temperature sintering takes only 10–30 s), representing a revolutionary advancement. This approach breaks the thermodynamic limitations of conventional sintering and enables precise control over material microstructures through nonequilibrium effects induced by external fields, opening new pathways for preparing high-performance ceramics and composites.

1.4.2 Precise microstructural control

Major progress has been made in precise microstructural control. Nanosintering technology enables accurate adjustment of grain sizes below 100 nm through precise process parameter control, providing an effective means for producing ultrafine-grained materials. At the same time, innovative dynamic pressure-assisted sintering enhances mechanical properties by introducing beneficial crystal defects. This precise microscale control capability not only achieves an excellent strength–toughness balance but also enables the customization of material properties, offering new high-performance material solutions for high-end applications such as aerospace and electronic devices. These advances mark a new era in sintering technology, transitioning from macroscale control to nanoscale precision.

1.4.3 Intelligentization and automation

Modern sintering technology has achieved groundbreaking advances in intelligence and automation. The adoption of multistation automatic transfer sintering equipment has significantly improved production efficiency. Meanwhile, artificial intelligence-based self-learning systems can autonomously generate optimized sintering process schemes tailored to the characteristics of different new materials. This intelligent production mode not only greatly enhances manufacturing efficiency but also enables dynamic optimization of process parameters through real-time data acquisition and analysis, substantially improving product consistency and reliability. This technological breakthrough provides a new intelligent solution for the materials manufacturing sector and accelerates the rapid development of sintering technology toward digitalization and flexibility.

1.4.4 High efficiency and energy savings

Modern sintering technology has achieved significant breakthroughs in high efficiency and energy conservation. Novel sintering processes, such as flash sintering and cold sintering, significantly reduce energy consumption by optimizing energy utilization efficiency. Meanwhile, innovative modular equipment design enables rapid switching between different sintering modes (e.g., hot isostatic pressing, HIP), greatly enhancing production flexibility and energy efficiency. These technological advances not only reduce production costs but also promote the transition of sintering processes toward green and sustainable manufacturing, providing the industry with material processing solutions that are both economical and environmentally friendly.

1.5 Paper structure

This paper provides a systematic exposition of novel field-assisted sintering technologies, primarily focusing on two major technical systems: mechanical field-assisted sintering and electric field-

assisted sintering. Mechanical field-assisted sintering encompasses techniques such as hot oscillatory pressing, cold sintering, and high/ultrahigh pressure sintering. Electric field-assisted sintering includes methods such as spark plasma sintering, ultrafast high-temperature sintering, and flash sintering. This paper offers a comprehensive introduction and in-depth analysis of these field-assisted sintering technologies from multiple perspectives, including technological advancements, sintering equipment, underlying mechanisms, technical advantages, typical material systems, and future prospects.

2 Mechanical field-assisted sintering techniques

2.1 Hot oscillatory pressing

2.1.1 Introduction to hot oscillatory pressing

(1) Concept of hot oscillatory pressing

As an important part of engineering materials and functional materials, advanced ceramic materials have broad application prospects in new energy, communication electronics, semiconductors, aerospace, and other industrial fields [26–29]. However, the wide application of advanced ceramic materials still faces many problems and challenges, among which reliability, density, and strength are the main constraints [30]. How to achieve rapid densification of materials at a low sintering temperature and prepare completely porosity-free ceramics with uniform structure, fine grains, and grain boundary strengthening has always been the goal pursued by ceramic material scientists [31]. For a long time, researchers have carried out extensive and in-depth research on the preparation and property control of nanoceramic powder, the ceramic sintering process and the basic theory of sintering, new sintering technology, and new equipment. The results show that the innovation of sintering technology is the key factor to further improve the properties of ceramic materials. In this regard, researchers have successfully developed a variety of ceramic sintering technologies, one of which is a very important type of mechanical field-assisted sintering technology, such as cold sintering [32], hot pressing [33], and spark plasma sintering [34]. All of these mechanical field-assisted sintering techniques use static constant pressure. While the introduction of static pressure in the sintering process facilitates the elimination of pores and enhances the density of ceramics, it remains challenging to entirely eliminate pores within advanced ceramic materials. Consequently, there are still constraints associated with the fabrication of ultrahigh strength, high toughness, and high reliability materials. Consequently, researchers in the field of materials continue to devote considerable effort to the development of novel mechanical field-assisted sintering technologies. It is exciting that Xie's group [35] at Tsinghua University pioneered the substitution of dynamic oscillatory pressure for static constant pressure during hot pressing and named this pressure sintering technology oscillatory pressure-assisted hot pressing (OPAHP) technology. Later, it was abbreviated as oscillatory pressure sintering (OPS) or hot oscillatory pressing (HOP).

(2) Development of hot oscillatory pressing

In 2014, Xie's group [35] published the first international paper on hot oscillatory pressing, which attracted widespread attention from the academic community. To date, over 90 papers on the subject of hot oscillatory pressing have been published. The majority of research on hot oscillatory pressing is conducted in China, with no reports from other countries due to the lack of

development of the corresponding sintering equipment. The technique of hot oscillatory pressing has been employed in the preparation of a diverse range of ceramic materials, including oxide ceramics [1,35–38], nitride ceramics [39–41], carbide ceramics [42–45], boride ceramics [46], high entropy ceramics [47], WC-based cemented carbide [48–50], W-based refractory metal [51–53], Mo-based refractory metal [54–56], Ni-based superalloy [57–59], Cu-based composite materials [60], and high entropy alloys [61–63]. All the results were found to be satisfactory. The microstructure of the materials is evidently optimized, and the mechanical properties are markedly enhanced. The results demonstrate that hot oscillatory pressing is a universal technique.

2.1.2 How to perform hot oscillatory pressing

Hot oscillatory pressing equipment is similar to hot pressing equipment; the main distinction is how the pressure is applied. There are two main design schemes for the application of oscillatory pressure. One is the application method of oscillatory pressure developed by Xie's group [64]. The fundamental principle underlying this technique is to superimpose an oscillatory pressure with adjustable frequency and amplitude under a relatively large constant pressure. The schematic of the applied oscillatory pressure of Xie's group is shown in Fig. 13(a). This design incorporates two distinct pressure control systems: a static pressure system and a dynamic pressure system. This design scheme has lower requirements for a dynamic pressure system and lower equipment costs. The other is the application method of oscillatory pressure designed by An's group [65], the fundamental principle of which is to completely replace constant pressure with oscillatory pressure rather than superposition of pressure (Fig. 13(b)). Under this method of pressure application, the dynamic pressure has a wider adjustable range, making it more suitable for fundamental research. This design requires only a dynamic pressure system, not a static pressure system. This design scheme has higher requirements for a dynamic pressure system and higher equipment costs.

2.1.3 Sintering parameters

The parameters of hot oscillatory pressing are similar to those of hot pressing, including temperature, time, and pressure. However, the definition and control of the pressure parameters in hot oscillatory pressing are more complex. It is decomposed into three interrelated dynamic components, including the median pressure, the amplitude pressure, and the frequency, as shown in Fig. 13(b). The median pressure refers to the average of the maximum and minimum oscillatory pressures, with units of MPa, usually ranging from 0 to 100 MPa. The amplitude pressure refers to the absolute value of the difference between the maximum pressure or the minimum pressure and the median pressure, with units of MPa, usually ranging from 0 to 100 MPa. The pressure frequency refers to the number of times the oscillatory pressure undergoes periodic changes within a unit of time, with units of Hz, usually ranging from 0 to 100 Hz.

2.1.4 Advantages of hot oscillatory pressing

(1) Promoting densification

The application of external pressure represents an effective method for enhancing the sintering properties of materials, particularly those that are challenging to sinter or necessitate high-temperature sintering. In comparison to hot pressing, hot oscillatory pressing has the potential to enhance the density of the sintered material, as illustrated in Table 4. For example, Fan *et al.*

[43] prepared highly dense B_4C ceramics via hot oscillatory pressing. In comparison to hot pressing, hot oscillatory pressing has been demonstrated to not only enhance the densification rate of the sample but also to elevate the final density of the sample (Fig. 14). Moreover, Liu *et al.* [37] investigated the impact of oscillatory pressure applied at distinct sintering stages on alumina density. Their findings indicated that oscillatory pressure applied at any stage of sintering can enhance density, with the application at the final stage of sintering proving particularly beneficial. Oscillatory pressure has been demonstrated to be an effective technique for promoting densification, with the potential to achieve higher relative densities at lower sintering temperatures. Han *et al.* [67] employed hot oscillatory pressing to produce zirconia ceramics with a close theoretical density and high strength at a temperature of 1300 °C, which is 150 °C lower than the temperature typically required for hot pressing.

(2) Optimization of microstructure

In the sintering of ceramic materials, the control of grain growth is of great importance, as the grain size, distribution, and uniformity have a significant effect on the properties of ceramics, including their mechanical properties. It is generally believed that the inhibition of grain growth by constant pressure is mainly due

to the reduction in sintering temperature, and the sintering trajectory is consistent with that of pressureless sintering [69]. Compared with constant pressure, oscillatory pressure not only promotes densification but also inhibits grain growth, thus increasing the ratio of density to grain size. For example, the grain size of high-purity Al_2O_3 ceramics [70] prepared via hot oscillatory pressing is 3.99 μm , which is 1 μm smaller than that of the sample sintered via hot pressing. The results of alumina toughened zirconia (ATZ) ceramics [36] show that for the ZrO_2 phase (Fig. 15), the grain growth constants of the samples sintered via hot pressing and hot oscillatory pressing are 2.1×10^{-18} and $8.5 \times 10^{-19} m^2/s$, respectively. In other words, the grain growth constant of the sample sintered via hot oscillatory pressing is twice as low as that of hot pressing, so the oscillatory pressure effectively inhibits grain growth. In some special cases, the oscillatory pressure did not inhibit the grain growth but promoted it. For example, in the sintering of Si_3N_4 [40,71,72], it was found that the sample prepared via hot oscillatory pressing had a faster growth rate in the width direction [2100] than the sample prepared via hot pressing. The researchers believe that this is because the grain plastic deformation and grain boundary sliding induced by oscillatory pressure promote the diffusion of Si and N (D_I),

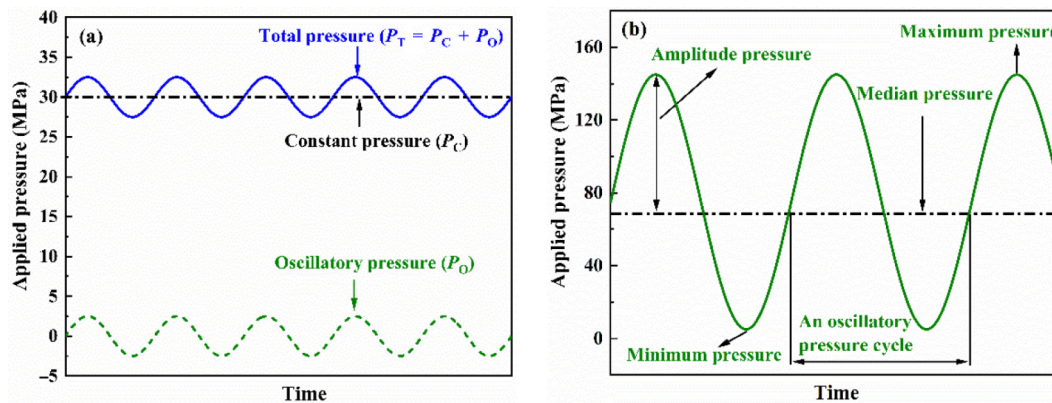


Fig. 13 Typical schematic diagram of applied oscillatory pressure: (a) Xie's group, (b) An's group. Reproduced with permission from Ref. [66] for (a), © Elsevier Ltd. and Techna Group S.r.l. 2017.

Table 4 Relative density of ceramic materials prepared via different sintering techniques

Sintering technology	Relative density (%)				
	ZrO ₂ [35]	Al ₂ O ₃ [68]	Al ₂ O ₃ -ZrO ₂ [38]	B ₄ C [43]	SiC [37]
Pressureless sintering	98.1	97.6	—	—	—
Hot pressing	99.2	99.0	95.9	98.8	97.8
Hot oscillatory pressing	99.7	99.6	96.9	99.6	98.5

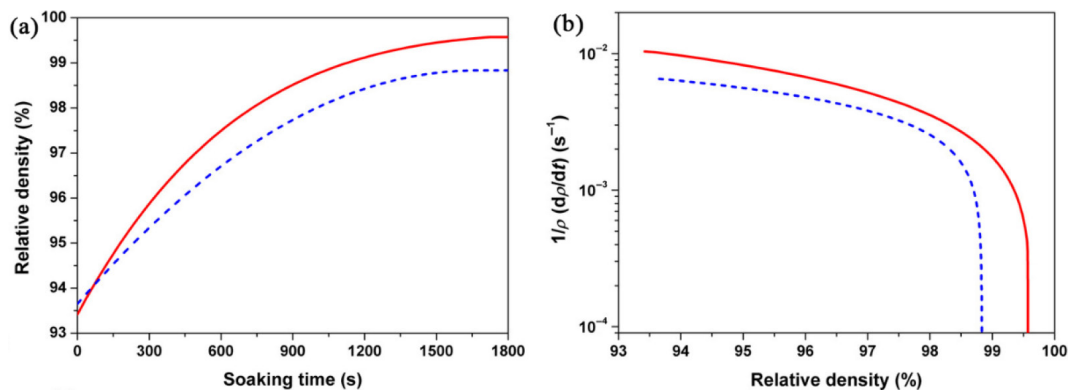


Fig. 14 Instantaneous (a) relative density and (b) densification rate of B_4C samples sintered via HOP (solid line) and HP (dashed line) at 1800 °C. Reproduced with permission from Ref. [43], © The American Ceramic Society 2022.

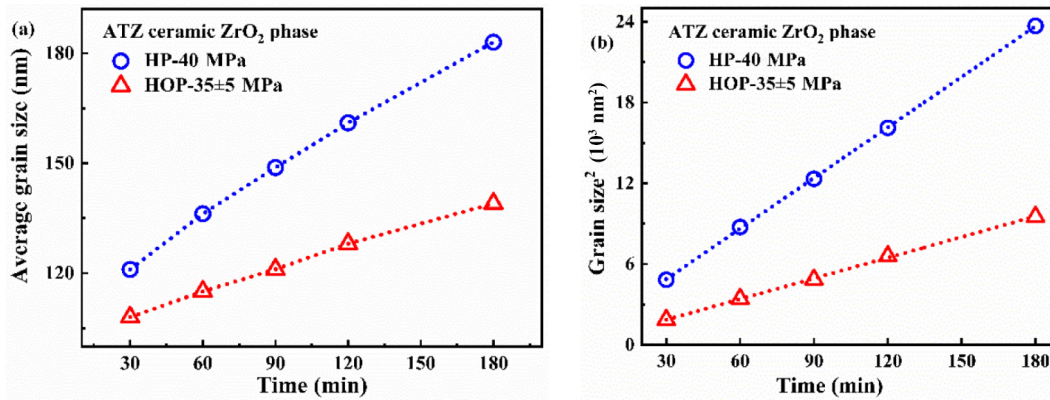


Fig. 15 (a) Average grain size and (b) grain growth kinetics of ZrO₂ for the samples sintered at 1400 °C via HP (circles) and HOP (triangles) as a function of sintering time. Reproduced with permission from Ref. [36], © Elsevier B.V. 2020.

resulting in a faster growth rate of Si₃N₄ grains in the width direction.

The oscillatory pressure also has the advantages of optimizing the microstructure (more uniform grain size distribution) and strengthening the grain boundaries. For example, the grain size distributions of WC–Ni–Fe cemented carbide [73] sintered via hot oscillatory pressing and hot pressing are 0.72–0.83 and 0.76–1.08 μm, respectively, and the results show that the sample sintered via hot oscillatory pressing has narrower grain size distributions and a more uniform microstructure. Compared with hot pressing, Zhu *et al.* [1] found that ZrO₂ ceramics sintered via hot oscillatory pressing contained a large number of coherent grain boundaries (Fig. 16). Since the binding energy of the coherent grain boundaries is significantly higher than that of the noncoherent grain boundaries, the grain boundary bonding strength is significantly improved. The improvement in grain boundary bonding strength contributes significantly to the mechanical properties of materials [65,74,75].

(3) Introducing more crystal defects in ceramics

Studies [76–79] have shown that presetting high-density dislocations in ceramics can simultaneously improve their strength, hardness, and toughness (this method has been widely studied and applied in metal materials [80]), which has become a research hotspot in the field of ceramics in recent years. For example, when the dislocation density in single-crystal SrTiO₃ is 10¹³/m², the hardness and fracture toughness can be increased by 20% at the same time [81], and when the dislocation density is increased to 10¹⁴/m², crack initiation on the (100) plane can be completely suppressed [76]. Due to the high bonding strength of ionic/covalent bonds that constitute ceramics, the Peierls stress to be overcome by dislocation nucleation and movement is extremely high, and there are fewer independent slip systems (< 5) in multiple crystals that are difficult to coordinate with each other; it is difficult for dislocations to nucleate and move in the process of ceramic preparation and deformation. Ceramics usually prepared by conventional sintering methods (such as pressureless sintering and hot pressing) tend to contain low-density (~ 10⁹/m²) dislocations [82], so it is very difficult to prefabricate such high-density (10¹⁴/m²) dislocations in ceramics, especially block ceramics.

It is noteworthy that it is easier to introduce crystal defects such as dislocations, twins, and stacking faults in materials under the coupling of oscillatory pressure and high temperature, and the effect is more significant in ceramic materials, which provides a new idea for the prefabrication of crystal defects in ceramics. Han *et al.* [60] compared the electron backscatter diffraction (EBSD) results of carbon nanotube-reinforced copper matrix nanocomposites prepared via hot pressing and hot oscillatory

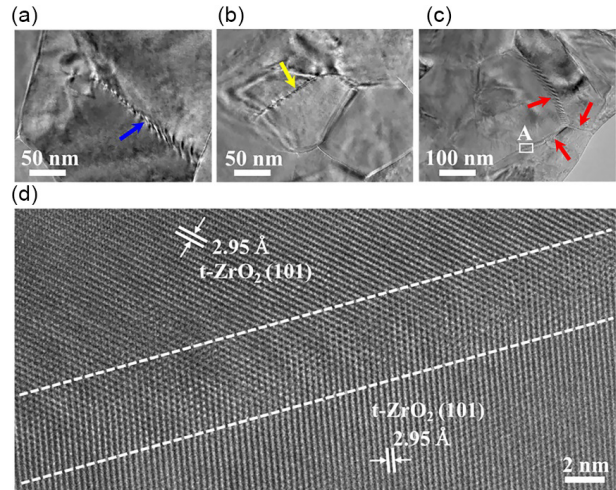


Fig. 16 TEM images of sample ZO-1450: (a) intragranular dislocation activity, (b) subgrain boundary, (c) moiré patterns, (d) high-resolution TEM image of region A in (c). Reproduced with permission from Ref. [1], © The American Ceramic Society 2021.

pressing at 700 °C and showed that the samples prepared via hot oscillatory pressing had a higher dislocation density due to repeated plastic deformation. Zhu *et al.* [1] observed a large number of dislocations and subgrain boundaries in ZrO₂ ceramics prepared via hot oscillatory sintering, while these crystal defects were rarely observed in samples sintered via hot pressing. Liu *et al.* [44] found that the dislocation density in SiC ceramics sintered via hot oscillatory pressing was approximately twice that of samples sintered via hot pressing (Fig. 17). In addition, SiC ceramics sintered via hot oscillatory pressing have more nanospaced stacking faults and high-order nanotwins. Zhao *et al.* [83] analyzed the microstructure of B₄C ceramics at different sintering temperatures and found that all the samples sintered via hot oscillatory pressing contained a large number of crystal defects, including dislocations, twins, and stacking faults. Moreover, the densification mechanism of different sintering stages is analyzed based on the Bernard–Granger model [84], and it is believed that these defects play an important role in the densification process of B₄C ceramics. Cheng *et al.* [85] observed a large number of dislocations in the ceramic matrix of WC–ZrO₂–Al₂O₃, which existed between WC grains. The dislocation may be related to the plastic deformation of ceramics promoted by oscillatory pressure at high temperatures.

(4) Improving performance

Based on the advantages of oscillatory pressure to promote densification and optimize microstructure, the materials prepared

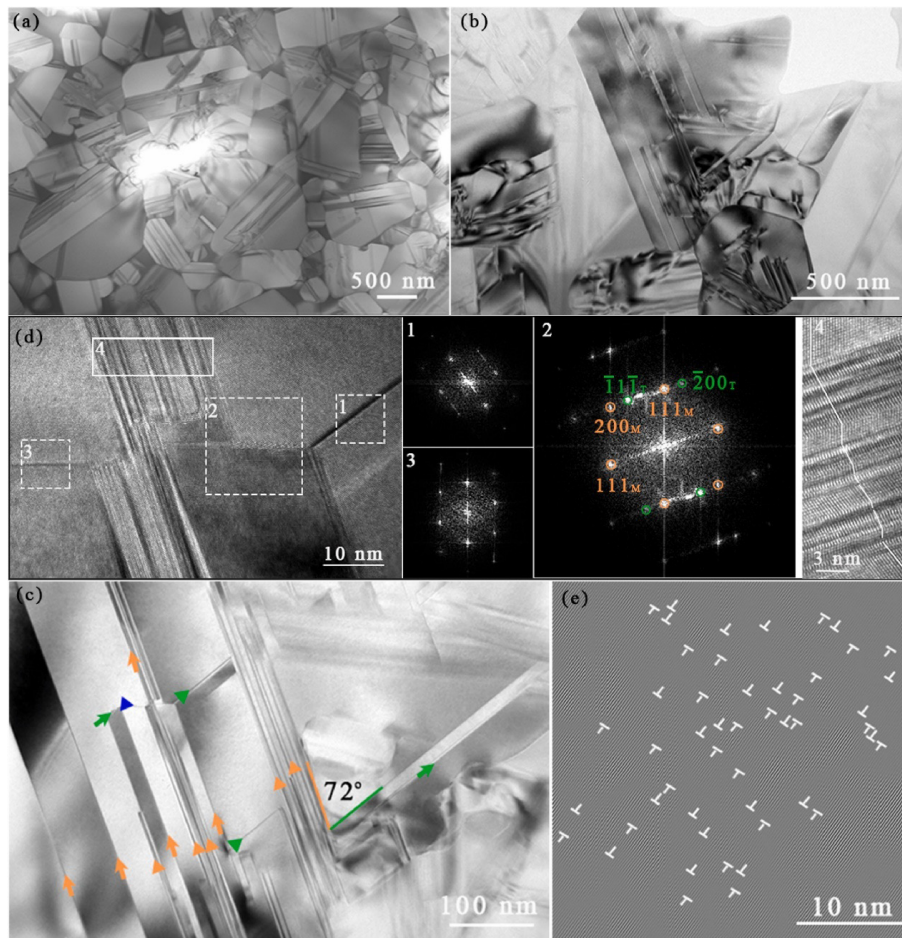


Fig. 17 TEM images of sample HOP-1850. (a) Bright-field TEM image at a relatively low magnification; (b, c) bright-field TEM images of a typical grain containing nano-spaced stacking faults (SFs) and nanotwins; (d) high-resolution TEM image of typical nSFs and high-order nanotwins and diffraction patterns corresponding to areas 1, 2, and 3, and the detailed atomic stacking sequences corresponding to area 4 in (d); (e) inverse Fourier transform image of area 2 depicted in (d). Reproduced with permission from Ref. [44], © Elsevier Ltd. and Techna Group S.r.l. 2023.

via hot oscillatory pressing generally have excellent properties, even breaking the limits of existing material properties. ZrO_2 ceramics [1] prepared via hot oscillatory pressing reached a record-breaking high flexural strength of 1.8 GPa (Fig. 18). The high flexural strength may be due to the effective elimination of micropores, the introduction of dislocations, subgrain boundaries, and grain boundary strengthening caused by coherent grain boundaries. B_4C ceramics [43] prepared via hot oscillatory pressing had a hardness of 39.5 ± 1.0 GPa, which was 3 GPa higher than that of the samples sintered via hot pressing and was also relatively high in Ref. [83,86,87]. The WC-GNP composites [50] prepared via hot oscillatory pressing showed good wear resistance with a low friction wear rate of $3.17 \times 10^{-7} \text{ mm}^3/(\text{N} \cdot \text{m})$. The electrical properties and thermoelectric (TE) properties of $\text{Ca}_3\text{Co}_4\text{O}_9$ prepared via hot oscillatory pressing are superior to those of samples sintered via conventional hot pressing [88], which may be attributed to the better alignment of plate-like grains in the samples sintered via hot oscillatory pressing. $\text{Na}_{0.5}\text{K}_{0.5}\text{NbO}_3$ ceramics [89] prepared via hot oscillatory pressing exhibit superior performance with a piezoelectric parameter (d_{33}) of 142 pC/N, an electromechanical coupling factor (k_p) of 0.41, and a relative permittivity ($\epsilon_{33}^T/\epsilon_0$) of 472–620. The W–Ni–Fe refractory alloy [90] prepared via hot oscillatory pressing has excellent corrosion resistance, and its corrosion current density is only $2.1 \times 10^{-6} \text{ A/cm}^2$. In addition, the oscillatory pressure significantly weakens the influence of powder on the mechanical

properties of materials and is expected to eliminate the dependence of high-performance materials on powder. For example, Zhang *et al.* [91] prepared high-strength FGH4096 superalloy materials via hot oscillatory pressing of powder with different particle sizes (51 and 120 μm). The results showed that the tensile strength and elongation of coarse powder samples prepared via hot oscillatory pressing were superior to those of high-cost fine powder samples prepared via hot pressing.

2.1.5 Possible mechanism

(1) Mechanism of oscillatory pressure promoting densification in solid-phase sintering

The continuous oscillatory pressure applied during the sintering process shortens the diffusion distance by promoting particle rearrangement and eliminating particle agglomeration, thus promoting sintering. For example, Feng *et al.* [39] assisted cold pressing with constant pressure and oscillatory pressure and obtained densities of AlN green bodies of 1.352 and 1.405 g/cm^3 , respectively. The results show that the oscillatory pressure can help to overcome the friction between particles, promote particle rearrangement, and increase the sample density.

The pressure applied in hot pressing can induce additional densification mechanisms such as diffusion enhancement under stress, grain boundary sliding, power law creep, and plastic deformation [17]. The oscillatory pressure enhances these mechanisms and increases the densification rate of the ceramic

material, thereby increasing the final density. Studies on the mechanism of zirconia [92] and alumina whisker-enhanced zirconia [38] show that although oscillatory pressure provides an additional driving force to improve the densification rate and density, it does not change the sintering mechanism (Figs. 19(a) and 19(b)). Zhao *et al.* [83] studied the densification mechanism of B_4C in hot oscillatory pressing, and the results showed that the densification mechanism of the samples varied with density (Fig. 19(c)). When the relative density of the sample is 62%–75%, the dominant densification mechanism is lattice diffusion/grain boundary diffusion. When the relative density of the sample is 75%–90%, the dominant mechanism of densification is grain boundary sliding by dislocation motion. When the relative density of the sample is 90%–95%, the dominant mechanism of densification is dislocation gliding and stacking faults. Of course, the dominant densification mechanism depends not only on the sintering parameters (such as the applied pressure) but also, to a large extent, on the intrinsic properties of the material (such as the plastic yield stress [93]) or other extrinsic properties (such as powder size [94]).

(2) Mechanism of oscillatory pressure promoting densification in liquid-phase sintering

In liquid-phase sintering, the oscillatory pressure accelerates the mass transfer in the liquid phase, thereby promoting phase transformation or solid solution. For example, Qin *et al.* [41] compared the X-ray diffraction (XRD) patterns of Si_3N_4 samples prepared via gas pressure sintering and hot oscillatory pressing and found that the transformation rate of $\alpha-Si_3N_4$ to $\beta-Si_3N_4$ in samples prepared via hot oscillatory pressing was faster than that of samples prepared via gas pressure sintering (Fig. 20). Liu *et al.* [44] used hot oscillatory pressing to prepare SiC, and the XRD results showed that the $\alpha-SiC$ content of the sample sintered via hot oscillatory pressing was higher than that of the sample sintered via hot pressing at 1850 °C, indicating that the oscillatory pressure facilitated the transformation of $\beta-SiC$ to $\alpha-SiC$. The results of the W–Ti alloy [53] and W–Ni–Fe alloy [90] show that the diffraction peak of the sample is shifted, which may be caused by the solid solution of solute atoms promoted via the oscillatory pressure.

(3) Mechanism of oscillatory pressure inhibiting grain growth

Although many studies clearly show that oscillatory pressure can inhibit grain growth, there are few studies on grain growth mechanisms. At present, the possible mechanism of oscillatory

pressure inhibiting grain growth is as follows: (1) Oscillatory pressure promotes plastic deformation, resulting in grain boundary flattening and increasing the curvature radius, thus reducing the grain growth rate. Fan *et al.* [36] calculated the grain growth constant of ATZ ceramics, and the results showed that the grain growth rate of the samples sintered via hot oscillatory pressing was several times lower than that of the samples sintered via hot pressing, which was attributed to the change in grain boundary curvature radius. Fan *et al.* [65] found that the proportion of straight grain boundaries was different in samples prepared by different oscillatory pressures, and suitable oscillatory pressures could significantly increase the proportion of flat grain

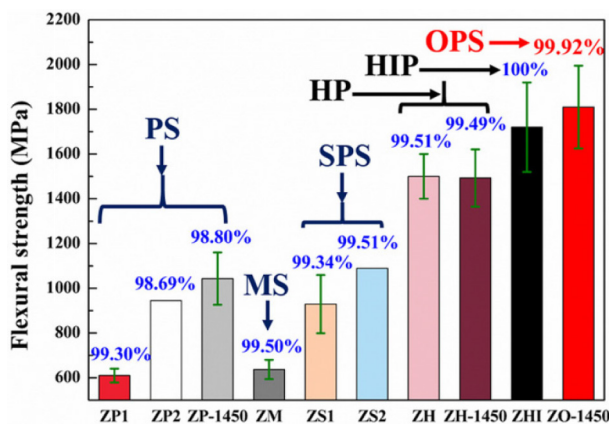


Fig. 18 A comparison of flexural strengths of ZrO_2 samples sintered via different technologies. PS—pressureless sintering, MS—microwave sintering, SPS—spark plasma sintering, HP—hot pressing, HIP—hot isostatic pressing, OPS—hot oscillatory pressing. Reproduced with permission from Ref. [1], © The American Ceramic Society 2021.

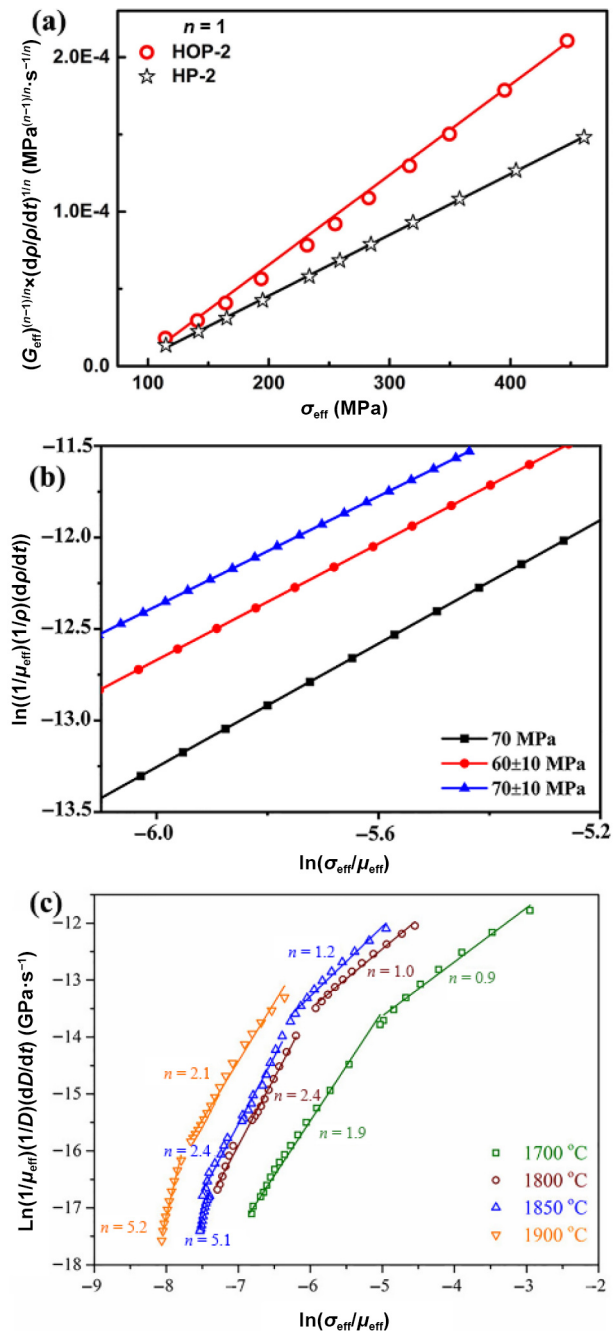


Fig. 19 Modified densification rates as a function of effective pressure: (a) ZrO_2 , (b) $Al_2O_{3W}-ZrO_2$, (c) B_4C . Reproduced with permission from Ref. [92] for (a), © The Authors 2022; from Ref. [38] for (b), © The Author(s) 2022; from Ref. [83] for (c), © Wiley-VCH GmbH 2024.

boundaries (Fig. 21). The higher the proportion of straight grain boundaries, the finer the grain. (2) Oscillatory pressure strengthens the pinning effect of solute atoms at grain boundaries. Gao *et al.* [51] found that the growth activation energy of W-Fe-Ni grains prepared via hot oscillatory pressing sintering was six times smaller than that of hot pressing, and it was believed that the oscillatory pressure promoted solutes to diffuse around the grain boundary and then pinned the grain boundaries, thus reducing the activation energy of grain growth.

2.1.6 Application of oscillatory pressure in ceramic processing

(1) Dynamic hot forging

Hot forging is a commonly used plastic process for forming and strengthening metal materials [95,96]. The main reason for the improvement of material properties by hot forging is the elimination of cracks, pores, and work hardening caused by high-density dislocation in the process of hot forging. However, hot forging requires a high plastic deformation capacity of materials. For ceramics, excessive shear strain will damage the specimen [97], which limits its wide application in ceramics.

Inspired by the effect of oscillatory pressure on accelerating mass transfer and improving plastic deformation ability in the

sintering process, Liu *et al.* [74] replaced the static constant pressure in the hot forging process with dynamic oscillatory pressure and named this type of oscillatory pressure-assisted hot forging dynamic hot forging. A schematic diagram of dynamic hot forging is shown in Fig. 22. The dense sample is placed between the upper and lower graphite punches, and then a small pressure is applied to ensure good contact between the graphite punch and the sample; then the sample is heated to a suitable temperature. Finally, an oscillatory pressure of a certain amplitude and frequency is applied for dynamic hot forging.

Dynamic hot forging not only significantly increases the strain of ceramics but also significantly increases the strain rate of ceramics, thus improving the plastic deformation capacity of ceramics. Taking zirconia as the model material, Liu *et al.* [74] and Fan *et al.* [98] preliminarily conducted hot forging experiments on dense ZrO_2 ceramics. Regardless of conventional hot forging or dynamic hot forging, ZrO_2 ceramics have obvious plastic deformation at 1300 °C (Fig. 23). As seen from Fig. 23(a), compared with conventional hot forging, the sample processed by dynamic hot forging has a larger axial strain, which increases from 15.9% of conventional hot forging to 29.0% of dynamic hot forging, and the strain increases by 13.1%. In addition, compared

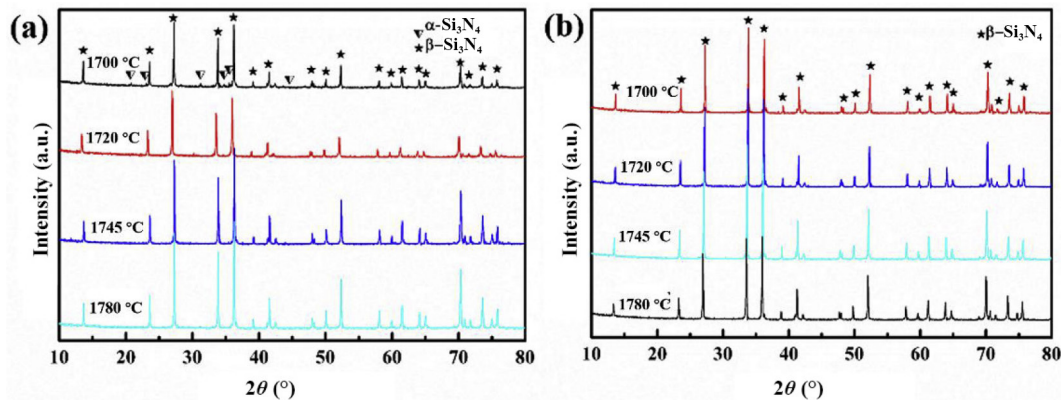


Fig. 20 XRD patterns of Si_3N_4 samples sintered via (a) GPS and (b) HOP at 1700, 1720, 1745, and 1780 °C. Reproduced with permission from Ref. [41], © Elsevier Ltd and Techna Group S.r.l. 2020.

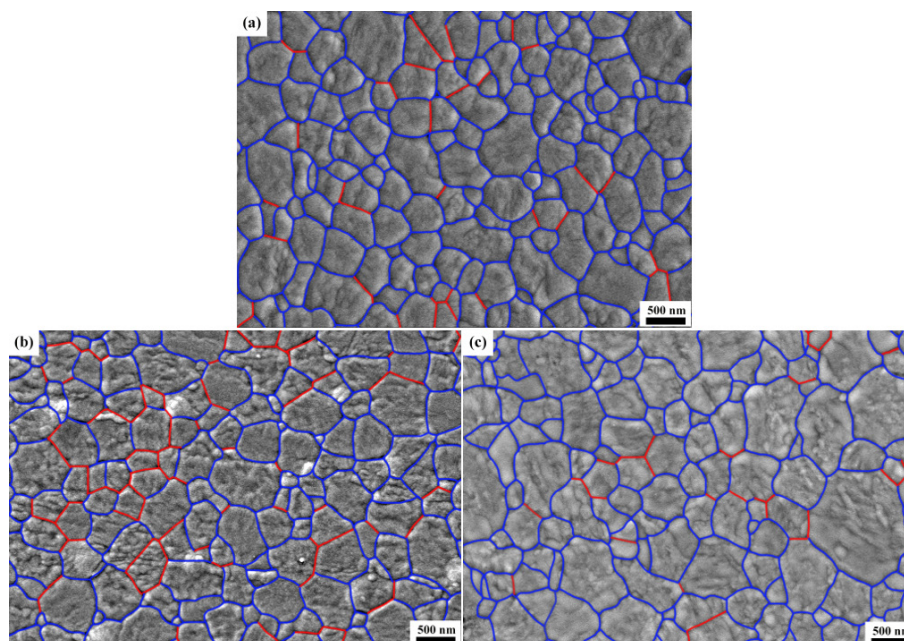


Fig. 21 Typical scanning electron microscopy (SEM) images of samples: (a) DJ40±35, (b) DJ75±35, (c) DJ75±70. The red lines mark flat grain boundaries, while the blue lines indicate curved grain boundaries. Reproduced with permission from Ref. [65], © Elsevier Ltd. 2024.

with conventional hot forging, the sample processed by dynamic hot forging has a larger axial strain rate, which is increased from $6.9 \times 10^{-3} \text{ s}^{-1}$ for conventional hot forging to $1.5 \times 10^{-2} \text{ s}^{-1}$ for dynamic hot forging, and the corresponding strain rate is doubled, effectively shortening the deformation time. Correspondingly, the radial strain and strain rate of the samples prepared by dynamic hot forging also showed a similar trend, as shown in Fig. 23(b). In addition, the results of He *et al.* [99] also show that the creep rate of alumina whisker-toughened zirconia ceramics is faster under oscillatory pressure.

Dynamic hot forging can effectively eliminate pores and microcracks in the material and strengthen the grain boundaries. Liu *et al.* [74] characterized the microstructure of zirconia ceramics before and after hot forging. As shown in Fig. 24(a), purchased commercial zirconia ceramics contain microcracks ranging in size from a few microns to tens of microns. After hot forging, almost no microcracks were observed in the sample, as shown in Fig. 24(b). In addition, the fracture morphology of the samples before and after hot forging was characterized. The original sample OS is mainly intergranular fracture, and only ~28% is transgranular fracture (Fig. 24(c)), while the proportion of transgranular fracture for sample DHF1400 is up to ~70% (Fig. 24(d)). The increase in the proportion of transgranular fracture indicates grain boundary strengthening [1,35], which may result from the repeated sliding of grain boundaries under oscillatory pressure [38].

Dynamic hot forging can also introduce a large number of

crystal defects in the material. Liu *et al.* [74] conducted TEM characterization of samples prepared by conventional hot forging and dynamic hot forging of zirconia ceramics (Fig. 25). The results show that both hot-forged samples contain a significant number of defects, such as dislocations and stacking faults (Figs. 25(a) and 25(b), and 25(g) and 25(h)). Although the two samples exhibit the same macroscopic strain, the sample prepared by dynamic hot forging has a higher defect density, which may be due to a greater degree of intracrystalline plastic deformation under oscillatory pressure. Fu *et al.* [100] also found a large number of crystal defects in $\text{ZrO}_2\text{-SiO}_2$ dual-phase nanocrystalline ceramics after dynamic hot forging treatment, including edge dislocations, subgrain boundaries, nanotwins, and stacking faults. The results of He *et al.* [99] show that the dislocation density in alumina whisker-toughened zirconia ceramics is higher under oscillatory pressure, indicating that the oscillatory pressure improves the motion ability of the dislocation.

After dynamic hot forging, defects such as pores and microcracks can be effectively eliminated, and the grain boundaries can be effectively strengthened. As a result, the properties of the material are significantly improved. Liu *et al.* [74] characterized the flexural strength of zirconia ceramics before and after hot forging, as shown in Fig. 26. The flexural strength of zirconia ceramic samples after hot forging is significantly higher than that of unforged samples. The flexural strength of ZrO_2 ceramics increases with increasing hot forging temperature, and the flexural strength of the sample ($1861 \pm 111 \text{ MPa}$) obtained by

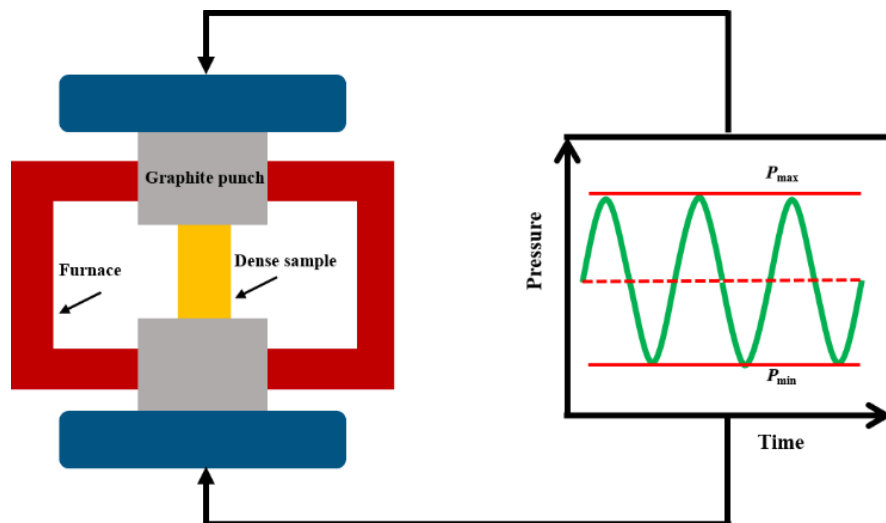


Fig. 22 Schematic diagram of dynamic hot forging.

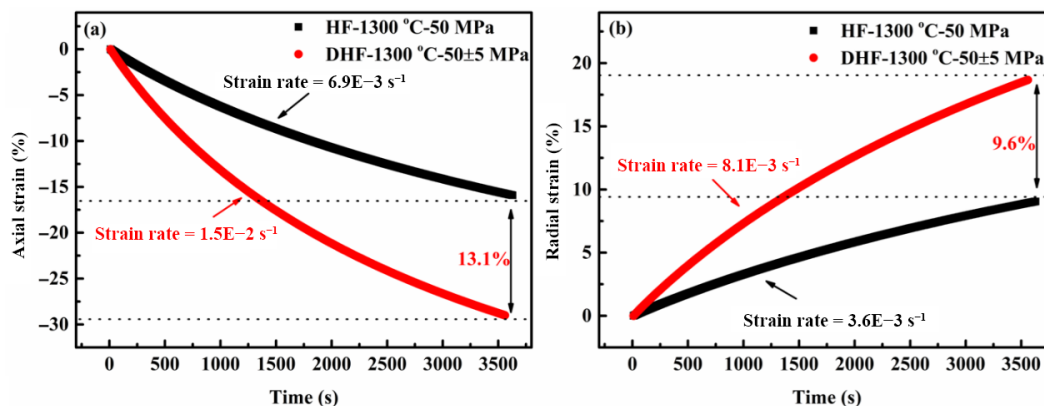


Fig. 23 Strain curves of hot-forged ZrO_2 ceramics with time: (a) axial strain; (b) radial strain. HF—conventional hot forging; DHF—dynamic hot forging.

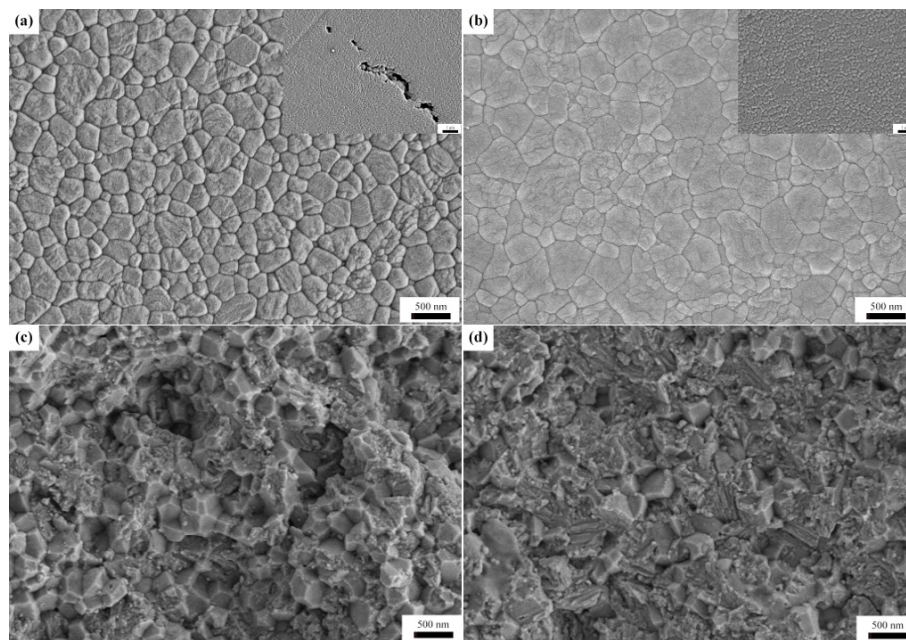


Fig. 24 Typical SEM images of (a) OS and (b) DHF1400, and fracture morphology of (c) OS and (d) DHF1400. Reproduced with permission from Ref. [74], © Elsevier Ltd. 2022.

dynamic hot forging at 1400 °C is slightly higher than the flexural strength of ZrO_2 (1810 ± 155 MPa) reported [1]. As the hot forging temperature continues to increase, the flexural strength decreases, which may be caused by grain growth or the introduction of vacancies. The flexural strength of the sample processed via dynamic hot forging is approximately 125 MPa higher than that of the sample processed via conventional hot forging, which indicates that the oscillatory pressure may further remove the grain boundary closed pores and eliminate cracks and other defects, which provides a possibility for the secondary processing of ceramics. Dynamic hot forging is also used to process B_4C ceramics [86], Si_3N_4 ceramics [101], Al_2O_3 -Ti(C,N)-CBN composites [102], WC-Co cemented carbides [103], and FGH4096 superalloys [104], all of which effectively improve material properties.

(2) Dynamic plastic joining

Due to their inherent low plasticity, poor toughness, and limited impact resistance, ceramic materials are difficult to process into large or complex-shaped components using conventional methods. Therefore, developing reliable ceramic joining technologies is of great significance for achieving integrated manufacturing of complex structures. The most widely used ceramic joining techniques include brazing, diffusion bonding, and superplastic bonding [105]. Among these, brazing offers simplicity and high efficiency, but the service temperature of the resulting joints is relatively low, limiting its application in high-temperature environments [106]. Although diffusion bonding and superplastic bonding can produce high-strength, heat-resistant joints with excellent performance, the prolonged processing time can lead to microstructural evolution and performance degradation in the base material [107,108]. With the expanding use of ceramic materials in extreme environments such as aerospace and energy equipment, there are increasingly stringent requirements for the high-temperature stability and comprehensive mechanical properties of joined joints. As a result, further development of efficient and highly reliable ceramic joining technology has become a critical research direction for enhancing the functional integration and expanding the application scope of ceramic structures.

Inspired by the ability of oscillatory pressure to enhance mass transfer and improve plastic deformation during dynamic hot forging, Fan *et al.* [65] innovatively replaced the conventional static constant pressure in plastic bonding with dynamic oscillatory pressure, thereby developing a novel dynamic plastic bonding technology. A schematic diagram of the experimental procedure is shown in Fig. 27: First, the samples to be joined are placed in a graphite mold, and a small preload is applied to ensure good contact between the graphite punch and the samples; then, the assembly is heated to a predetermined temperature; finally, oscillatory pressure with a specific amplitude and frequency is applied to achieve dynamic joining. The experimental results show that significant plastic deformation occurs in the zirconia ceramic samples during the dynamic plastic joining process, with a shear strain as high as 0.82. No obvious pores are observed in the bonded interface layer, and the sample remains highly dense. The flexural strength of the joined sample reaches 1552 ± 225 MPa, which is 110% higher than that of the parent material. This improvement in performance can be attributed to three mechanisms: first, the elimination of large pores under high shear strain; second, the enhancement of grain boundary strengthening effects; third, the introduction of high-density dislocations and their associated strengthening contributions.

2.1.7 Challenges and opportunities for hot oscillatory pressing

(1) Challenges

First, hot oscillatory pressing equipment is expensive, and the initial investment is large, which is not a small burden for many enterprises and research institutions. Therefore, to optimize the equipment structure and reduce the cost, it is possible to promote the technology on a large scale.

Second, the technology has high requirements for temperature and pressure control, and the operation technology is complex, requiring professional technicians to operate and maintain. Therefore, the integration of equipment and intelligent control should be improved, and the dependence on professional and technical personnel should be effectively reduced.

Third, in the preparation of large samples, the large oscillatory pressure required is difficult to match with the existing pressure

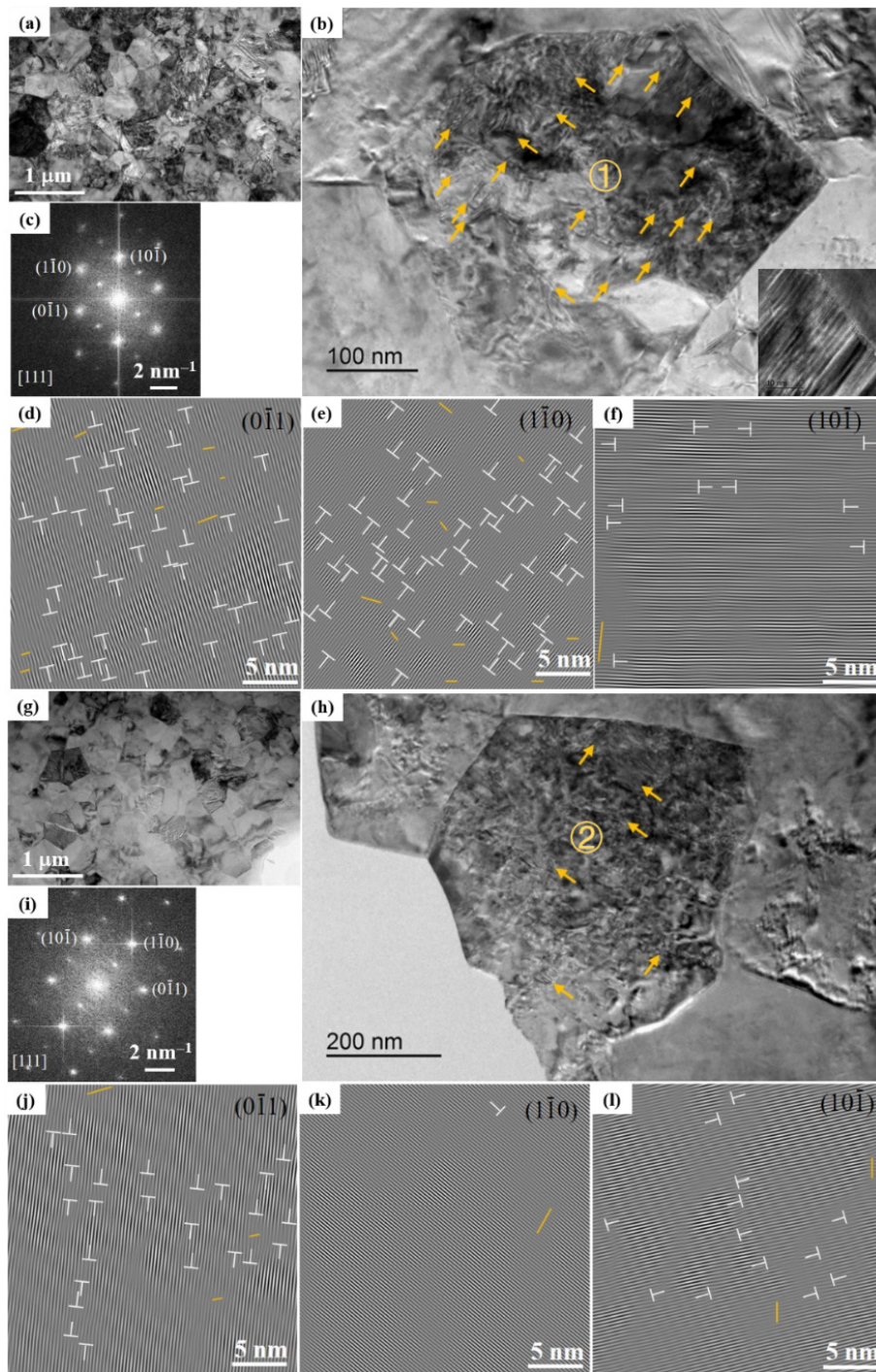


Fig. 25 Typical TEM images of (a–f) DHF1400 and (g–l) CHF1400: (a, b, g, h) high-resolution TEM (HRTEM) images; (c, i) single-crystal electron diffraction patterns of grains ① and ② in (b) and (h), respectively; (d–f) and (j–l) inverse fast Fourier transform diagrams of crystal planes (011), (110), and (101) of grains ① and ② in (b, h), respectively. Reproduced with permission from Ref. [74], © Elsevier Ltd. 2022.

system, which is also a great challenge for the manufacturing industry. Therefore, the development of hot oscillatory pressing also depends on the continuous progress of other technologies.

Finally, the technology places higher demands on molds such as graphite, and under the multiple effects of creep and fatigue, the mold is more easily damaged, and the service life is significantly shortened. Due to the limitations of the mold, the effective pressure applied by hot oscillatory pressing is generally no more than 80 MPa. Therefore, the development of higher-performance molds, such as carbon–carbon composite molds, will provide the opportunity to produce extremely high-performance materials.

(2) Opportunities

Hot oscillatory pressing has attracted much attention for its wide range of applications, ranging from ceramics to cemented carbide and metal powder metallurgy. The technology significantly improves the density, hardness, wear resistance, and strength of materials through oscillatory pressure, breaking the limits of existing material properties, and is expected to be applied in electronics, optics, life sciences, cutting tools, aerospace, and transportation manufacturing. At the same time, with the progress of materials science, hot oscillatory pressing has also shown great potential in the research and development of new materials, such

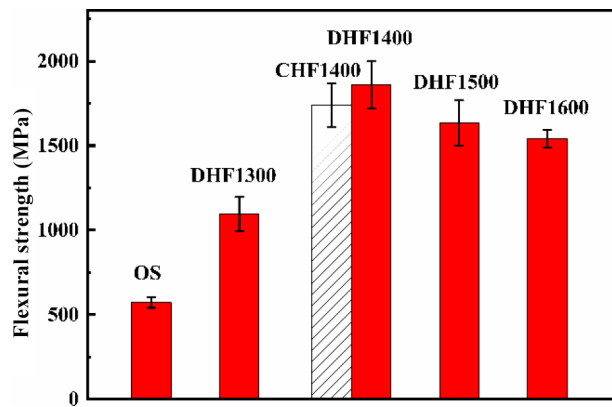


Fig. 26 Flexural strengths of original sample and samples forged at different temperatures. Reproduced with permission from Ref. [74], © Elsevier Ltd. 2022.

as nanomaterials and semiconductor materials, indicating its future expansion and application in more fields.

2.2 Cold sintering

2.2.1 Development of cold sintering

During the Late Paleolithic period, approximately 25,000 years ago, humans had already mastered the skill of creating ceramics by heating clay materials. This remarkable feat greatly satisfied the needs of daily life during that time [109–111]. Ceramics have significantly expanded their applications in recent times, seamlessly integrating into decorative and structural aspects of the construction industry. Furthermore, they play a critical role in cutting-edge technological fields as functional materials, such as new energy development, communication electronics, semiconductor technology, the military industry, and aerospace [112–114]. In many of these applications, the sintering process is required to densify the ceramic material for functionality. However, conventional sintering of ceramics requires prolonged high-temperature processing, leading to high energy consumption and substantial carbon emissions. Moreover, high temperatures tend to cause abnormal grain growth and volatilization of some elements (Na, K, Bi, etc.), negatively impacting the performance of ceramics [115–117]. To reduce sintering temperatures, shorten sintering time, and decrease energy consumption, researchers

have developed various sintering technologies over the past decades [118,119], primarily including liquid-phase sintering [120,121], high-pressure sintering [122], microwave sintering [123,124], two-step sintering [125,126], spark plasma sintering [127,128], flash sintering [13,129,130], and multi-technique sintering [131–134].

In fact, nature exhibits processes similar to sintering, such as the formation of fossils and diamonds, the growth of teeth and pearls, and the agglomeration of salt/sugar particles [135]. These phenomena are characterized by densification occurring under the assistance of liquid-phase environments. A number of scientists have directed considerable efforts toward the preparation of dense and sturdy ceramics at low temperatures with aqueous solutions. In 1986, Yamasaki *et al.* [136] designed two types of autoclaves and proposed hydrothermal hot-pressing (HHP). However, the densities of ceramics are relatively low using the HHP process.

Inspired by natural sintering-like phenomena and hydrothermal hot-pressing [137], Randall, Guo, and colleagues at Pennsylvania State University [32] proposed the cold sintering process (CSP) in 2016. They mixed ceramic powders with a small amount of aqueous solutions (named transient liquid phase or mediated liquid phase), such as deionized water, acid, and aqueous alkali, and put the wetted powders in a steel mold with axial pressure and heat. Densification occurs under mild sintering conditions with a temperature much lower than that of conventional thermal sintering. They have demonstrated a large number of ceramics to be well densified using this method, including Li_2MoO_4 , $\text{K}_2\text{Mo}_2\text{O}_7$, $\text{Na}_2\text{Mo}_2\text{O}_7$, NaCl , V_2O_5 , and so on, which is termed “cold sintering process” to highlight the extremely low sintering temperature [32,135,138–142].

In this review, we first illustrate the apparatus and densification mechanism of the cold sintering process and then summarize the recent developments of cold-sintered ceramics and composites in various applications. Finally, the advantages and disadvantages of the cold sintering process as well as future challenges are discussed. This review not only provides comprehensive scientific explanations for CSP phenomena but also offers practical guidelines for future researchers.

2.2.2 Setup and mechanism of cold sintering

(1) Setup of cold sintering

The cold sintering process can be applied using a simple and

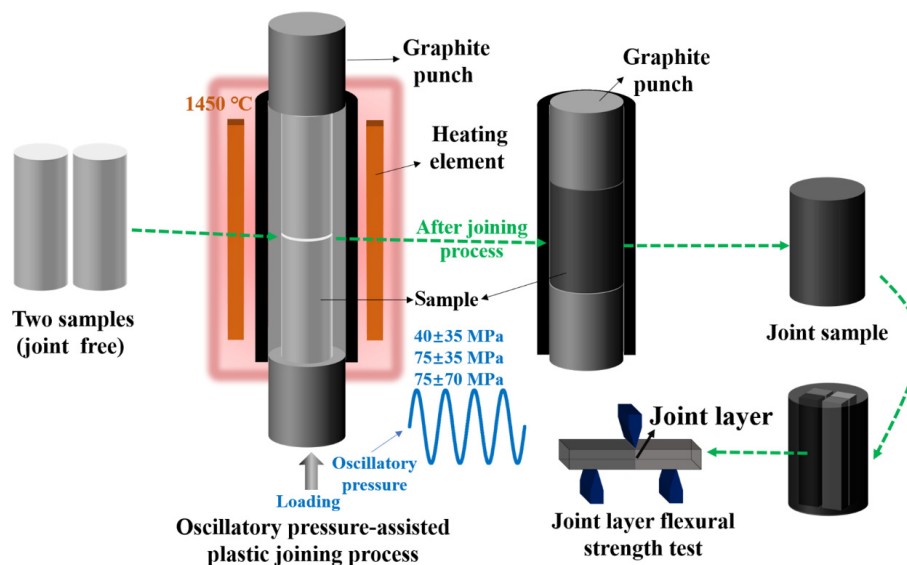


Fig. 27 Diagram of the experimental procedures. Reproduced with permission from Ref. [65], © Elsevier Ltd. 2022.

inexpensive apparatus. The primary steps involved in the cold sintering process are as follows: First, an adequate amount of solvent is introduced into the ceramic powders to ensure uniform wetting of the particle surfaces. Subsequently, the wetted powders are transferred into a mold. Afterwards, uniaxial pressure is applied to the material within the mold. Upon reaching the maximum load, the heating temperature is typically maintained below 300 °C, which can be accomplished using upper and lower heating plates or a heating jacket. A schematic diagram of the experimental setup for the cold sintering process, as well as images of the heating plates and heating jacket, are shown in Fig. 28.

(2) Densification mechanism of cold sintering

In the study of cold sintering mechanisms, two primary theories have been proposed, including the dissolution-

precipitation mechanism and plastic deformation. Pressure solution creep originating from the dissolution–precipitation process is also proposed because pressure together with the transient liquid phase is usually utilized during the cold sintering process.

Figure 29(a) shows the three stages of the dissolution–precipitation process [32,135]. The first stage involves particle rearrangement. During this stage, the introduction of a transient liquid phase enhances the lubricity of ceramic particle surfaces, especially sharp edges. The preferential dissolution of particles at edges with a small radius of curvature causes the particles to be rounded and thus facilitates particle sliding. When external pressure is applied, the transient liquid phase redistributes and fills the interstitial spaces, resulting in a closer packing of particles. The

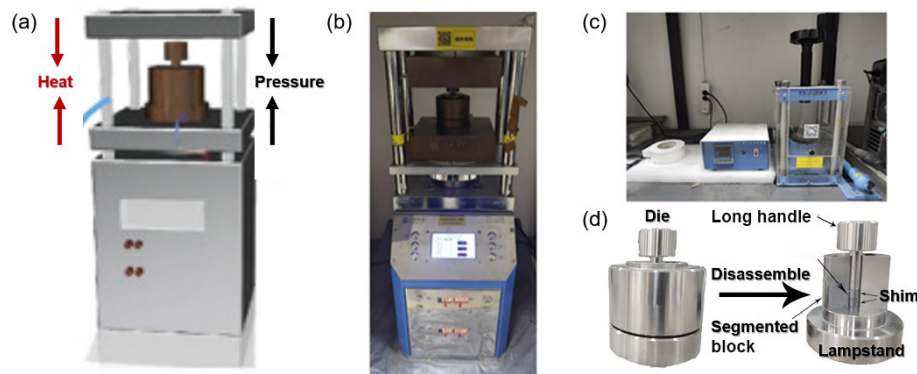


Fig. 28 (a) Schematic illustration and (b) photograph of experimental setup for cold sintering process. Photographs of (c) heater jacket and (d) die. Reproduced with permission from Ref. [216], © Elsevier Ltd. 2022.

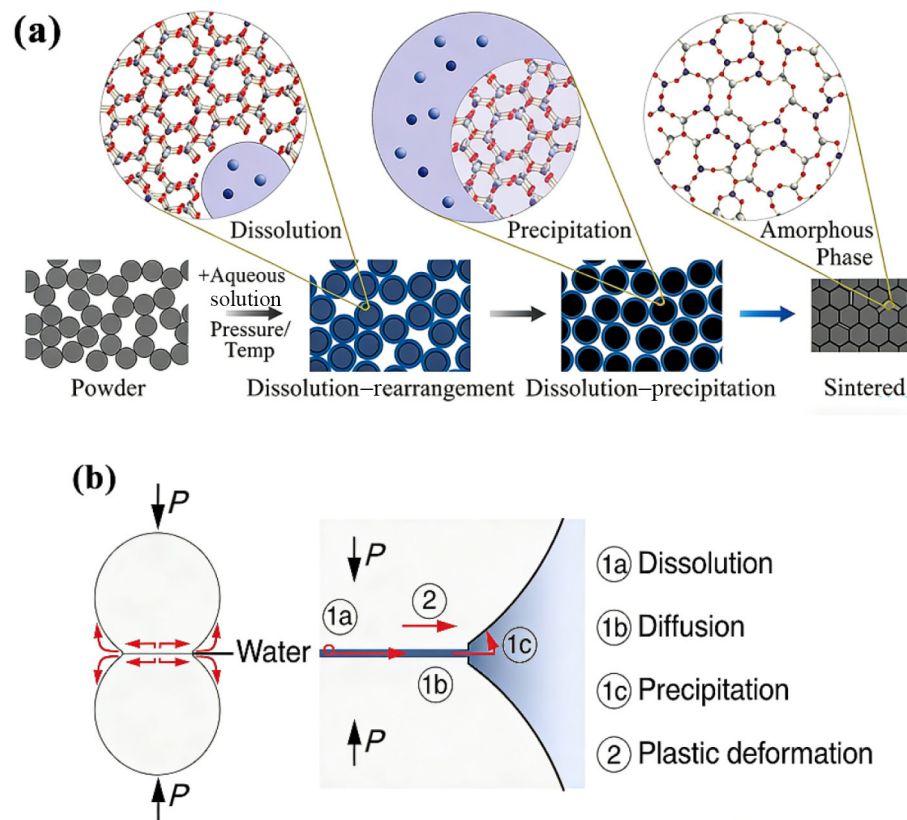


Fig. 29 (a) Schematic diagram of dissolution–precipitation mechanism during cold sintering process. Reproduced with permission from Ref. [32], © WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim 2016. (b) Schematics of plastic deformation mechanism. Reproduced with permission from Ref. [145], © The Author(s) 2017.

second stage encompasses the dissolution–precipitation process. Solid particles are partially dissolved in the transient liquid phase. As the temperature rises, the diffusion and evaporation of the transient liquid phase are accelerated, resulting in the dissolved ions precipitating at the surfaces of particles from the saturated solution. Concurrently, ions dissolved in the liquid migrate outwards and precipitate in regions characterized by diminished chemical potentials. This process promotes grain growth and significantly enhances densification. The third stage consists of the evaporation of residual liquid and the conversion of transient phases into crystalline phases, which may take place simultaneously during cold sintering or in subsequent post-heat treatments. If the amount of transient liquid phase is high enough and the sintering time is short, the liquid phase persists during the whole cold sintering process, and residual remains in the final ceramics. Drying in an oven or annealing in a furnace is usually utilized to remove the residual. If the amount of transient liquid phase is low or the sintering time is long enough, the liquid phase is completely consumed in the final stage of CSP. The amorphous phase formed during CSP will be gradually crystallized under mild temperature and pressure conditions. However, the rate of crystallization is low due to the low sintering temperature, and an amorphous phase may exist in the final ceramics.

The plastic deformation mechanism was developed by Hong *et al.* [144], Bouville and Studart [145], Haug *et al.* [146], and Gonzalez-Julian *et al.* [147] during their studies on the cold sintering densification, as shown in Fig. 29(b). Densification is triggered by stress-induced dislocation movement from grains towards grain boundaries, leading to grain boundary migration. Randall, Guo, and colleagues summarized the potential mechanisms for densification during the cold sintering process, highlighting three key processes: (1) liquid-enhanced creep at solid–solid interfaces; (2) Marangoni flow at liquid–liquid interfaces describing mass transfer driven by surface tension gradients; and (3) gradient diffusion at solid–liquid interfaces [138]. Furthermore, pressure-induced plastic deformation may

also play a role in the densification process. In the study of cold-sintered ceramics–polymer composites, the fluidity of the polymer has also been considered as one of the factors promoting densification. The densification of $(1-x)\text{LM}-x\text{PTFE}$ ($\text{LM} = \text{Li}_2\text{MoO}_4$, $\text{PTFE} = \text{polytetrafluoro-ethylene}$), $(1-x)\text{LAGP}-x(\text{PVDF-HFP})$ ($\text{LAGP} = \text{Li}_{1.5}\text{Al}_{0.5}\text{Ge}_{1.5}(\text{PO}_4)_3$, $\text{PVDF-HFP} = \text{poly(vinylidene fluoride-co-hexafluoropropylene)}$), and $(1-x)\text{V}_2\text{O}_5-x\text{PEDOT:PSS}$ ($\text{PEDOT:PSS} = \text{poly(3,4-ethylenedioxythiophene) polystyrene sulfonate}$) composites with high volume fractions of ceramics can be improved by CSP, in contrast to the hot press process (no water addition) [148]. The simultaneous contribution from the dissolution–precipitation process and viscous flow accelerates the densification of the $(1-x)\text{BaTiO}_3-x\text{PEI}$ ($\text{PEI} = \text{poly(ether imide)}$) nanocomposites [142]. Given that the study of the cold sintering process is in an early stage, a comprehensive understanding of the underlying mechanisms continues to be an important aspect of future research.

(3) Sintering parameters

The added transient liquid phase and corresponding sintering pathways vary substantially with different materials. Guo HZ *et al.* [135], Guo J *et al.* [138], and Ding *et al.* [149] proposed several potential cold sintering pathways based on the solubility characteristics of different material systems, as illustrated in Fig. 30. For congruently dissolving materials, where the dissolution and precipitation rates of anions and cations are similar (especially those with high solubility in water, such as KH_2PO_4 , NaNO_2 , NaCl , and alkali metal molybdates), the cold sintering process is relatively straightforward. This process involves the use of an adequate amount of aqueous solution as the transient liquid phase to aid in densification. The ceramic particles are dissolved readily into water with uniform stoichiometry, and the dissolved substances precipitate progressively onto the surfaces of growing particles within the cold sintering environment.

However, for incongruently dissolving materials, where there is a notable disparity in the dissolution and precipitation rates of

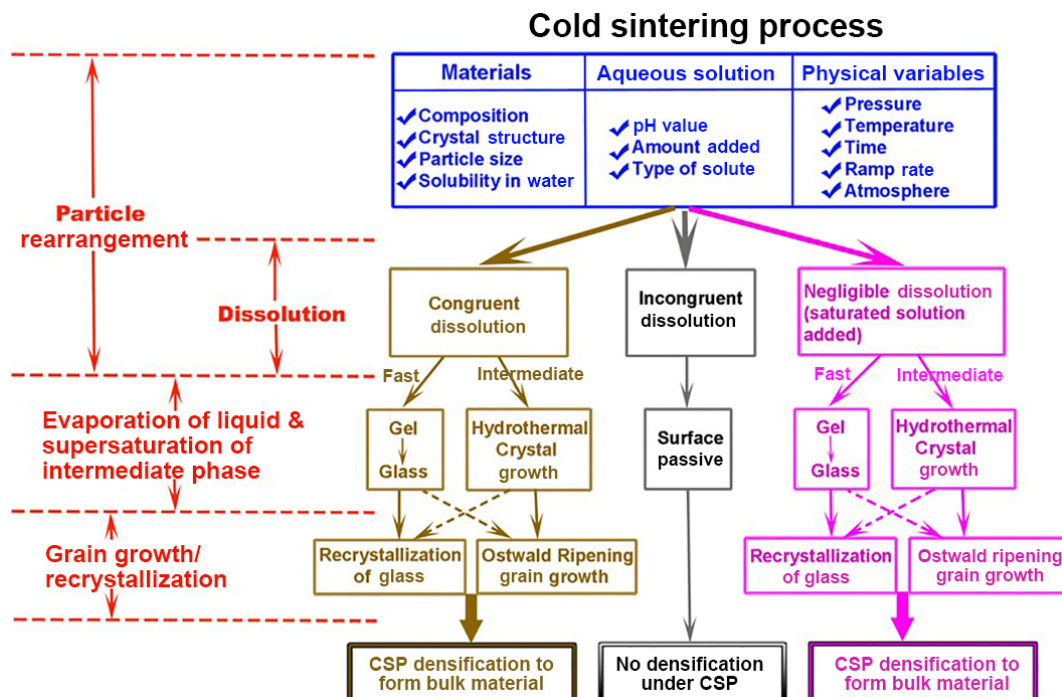


Fig. 30 Flowchart summary of the cold sintering process regarding the fundamental mechanisms and possible routes for different types of material systems. Reproduced with permission from Ref. [135], © The American Ceramic Society 2016.

anions and cations, or materials that form transient phases during dissolution, the formation of passivating surfaces can separate the liquid phase from the ceramic particles, which impedes the precipitation process and further densification. The cold sintering of BaTiO₃ ceramics demonstrates that this issue can be overcome by introducing a suitable transient liquid phase, such as a saturated solution enriched with the corresponding chemical components of the target compound. BaTiO₃ is a representative material that exhibits inconsistent dissolution rates of Ba²⁺ and Ti⁴⁺, leading to surface passivation [135,139,140]. The use of a suspension containing Ba(OH)₂ and TiO₂ can mitigate the inconsistent dissolution of BaTiO₃ particles and minimize surface passivation but may result in the formation of the BaCO₃ impure phase, which requires annealing at high temperatures [139]. Alternatively, one-step cold sintering at 300 °C with a NaOH–KOH eutectic flux can densify BaTiO₃ ceramics. However, this flux is corrosive, especially at 300 °C, and may damage the steel mold [150]. Ba(OH)₂·8H₂O and TiO₂/H₂TiO₃ are promising transient liquid phases for the cold sintering of BaTiO₃ at 150–300 °C without further heat treatment [151–153]. Comparable sintering techniques can be applied to materials with low solubility, such as TiO₂. The hydrolysis reaction of tetrabutyl titanate can be utilized to cold-sinter TiO₂ ceramics [154]. Additionally, acidic or alkaline solutions are effective transient liquid phases to enhance material surface activity. For instance, ZnO shows only moderate densification (65%) when deionized water is used as the transient liquid phase but achieves high densities of 99% when acetic acid or zinc acetate solutions are employed [155]. Consequently, selecting the appropriate transient liquid phase for different material systems is vital, as it directly determines the final densities and performances of ceramics. It is noted that materials that are easy to dissolve can be densified using the cold sintering process. If the materials are not easily dissolved, the cold sintering process may be challenging. With suitable acidic solutions, alkaline solutions, or saturated solutions enriched with the target compound, the non-dissolvable materials may be densified using CSP. It is anticipated that an increasing number of materials will be fabricated using the cold sintering process with the development of transient liquid phases.

2.2.3 Current status of cold sintered materials

(1) Features of cold sintering

The cold sintering process shows significant advantages in the extremely low sintering temperature of room temperature–300 °C and rapid densification of tens of minutes or even several minutes, and thus, it is promising for energy saving, environmental protection, and green manufacturing. In comparison to other sintering methods, the cold sintering process significantly reduces energy consumption, with its sintering energy consumption being merely 1/10 to 1/100 of that of conventional thermal sintering [156]. The exceptionally low sintering temperature of the cold sintering process provides new opportunities that are challenging using other sintering methods. Specifically, it is feasible to integrate ceramics with disparate materials, such as ceramic–metal, ceramic–polymer, ceramic–two-dimensional (2D) material, ceramic–nanofiber, and so on, which facilitates the development of novel high-performance composites and caters to the demands of high-performance device development. Furthermore, the low temperature and short time of the cold sintering process can effectively inhibit excessive growth of ceramic grains and circumvent the uncontrollable defects often caused by the volatilization of chemical additives during conventional high-temperature sintering processes.

A wide variety of materials in different applications have been

developed using the cold sintering process, as shown in Fig. 31. Currently, research on the cold sintering process is gradually maturing. A large number of ceramics have been reported to be densified using the cold sintering process, such as oxide, chloride, fluoride, bromide, carbonate, molybdate, tungstate, vanadate, and phosphate [157–159]. Due to the low-temperature characteristics of polymers, it is challenging to co-fire ceramics with polymers using conventional sintering methods at high temperatures. However, the cold sintering process enables the densification of ceramic-based composites with the addition of a small amount of polymer [151–153,160,161]. Furthermore, Guo *et al.* [162] utilized the cold sintering process to fabricate a multilayer ceramic structure formed by co-firing ceramics with silver electrodes. This structure serves as a prototype for multilayer devices, indicating that the cold sintering process can be applied to the heterogeneous co-firing of multilayer components. The cold sintering process opens up a new direction for the development of ceramics and ceramic-based composites [163]. The research status of the cold sintering process in different applications (from microwave dielectric materials to optical materials) will be systematically introduced.

(2) Microwave dielectric materials

Microwave dielectric ceramics are electronic functional materials that are utilized in electronic devices at microwave frequencies (300 MHz–300 GHz). They play an important role in wireless communication technology and are critical materials in resonators, filters, waveguides, antennas, etc. [164,165]. Good microwave dielectric ceramics usually meet the following characteristics: a suitable dielectric constant (ϵ_r), a high quality factor (Q), and a near-zero temperature coefficient of resonance frequency (TCF/τ_f). As shown in Fig. 32, typical dielectric ceramics can be categorized into high-temperature co-fired ceramics (HTCC), low-temperature co-fired ceramics (LTCC), ultralow-temperature co-fired ceramics (ULTCC), and cold-sintered co-fired ceramics (CSCC) based on the numerical difference in sintering temperature. The typical sintering temperatures for HTCC, LTCC, ULTCC, and CSCC are 1200–1800, 900–1000, 400–700, and 20–300 °C, respectively [162]. Due to the low sintering temperature of the cold sintering process, microwave dielectric ceramics can be co-fired with metal

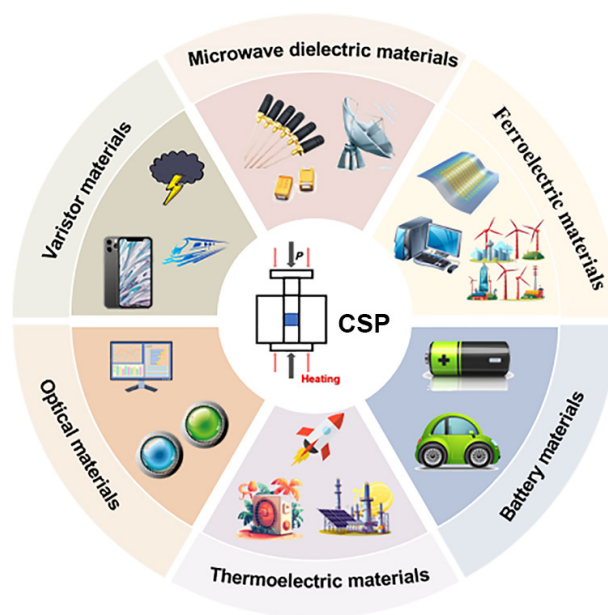


Fig. 31 Applications of the cold sintering process in a wide range of fields.

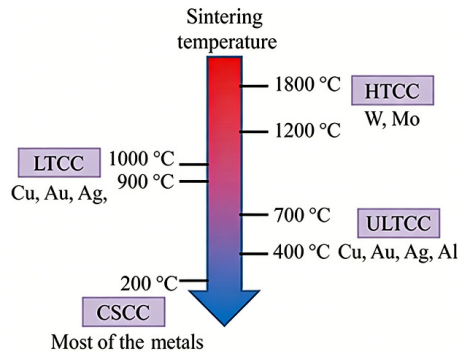


Fig. 32 Schematic for the development of co-fired ceramic technology with decreasing sintering temperature. Reproduced with permission from Ref. [162], © The American Ceramic Society 2016.

electrodes with high conductivity and low melting points, such as Cu, Ag, and Al, to meet the requirements for the miniaturization of microwave components. Sections (i)–(vi) introduces the application of the cold sintering process in different microwave dielectric material systems, including molybdate, tungstate, borate, chloride, fluoride, and phosphate, most of which can be densified using the cold sintering process with deionized (DI) water or hydrate, as shown in Table 5. However, the cold sintering of TiO_2 , HBO_2 , and Al_2O_3 may involve tetrabutyl titanate, ethanol, and NaOH/acetic acid, respectively. Compared with conventional thermal sintering, cold-sintered ceramics with lower sintering temperatures and shorter sintering time show comparable or better microwave dielectric properties, making them promising for 5G/6G applications.

(i) Molybdate-based microwave dielectric materials

Molybdate-based ceramic has excellent microwave dielectric properties and is the first material system prepared by the cold sintering process. It is demonstrated that Li_2MoO_4 , $\text{Na}_2\text{Mo}_2\text{O}_7$, and $\text{K}_2\text{Mo}_2\text{O}_7$ ceramics could be well densified with relative densities of > 90% prepared by the cold sintering process at 120 °C using DI water as the transient liquid phase. The permittivities and $Q \times f$ values of cold-sintered Li_2MoO_4 , $\text{Na}_2\text{Mo}_2\text{O}_7$, and $\text{K}_2\text{Mo}_2\text{O}_7$ ceramics are comparable to those of conventional thermally sintered samples [32]. On this basis, Guo *et al.* [162] prepared Li_2MoO_4 -PTFE and $(\text{LiBi})_{0.5}\text{MoO}_4$ -PTFE composites using a cold sintering process. The TCF value is increased from -170×10^{-6} to $-7.2 \times 10^{-6} \text{ } ^\circ\text{C}^{-1}$ when PTFE rises from 0 to 70%, indicating that the addition of PTFE improves the temperature stability of Li_2MoO_4 ceramic. Wang *et al.* [166] prepared $\text{Na}_{0.5}\text{Bi}_{0.5}\text{MoO}_4$ - Li_2MoO_4 composite ceramics using the cold sintering process and achieved a zero TCF value by adjusting the weight fraction of Li_2MoO_4 . The potential applications of novel RF

substrates and graded radial index (GRIN) dielectric lenses are demonstrated. Li *et al.* [167] prepared dense $x\text{Li}_2\text{MoO}_4$ – $(1-x)(\text{LiBi})_{0.5}\text{MoO}_4$ microwave dielectric ceramics by a cold sintering process under mild sintering conditions (120 °C/30 min/200 MPa). Li_2MoO_4 – $(\text{LiBi})_{0.5}\text{MoO}_4$ ceramics are compatible with Ag and Al electrode materials under cold sintering conditions. The specimens exhibit good microwave dielectric properties, and a near-zero TCF (+0.7 ppm/°C) can be obtained with $x = 50 \text{ vol}\%$. This indicates that the cold sintering process is promising for the development of temperature-stable microwave dielectric ceramics with low CO_2 emission.

(ii) Tungstate-based microwave dielectric materials

Tungstate shows excellent microwave dielectric properties. However, due to the high melting point, its sintering temperature is typically high. The emergence of the cold sintering process provides a new opportunity to solve the aforementioned problem. Hao *et al.* [168] successfully prepared Na_2WO_4 ceramics with controllable grain sizes via a cold sintering process at 240 °C. In addition, Na_2WO_4 ceramics can be densified using the cold sintering process with $\text{Na}_2\text{WO}_4 \cdot 2\text{H}_2\text{O}$ dry powder as the “transient liquid phase”. The $\text{Na}_2\text{WO}_4 \cdot 2\text{H}_2\text{O}$ hydrate exhibits the advantage of uniform dispersion of liquid water. Insufficient water content leads to incomplete cold sintering, whereas excessive water content results in reduced density. The sintering temperature influences the densification mechanisms of plastic deformation and dissolution–precipitation. External pressure accelerates the densification process, and sufficient sintering time ensures the completion of the cold sintering process. Ultimately, the optimum cold sintering parameters for Na_2WO_4 ceramics using $\text{Na}_2\text{WO}_4 \cdot 2\text{H}_2\text{O}$ powders are determined to be 150 °C and 240 MPa for 60 min [169].

(iii) Borate-based microwave dielectric materials

The boron ions exhibit extremely low polarizability, and most borates have a relative permittivity of less than 8, making them promising for millimeter-wave applications. HBO_2 ceramic can be prepared using the conventional sintering method but shows a relatively low relative density of less than 80%, which results from the presence of pores, residual H_3BO_3 , and amorphous B_2O_3 [170]. Since HBO_2 reacts with H_2O to produce H_3BO_3 , Hong *et al.* [171] used 10 wt% dehydrated ethanol instead of water as the transient liquid phase and employed a cold sintering process to prepare dense HBO_2 -II ceramics. A dense HBO_2 -II ceramic with a relative density of 94.7% can be obtained using a cold sintering process at 120 °C. In comparison, HBO_2 -II ceramics prepared without the addition of ethanol are fragile, with the relative density reduced by 6.0%–10.2%, indicating that ethanol has a significant impact on the densification process. Cold sintering at 120 °C and 500 MPa for 10 min results in the best microwave dielectric properties for

Table 5 Comparisons of cold sintered materials in microwave dielectric applications (some of the materials may be utilized in other applications, such as Al_2O_3 and fluoride)

Material	Cold sintering transient liquid phase	Key parameters compared with those prepared by TS	Performances compared with those prepared by TS
Molybdate, tungstate and phosphate	DI water or hydrate (easy to be densified)	Much lower ST and shorter T	Comparable
TiO_2	Tetrabutyl titanate and DI water (annealing at higher temperatures may be required)	Lower ST and shorter T	
Chloride	DI water (easy to be densified)	Much lower ST and shorter T	Better
Fluoride and calcite	DI water (annealing at higher temperatures may be required)	Lower ST and shorter T	
Borate	DI water, ethanol or acetic acid (annealing may or may not be required)	Lower ST and shorter T	
Al_2O_3	DI water, hydrate, NaOH, or acetic acid (annealing at higher temperatures may be required)	Lower ST and shorter T	

Note: ST, sintering temperature. T , total time for sintering. TS, traditional thermal sintering.

single-phase HBO_2 -II ceramic: $\epsilon_r = 4.21$, $Q \times f = 47,500$ GHz, and $\tau_f = -70.0 \times 10^{-6} \text{ } ^\circ\text{C}^{-1}$. Both the relative density and $Q \times f$ value of cold sintered samples are significantly higher than those of reported HBO_2 ceramics prepared by the conventional sintering route (70.3% and 32,700 GHz, respectively). Li *et al.* [172] utilized acetic acid solution as a transient liquid phase to prepare $\text{Zn}_3\text{B}_2\text{O}_6$ ceramics via CSP at 200 $^\circ\text{C}$ for 60 min under 300 MPa, achieving a relative density of 92%. Subsequent annealing at 550 $^\circ\text{C}$ further improved the room-temperature microwave dielectric properties of $\text{Zn}_3\text{B}_2\text{O}_6$ ceramics, resulting in $\epsilon_r = 5.95$, $Q \times f = 20,620$ GHz, and $\tau_f = -67 \times 10^{-6} \text{ } ^\circ\text{C}^{-1}$.

(iv) Chloride-based microwave dielectric materials

NaCl is difficult to densify at temperatures lower than 600 $^\circ\text{C}$. However, Guo *et al.* [32] prepared dense NaCl ceramics (90%, Figs. 33(a) and 33(b)) at room temperature using a cold sintering process with DI water as the transient liquid phase, which had a higher relative density than that (84%, Figs. 33(c) and 33(d)) sintered at 600 and 700 $^\circ\text{C}$. Hong *et al.* [144] investigated the microwave dielectric properties of cold sintered and dry pressed NaCl ceramics. The $Q \times f$ value of cold sintered NaCl ceramics is significantly higher than that of dry-pressed ceramics, as the presence of a transient liquid phase promotes microstructural uniformity. The optimal microwave dielectric properties of cold sintered NaCl ceramics are $\epsilon_r = 5.55$, $Q \times f = 49,600$ GHz, and $\tau_f = -173 \times 10^{-6} \text{ } ^\circ\text{C}^{-1}$. Furthermore, Santha *et al.* [173] fabricated dense MgTiO_3 - NaCl composites exhibiting good quality factors via a cold sintering process. $Q \times f$ exhibited nonlinear variations, peaking at 29,500 GHz with $x = 0.2$.

(v) Fluoride-based microwave dielectric materials

LiF is a low-firing fluoride with excellent microwave dielectric properties; however, the densification of LiF ceramics is challenging owing to their low surface free energy. Liu *et al.* [174] utilized the cold sintering process (150 $^\circ\text{C}$, 250 MPa, 60 min) to prepare compact LiF ceramics. Annealing treatment was performed on the cold-sintered sample to further achieve densification, and the relative density increased to 92%. The $Q \times f$ value increases with the annealing temperature, reaching a maximum value of 110,800 GHz at 800 $^\circ\text{C}$, which is 1.5 times higher than that of conventional sintering (78,800 GHz). The high $Q \times f$ value and compatibility with Ag electrodes indicate that cold-sintered LiF has great potential as a component or additive in low-temperature co-fired ceramic formulations. CaF_2 ceramics with a $Q \times f$ value of 80,522 GHz can be densified by the cold sintering process [175], which is three times higher than that of conventional sintering at 1000 $^\circ\text{C}$ (26,448 GHz). In addition, CaF_2 ceramics assisted by CSP can be utilized as the dielectric substrate to prepare C-band microstrip patch antennas, revealing their potential application prospects in antennas.

(vi) Other microwave dielectric materials

Wang *et al.* [176] prepared dense LiMgPO_4 ceramics and

LiMgPO_4 - CaTiO_3 - K_2MoO_4 composites using the cold sintering process. As the sintering temperature and time increased, the relative density of the LiMgPO_4 ceramics rose from 82% at 150 $^\circ\text{C}$ for 30 min to 93% at 250 $^\circ\text{C}$ for 60 min, with ϵ_r and $Q \times f$ values of 6.5 and 16,000 GHz, respectively. Under identical sintering conditions, the cold-sintered LiMgPO_4 - CaTiO_3 - K_2MoO_4 composite achieved a relative density of 97%, exhibiting $\epsilon_r = 9.1$, $Q \times f = 8500$, and $\text{TCF} = -6 \times 10^{-6} \text{ } ^\circ\text{C}^{-1}$. MgTiO_3 -based ceramic with a relative density of more than 96% can be obtained using a cold sintering process at 200 $^\circ\text{C}$ under an applied pressure of 250 MPa for 1 h, followed by postannealing at 1100 $^\circ\text{C}$ [177]. The MgTiO_3 -15wt% B_2O_3 ceramic exhibits good microwave dielectric properties, including an ϵ_r of 14.5, a high $Q \times f$ value of 39,000 GHz, and a TCF value of $-54.36 \times 10^{-6} \text{ } ^\circ\text{C}^{-1}$.

(3) Ferroelectric and piezoelectric materials

Piezoelectric ceramics are a type of functional material capable of mutually converting mechanical energy and electrical energy. Among piezoelectric ceramics, there is a class of ceramics that exhibit ferroelectric properties, which have spontaneous polarization, referred to as ferroelectric ceramics. Ferroelectric and piezoelectric ceramics have extensive applications in sensing, energy conversion, energy storage, cooling, catalysis, flexible devices, and so on [178–183]. Some typical ferroelectric and piezoelectric materials are discussed below, including BaTiO_3 and $\text{Pb}(\text{Zr,Ti})\text{O}_3$ (PZT). The cold sintering of these ceramics with perovskite structures is complicated due to incongruent dissolution, as shown in Table 6. With lower sintering temperatures and short sintering time, the cold sintering process is promising for the fabrication of high-performance piezoelectric and ferroelectric materials with low CO_2 emissions and energy consumption.

(i) BaTiO_3 -based materials

BaTiO_3 ceramic serves as a quintessential example of incongruent dissolution material, where the dissolution rates of Ba^{2+} and Ti^{4+} diverge, ultimately leading to surface passivation and hindering the dissolution-precipitation process during the cold sintering process. To address this challenge, various solutions have been proposed, such as $\text{Ba}(\text{OH})_2/\text{TiO}_2$, molten Na/K mixed alkalis, $\text{Ba}(\text{OH})_2 \cdot 8\text{H}_2\text{O}$ and $\text{Ba}(\text{OH})_2 \cdot 8\text{H}_2\text{O}/\text{H}_2\text{TiO}_3$.

The suspension of $\text{Ba}(\text{OH})_2$ and TiO_2 can be used as a transient liquid phase to prevent incongruent dissolution of BaTiO_3 particles, ultimately enabling the densification of BaTiO_3 ceramics [135,184]. However, the presence of the BaCO_3 phase necessitated subsequent high-temperature annealing to eliminate impurities. To circumvent the formation of the BaCO_3 phase during the cold sintering process, Tsuji *et al.* [150] utilized molten Na/K mixed alkalis as the transient liquid phase, successfully preparing pure-phase BaTiO_3 ceramics without further heat treatment. At room temperature and a frequency of 10^6 Hz, BaTiO_3 ceramics cold-sintered at 300 $^\circ\text{C}$ exhibit a permittivity ranging from 700 to 1800

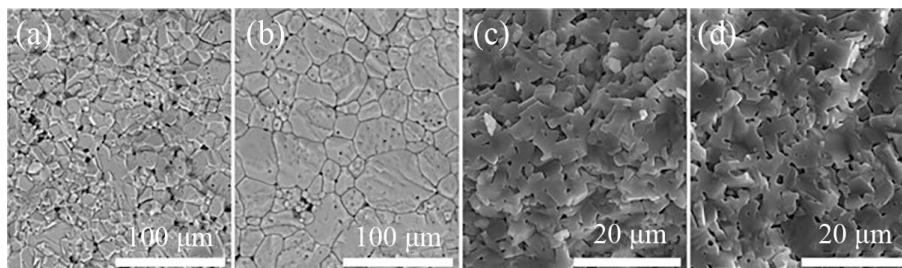


Fig. 33 Microstructures of NaCl [32]: (a) Cold sintered at room temperature and 75% relative humidity, (b) cold sintered at room temperature and 85% relative humidity, (c) conventionally thermal sintered at 600 $^\circ\text{C}$, (d) conventionally thermal sintered at 700 $^\circ\text{C}$. Reproduced with permission from Ref. [32], © WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim 2016.

and a dielectric loss tangent ($\tan\delta$) of ~ 0.04 , which is comparable to the highest-quality data previously reported for BaTiO₃ ceramics prepared by conventional high-temperature sintering techniques ($> 1000^\circ\text{C}$). Ullah *et al.* [185] used Ba(OH)₂·8H₂O as a solvent material for BaTiO₃ during a CSP pretreatment prior to subsequent heat treatment. With the pretreatment, the relative density of BaTiO₃ was significantly enhanced with controlled grain size without harmful secondary phases, allowing for a higher dielectric constant compared to the case without the CSP pretreatment.

For BaTiO₃-polymer composites, the polymer is prone to degradation under strong alkaline conditions at 300°C . To address this issue, Sada *et al.* [153] selected Ba(OH)₂·8H₂O as the transient liquid phase and achieved dense BaTiO₃-PTFE nanocomposites with excellent electrical properties at a cold sintering temperature 50% lower than that using Na/K mixed alkalis. The incorporation of polymers significantly improved the performance of BaTiO₃ ceramics. With a polymer volume fraction of 5%, the resistivity exceeded $10^{12}\ \Omega\cdot\text{cm}$, which is four orders of magnitude higher than that of pure BaTiO₃ ceramics. The BaTiO₃-PTFE nanocomposite boasted a dielectric permittivity as high as 790 at 1 MHz and a low dielectric loss of 0.014, demonstrating the potential of CSP in designing novel ferroelectric nanocomposites. Furthermore, Sada *et al.* [152] mixed surface-modified BaTiO₃ with polyphenylene oxide (PPO) using Ba(OH)₂·8H₂O as the transient liquid phase, resulting in dense BaTiO₃-PPO nanocomposites with outstanding dielectric properties. These nanocomposites exhibit high permittivities (up to 1000–2000) and low dielectric losses (as low as 0.05) at room temperature, along with resistivity of 10^{13} – $10^{14}\ \Omega\cdot\text{cm}$. Vinoth *et al.* [186] employed CSP to produce BaTiO₃-PVDF nanocomposites with a relative density of 94.8%. These composites show excellent dielectric properties at 1 GHz, featuring a permittivity of 71.1 and a low dielectric loss of 0.04. Li *et al.* [142] prepared $(1-x)\text{BaTiO}_3$ - $x\text{PEI}$ nanocomposites by a one-step cold sintering

process using Ba(OH)₂·8H₂O and H₂TiO₃ as the transient liquid phase. Superior performances with a relative permittivity of 163 and a low dielectric loss of 0.014 are obtained in 80BaTiO₃-20PEI nanocomposites, and the electrical breakdown strength is increased by 80.65% compared with BaTiO₃.

(ii) PZT-based materials

Conventionally, the sintering temperature of PZT often exceeds 1200°C . Researchers have delved into various approaches to lower the maximum processing temperature of PZT, including hot pressing, liquid-phase sintering, microwave sintering, and field-assisted sintering (such as spark plasma sintering). However, the sintering temperature of PZT ceramics still exceeds 1000°C [187–189]. The advent of the cold sintering process has significantly reduced the sintering temperature of PZT materials. Given that PZT materials share a similar perovskite structure with BaTiO₃ and exhibit incongruent dissolution characteristics, identifying a suitable transient liquid phase is of paramount importance. Several transient liquid phases were developed for the cold sintering process of PZT, such as Pb(NO₃)₂, Pb(CH₃COO)₂·3H₂O, and Ba(OH)₂·8H₂O.

Pb(NO₃)₂: Wang *et al.* [190] employed Pb(NO₃)₂ solution as a transient liquid phase to densify PZT powders. Through the cold sintering process at 300°C and 500 MPa, the resultant PZT ceramics achieved a relative density of 89%. After annealing at 900°C for 3 h, the relative density of the samples increased to 99%, and the resulting permittivity reached 1300. The samples displayed a clear ferroelectric hysteresis loop with a remanent polarization strength of $28\ \mu\text{C}/\text{cm}^2$, which is higher than that prepared by conventional thermal sintering, as illustrated in Fig. 34. Upon polarization, the piezoelectric coefficient d_{33} was approximately 200 pC/N. Additionally, Wang *et al.* prepared 5–10 μm thick PZT/metal film (Ni, Ag, and Cu) composites via a cold sintering process using Pb(NO₃)₂ solution as the transient liquid phase [190]. CSP effectively prevented cracking and delamination of PZT on metal surfaces. Co-fired PZT/Cu

Table 6 Comparisons of cold-sintered materials in ferroelectric and piezoelectric applications

Material	Cold sintering transient liquid phase	Key parameters compared with those prepared by TS	Performances compared with those prepared by TS
BaTiO ₃	Ba(OH) ₂ /TiO ₂ , molten Na/K mixed alkalis, Ba(OH) ₂ ·8H ₂ O or Ba(OH) ₂ ·8H ₂ O/H ₂ TiO ₃ (annealing is not required for most of compositions)	Much lower ST and shorter T	
Pb(Zr,Ti)O ₃ (PZT)	Pb(NO ₃) ₂ , Pb(CH ₃ COO) ₂ ·3H ₂ O or Ba(OH) ₂ ·8H ₂ O (annealing at higher temperatures may be required)	Lower ST and shorter T	Comparable
(K,Na)NbO ₃ (KNN)	DI water or NaOH–KOH (Annealing at higher temperatures may be required)	Lower ST and shorter T	

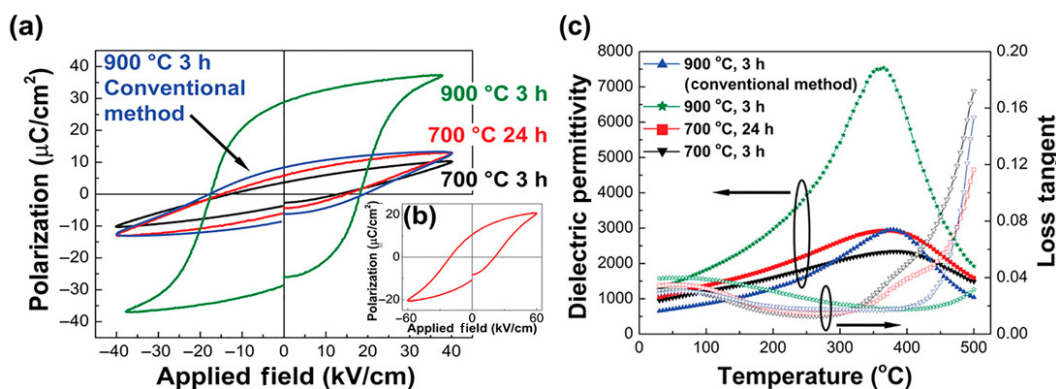


Fig. 34 (a) Polarization–electrical (P–E) hysteresis loops of PZT post-annealed at 700°C for 3 and 24 h and at 900°C for 3 h. The samples were measured at 10 Hz. (b) P–E loop of $700^\circ\text{C}/24\text{ h}$ annealed PZT with a higher maximum applied field (60 kV/cm) at 10 Hz. (c) Permittivity and loss of the post-annealed PZT at different temperatures under 100 kHz. Reproduced with permission from Ref. [192], © Author(s) 2017.

exhibited a lower coercivity and a larger effective transverse piezoelectric constant e_{31f} of -4.7 C/m^2 .

$\text{Pb}(\text{CH}_3\text{CO}_2)_2 \cdot 3\text{H}_2\text{O}$: Gupta *et al.* [191] employed lead acetate trihydrate as the transient liquid phase for CSP, achieving a relative density of 84% for PZT cold-sintered at 200°C . After post-annealing at 900°C , the relative density of the cold-sintered samples surpassed 97%. The annealed samples exhibited a room-temperature permittivity of approximately 1600 at 100 Hz with a tangent loss of approximately 0.038. Notably, cold-sintered PZT with lead acetate trihydrate exhibited a higher permittivity and lower loss tangent than those cold-sintered with $\text{Pb}(\text{NO}_3)_2$.

$\text{Ba}(\text{OH})_2 \cdot 8\text{H}_2\text{O}$: Ichangi *et al.* [193] utilized $\text{Ba}(\text{OH})_2 \cdot 8\text{H}_2\text{O}$ as a transient liquid phase to fabricate PZT ceramics with well-defined grain morphology via CSP. The XRD patterns of cold-sintered samples at 290°C revealed the presence of a lead oxide phase, confirming the dissolution of Pb ions on the surface of the PZT particles. Annealing at 900°C resulted in pure-phase materials with a perovskite structure, where Ba ions diffused into the PZT lattice. The annealed samples exhibited polyhedral grains in three-point equilibrium, with a relative density of 93%. The d_{33} of the samples increased with the Ba content in PZT, reaching $201 \pm 80 \text{ pm/V}$. The annealed samples displayed ferroelectric switching current behavior with a saturated polarization loop.

(iii) KNN-based materials

High-performance lead-free piezoelectric ceramics have garnered considerable attention and can be utilized in numerous applications, such as sensors and ultrasonic transducers [194–196]. Among all lead-free piezoelectric ceramics, KNN-based ceramics have received significant attention owing to their high Curie temperature and excellent piezoelectric performance. Since KNN exhibits an ABO_3 perovskite structure similar to BaTiO_3 , it also shares similar dissolution characteristics with BaTiO_3 .

Ma *et al.* [197] fabricated KNN ceramics with a relative density of 98% using the cold sintering-assisted sintering (CSAS) method. Compared to conventional sintering, CSP with deionized water as the transient liquid phase significantly enhances the green density of KNN ceramics. The increase in green density effectively lowers the sintering temperature and greatly elevates the sintered density of the final product. CSAS ceramics sintered at 1115°C exhibit excellent ferroelectric, piezoelectric, and dielectric properties, with a piezoelectric coefficient of 131 pC/N , a permittivity of 10,922, a dielectric loss of 0.0486 at 10 kHz, a residual polarization of 17.3 C/cm^2 , and a coercive field of 8.3 kV/cm .

Tsuji *et al.* [198] employed a cold sintering process utilizing NaOH–KOH as a transient additive to prepare KNN ceramics with a relative density ranging from ~92% to 96% under sintering conditions of $230\text{--}300^\circ\text{C}$ and 400 MPa. The NaOH–KOH fluxes are designed around the eutectic composition and overcome the incongruent dissolution problems of the perovskite structure, enabling the dissolution and reprecipitation process of the cold sintering process. The low-field dielectric and electrical properties of cold-sintered KNN ceramics are comparable to those of conventionally sintered samples. A high electric field strength can be applied at $\sim 80 \text{ kV/cm}$, without high losses, indicating the existence of a reasonable dielectric strength. Transmission electron microscopy (TEM) studies revealed high-density linear defects in

most of the grains, with dislocations within grains limiting polarization and domain wall motion, which suggests that future work on piezoelectric ceramics needs to consider not only grain boundary interfaces but also the types and concentrations of defects.

Nakagawa *et al.* [199] prepared KNN ceramics with a relative density of 93% using NaOH–KOH as transient additives at 300°C under 400 MPa. After secondary heat treatment (annealing at 700°C), the ferroelectric hysteresis and piezoelectric properties of the sample were significantly improved. The annealed sample has $\epsilon_r = 620$ and $\tan\delta = 0.024$, similar to those before annealing, but the annealed sample exhibits $d_{33} = 88 \text{ pC/N}$, which is significantly higher than that of the unannealed samples ($d_{33} = 35 \text{ pC/N}$). In addition to densification, an unusual *in situ* plastic deformation process occurred during the CSP. High-density dislocations within grains led to multiple pinning sites, affecting high-field dielectric and piezoelectric properties. Annealing significantly reduced the dislocation density in high-defect crystals, bringing the piezoelectric and ferroelectric properties of KNN closer to those of conventionally sintered KNN.

(4) Varistor materials

Varistor ceramics are a type of semiconducting ceramic with significant nonlinear current–voltage (I – V) characteristics. Varistors are typically connected in parallel with electronic devices to provide protection against surges or stabilize operating currents in circuits. Currently, varistors are widely utilized in power electronic systems, with typical applications including lightning arresters employed in large buildings, lightning protection devices in railway systems, and surge protection devices for internal circuits of computers and smartphones [200]. The primary materials used for varistors include semiconducting metal oxides, such as ZnO, TiO_2 , SnO_2 , WO_3 , and $\text{CaCu}_3\text{Ti}_4\text{O}_{12}$ (CCTO) [201–206]. Cold-sintered ZnO-based varistors usually show a higher breakdown electric field and a comparable nonlinear coefficient, as summarized in Table 7. With formic acid as the transient liquid phase, the mechanical properties can be further improved. It has been demonstrated that the cold sintering process is not only environmentally friendly but also improves the electrical properties of varistors.

(i) Cold sintering parameters of ZnO

Using water as the transient liquid phase for cold sintering of ZnO ceramic results in a relative density of only 65% [207]. However, the addition of acetic acid aqueous solution significantly alters the density and grain structure of the ZnO ceramic, achieving a relative density exceeding 90%. The average conductivity of ZnO samples cold-sintered at approximately 300°C was 9 S/cm , higher than that (5 S/cm) of conventionally sintered ZnO at 1400°C , indicating that the introduction of acetic acid did not deteriorate the electrical properties of ZnO ceramics. The densities and grain sizes of ZnO can be predicted using a sintering simulation model [208]. The sintering curves of ZnO show two densification regimes, with one before 115°C (regime 1) and one after 115°C (regime 2), at which the transient liquid phase is eliminated. The stress exponents of the creep law reveal that there are two concomitant diffusion mechanisms for regime 1, including grain boundary diffusion and grain boundary sliding, and there is only grain boundary diffusion for regime 2.

Table 7 Comparisons of cold-sintered materials in varistor applications

Materials	Cold sintering transient liquid phase	Key parameters compared with those prepared by TS	Performances compared with those prepared by TS
ZnO based composites	Acetic acid, zinc acetate or formic acid (annealing is not required)	Much lower ST and shorter T	Higher breakdown electric field, better mechanical properties with formic acid

Kang *et al.* [209] explored the impact of ion species, concentration, and pH in the cold sintering liquid phase on the density of ZnO ceramics. They found that when aqueous solutions of HCl, H₂SO₄, ZnCl₂, and ZnSO₄ were used for the densification of ZnO ceramics at room temperature, the maximum density values ranged between 70% and 75%, while using acetic acid as the transient liquid phase yielded a relative density of up to 96%. The optimal concentration of the transient liquid phase during CSP depends on material properties, as well as factors such as extrusion speed, initial particle size, and evaporation rate. Under a uniaxial pressure of 530 MPa at 120 °C for 0.5 h, the ZnO ceramic obtained using water with a pH of 7 had a relative density of only 71%, whereas samples using a 0.83 mol/L zinc acetate solution with a pH of 7 achieved a relative density of 99%. This suggests that the pH of the liquid phase has a limited influence on the density of ZnO ceramics, and instead, the sufficient presence of Zn²⁺ and acetate ions in the liquid phase is crucial for achieving densification. Previous studies on the cold sintering of ZnO typically employed aqueous solutions of acetic acid or zinc acetate at appropriate concentrations as the transient liquid phase.

Recently, Jabr *et al.* [210] investigated the effects of formic acid, acetic acid, and citric acid on the densification, microstructural evolution, and mechanical response of ZnO ceramics during CSP. ZnO ceramics using aqueous solutions of formic acid and acetic acid as the transient liquid phase exhibited higher relative densities (~96%), whereas those using citric acid aqueous solutions had lower relative densities (< 80%). Compared to densification with acetic acid, samples densified using formic acid retain their grain size over all temperature regimes. Employing formic acid as the transient liquid phase for the cold sintering of ZnO enhanced the mechanical properties, with the biaxial characteristic strength of formic acid-sintered samples reaching ~90 MPa, twice that of acetic acid-sintered samples (~40 MPa), and approximately 40%

higher than previously reported data using zinc acetate. In samples densified using acetic acid (Fig. 35(a)), fracture typically originates from delaminated regions in the microstructure. Figure 35(b) shows a sample densified using formic acid, where the failure originated from a flaw (crack) near the surface. Compared to samples densified with acetic acid, formic acid results in a homogeneous microstructure with fracture surfaces showing features similar to a well-densified material. It may be hypothesized that the superior mechanical performance achieved with formic acid could be associated with better grain boundary cohesion.

(ii) Cold-sintered ZnO specimens

The nonlinear electrical characteristics of ZnO varistors stem from their unique grain boundary structure, which is typically achieved by incorporating metal oxides such as Bi₂O₃ or Pr₆O₁₁ [211,212]. These metal oxides are distributed at grain boundaries, forming Schottky barriers that give rise to nonlinearity. Conventional sintering temperatures for ZnO ceramics usually exceed 1000 °C, which consumes significant energy and can lead to the evaporation of low-melting-point additives, deteriorating the electrical properties of the material [117,213]. Furthermore, high sintering temperatures restrict the introduction of materials with low melting points. The advent of the cold sintering process effectively addresses these challenges.

Zhao *et al.* [214] prepared ZnO–PTFE composites with a high relative density using acetic acid solution as the transient liquid phase at 285 °C and 300 MPa. Notably, the ZnO–PTFE composites, with a PTFE volume fraction of 5%, exhibited the highest nonlinear coefficient of approximately 7 and a breakdown electric field of 2304.6 V/mm. The maximum breakdown electric field of ZnO–PTFE composites reached 3225.4 V/mm, surpassing ZnO–Bi₂O₃, SrTiO₃, and other materials by 1–2 orders of magnitude. It is feasible to fabricate dense ZnO-based varistors with controllable grain growth using CSP [215]. The cold sintered

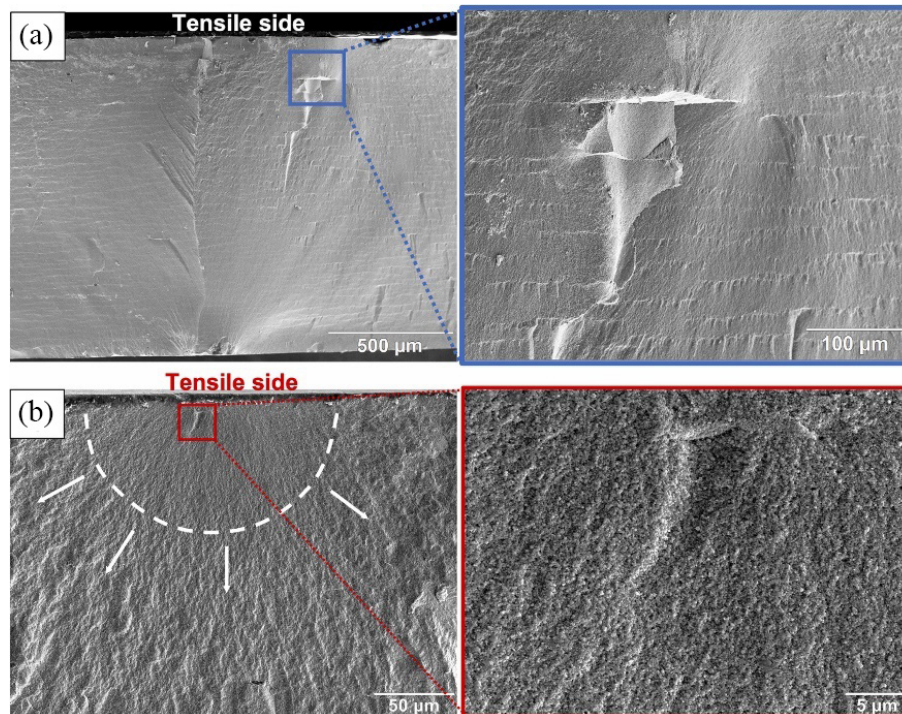


Fig. 35 Fracture surfaces and enlarged view of fracture origins in (a) a sample cold-sintered with acetic acid solution with a failure stress of 36 MPa. The fracture origin is a delaminated area in the subsurface. (b) Cold sintered with formic acid solution with a failure stress of 91 MPa. Reproduced with permission from Ref. [210], © The Author(s) 2022.

ZnO–Bi₂O₃–Mn₂O₃–CoO ceramics demonstrated a nonlinear coefficient of 33.5 and a high breakdown electric field of 3550 V/mm.

In the case of cold-sintered ZnO/poly-ether-ether-ketone (PEEK) composites, two different methods, including direct mixing and dissolution methods, were utilized to mix PEEK with ZnO [143]. The PEEK particles with the dissolution method are smaller, while those in the direct mixing method are larger. The von Mises stresses are concentrated at the interface between ZnO and PEEK at the grain boundaries of ZnO, and the stress increases with increasing PEEK particle size, from several hundred MPa to over GPa. The breakdown electric field of the ZnO–PEEK composite prepared by the dissolution method is much higher than that of samples prepared by the direct mixing method. Moreover, Si *et al.* [161] incorporated PEEK and metal oxides (Bi₂O₃, MnO₂, Co₂O₃, and Cr₂O₃) that can be incorporated into ZnO ceramics through the cold sintering process, engineering the grain boundary structure and enhancing the electrical properties. The composite varistors achieved a breakdown electric field of up to 13 kV/mm and a maximum nonlinear coefficient exceeding 300.

Apart from organics and metal oxides, Guo *et al.* [216] adopted CSP to combine C₆₀ with ZnO. The ZnO–C₆₀ composites boasted a relative density above 95%, with 10 nm thin C₆₀ layers observed within the cold-sintered composites. C₆₀ modified the interfacial structure of ZnO ceramics, significantly enhancing their electrical properties, with the nonlinear coefficient rising from 2.3 to 8.5 and the breakdown electric field increasing from 80 to 2100 V/mm. Finite element analysis shows that the current density at the ZnO–C₆₀ interface surpasses that in other regions, and the current density in the aggregated region is higher than that in the nonaggregated region. This research demonstrates that CSP is a feasible route to tailor the interfacial properties of functional materials via the integration of C₆₀ and ceramics. In addition, ultrastiff nanodiamond particles can be incorporated into the ZnO matrix using the cold sintering process at 300 °C with acetic acid as the transient liquid phase [217]. The piezoelectric polarization induced by a large internal stress decreases the double Schottky barrier, which ultimately forms a ZnO-based varistor with a low switch voltage. The internal stress also enhances the strength of the ZnO-based composite by more than 230% compared with the monolith.

In addition to varistor characteristics, the thermal properties of ZnO may be manipulated with polyacrylonitrile (PAN) grain boundaries [218]. Using the cold sintering process with acetic acid as the transient liquid phase, ZnO–PAN composites show a 12.5-fold increase in the threshold electric field and the lowest recorded thermal conductivity of 0.72 W/(m·K) at room temperature, indicating that the ZnO–PAN composite is promising for the application of surge protection material with integrated thermal insulation.

Tai *et al.* [219] prepared a (1–*x*)ZnO–*x*polystyrene (PS) composite material with a high threshold electric field by a cold sintering process at 220 °C for 50 min using lightweight PS to adjust the grain boundary structures of ZnO varistors. As shown in Fig. 36, PS thin layers with a thickness of 2–10 nm are distributed at the grain boundaries of ZnO, forming the Schottky barrier, which triggers nonlinear current–voltage responses. In addition, the high insulation of PS enhances the breakdown electric field of composites, which is beneficial for the miniaturization development of varistor devices.

(5) Battery materials

The all-solid-state battery is an important next-generation battery, as it can increase safety and potentially allow for higher

power and energy density over a broader operating voltage and temperature range [220–222]. However, the densification temperature of solid electrolytes and cathode materials prepared through conventional sintering methods is typically high, especially for oxide-based materials. Field-assisted sintering technology (FAST), SPS, and CSP have been used to reduce sintering temperature and shorten sintering time [133,223]. Among these methods, the cold sintering process is a sustainable and energy-efficient sintering technology that represents a paradigm shift for the processing of solid electrolytes and cathodes. A wide variety of lithium- and sodium-based solid-state batteries with enhanced ionic conductivity and relative density have been developed, demonstrating the potential of CSP for applications in next-generation batteries [224]. To date, cold-sintered lithium-ion systems have good conductivity and volumetric capacity, where the performances of cold-sintered NASICON need to be further improved, as shown in Table 8.

(i) Li-ion materials

Lithium-ion batteries have been widely applied in various fields, ranging from portable electronic devices to electric vehicles, owing to their high energy density and long cycle life [225–228]. Guo *et al.* [148] utilized DI water as the transient liquid phase to cold sinter Li_{1.5}Al_{0.5}Ge_{1.5}(PO₄)₃ powders with PVDF–HFP at 120 °C under 400 MPa for 1 h, resulting in samples with a relative density of 80%–88%, higher than that obtained through hot-press sintering. CSP can co-fire Li_{1.5}Al_{0.5}Ge_{1.5}(PO₄)₃ and polymers, mitigating the adverse effects of composite material properties caused by poor physical contact and polymer decomposition. Seo *et al.* [229] found that the cold sintering process with LiOH solution as the transient liquid phase at 240 °C could effectively densify pure LiFePO₄ active material as well as cathode composites mixed with conductive carbon and PVDF. CSP yielded LiFePO₄ ceramics and LiFePO₄–PVDF cathode composites with relative densities exceeding 85%. It is noteworthy that the LiFePO₄–PVDF cathode composite exhibited a volumetric capacity of 340 (mA·h)/cm³ at 0.03 C to 0.1 C (where C represents the battery charge–discharge rate).

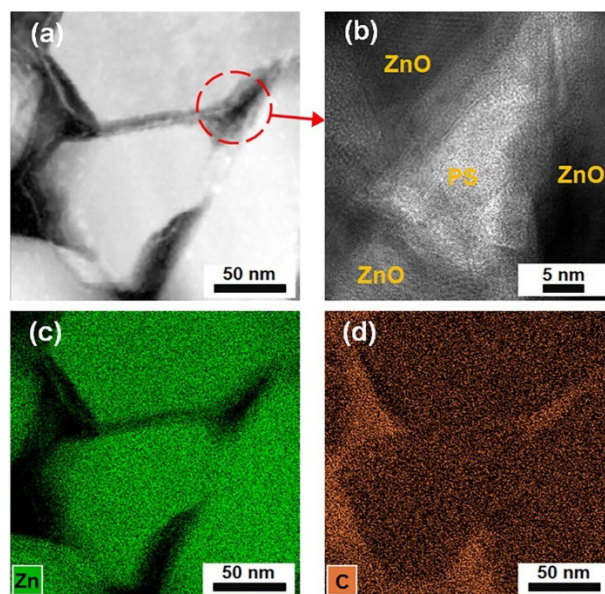


Fig. 36 TEM micrographs and EDS mappings of cold-sintered 93ZnO–7PS composite. (a) High-angle annular dark-field scanning TEM (HAADF-STEM) and (b) HRTEM images of grain boundaries. EDS mappings of (c) Zn and (d) C elemental distributions. Reproduced with permission from Ref. [219], © American Chemical Society 2024.

Table 8 Comparisons of cold-sintered materials in battery applications

Materials	Cold sintering transient liquid phase	Key parameters compared with those prepared by TS	Performances compared with those prepared by TS
Lithium-ion battery	DI water, LiOH solution or dimethylformamide (Annealing may or may be not required)	Lower ST and shorter T	Comparable or higher conductivity
NASICON	NaOH or KOH solution (Annealing may or may be not required)	Lower ST and shorter T	Relatively Lower conductivity

$\text{Li}_4\text{Ti}_5\text{O}_{12}$ enhances anode safety and is regarded as one of the most promising anode materials to replace conventional graphite anodes. The $\text{Li}_4\text{Ti}_5\text{O}_{12}$ /CNF composite anode achieved a relative density of 87% using CSP under 500 MPa at 120 °C with DI water as the transient liquid phase [230]. This cold-sintered anode demonstrated a high capacity of ~ 380 (mA·h)/ cm^3 at low current densities and maintained excellent cycle stability with 225 (mA·h)/g after 150 cycles at 0.2 C, which suggests that CSP is an effective means of enhancing the volumetric capacity of lithium-ion batteries.

Garnet-type $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$ stands as one of the most compelling highly conductive ceramic materials for all-solid-state lithium batteries. However, conventional sintering of $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$ materials necessitates high temperatures and lengthy sintering times and exhibits sensitivity to air and moisture, posing formidable challenges for practical applications. $\text{Li}_{7-2x+y}\text{Mg}_x\text{La}_{3-y}\text{Sr}_y\text{Zr}_2\text{O}_{12}$ can be densified through CSP at 120 °C using dimethylformamide as the transient liquid phase with a conductivity of 10^{-4} S/cm [231]. In addition, cold-sintered composite electrolytes have high conductivity over a wide temperature range and have good microstructure and grain boundary interfaces. Based on this, high-capacity cold-sintered composite all-solid-state lithium batteries were fabricated, achieving a capacity of approximately 140 (mA·h)/g.

$\text{Li}_{1.3}\text{Al}_{0.3}\text{Ti}_{1.7}(\text{PO}_4)_3$, with its NASICON (sodium superionic conductor) structure, has emerged as a promising solid-state electrolyte due to its high ionic conductivity (ranging from 10^{-4} to 10^{-3} S/cm at room temperature), exceptional stability against air and moisture, and low manufacturing costs. However, it is challenging to densify $\text{Li}_{1.3}\text{Al}_{0.3}\text{Ti}_{1.7}(\text{PO}_4)_3$ ceramics at reduced sintering temperatures while preserving their electrical properties. Cai *et al.* [232] employed CSP to predensify $\text{Li}_{1.3}\text{Al}_{0.3}\text{Ti}_{1.7}(\text{PO}_4)_3$ at 250 °C. Under optimal CSP conditions (DI water, 250 °C, 250 MPa, 1 h), the prepared samples exhibited a conductivity of 2.01×10^{-6} S/cm and a relative density of 83.0%. The optimal conductivity can be achieved from the annealed material (900 °C, 1 h) with 4.29×10^{-4} S/cm, surpassing the conductivity of conventionally sintered $\text{Li}_{1.3}\text{Al}_{0.3}\text{Ti}_{1.7}(\text{PO}_4)_3$ (8.51×10^{-5} S/cm). This underscores that the cold sintering-assisted process can produce highly conductive $\text{Li}_{1.3}\text{Al}_{0.3}\text{Ti}_{1.7}(\text{PO}_4)_3$ ceramics at temperatures lower than those of conventional sintering methods.

(ii) Other battery materials

The market witnesses relatively high prices for lithium, whereas metallic sodium boasts advantages of cost-effectiveness and abundant availability. Considering cost factors, sodium ion conductors can be considered alternatives to existing lithium-ion conductors. Scandium-substituted $\text{Na}_{3.4}\text{Sc}_{0.4}\text{Zr}_{1.6}\text{Si}_2\text{PO}_{12}$ (NASICON) is a promising electrolyte material for sodium-ion solid-state batteries, and it is the NASICON material with the highest ionic conductivity reported to date. Low-temperature densification and microstructural control are important factors in achieving low-cost manufacturing. Silva *et al.* [233] utilized CSP, with a KOH aqueous solution as a transient liquid phase, to prepare NASICON samples with a relative density greater than 85% at 250 °C. Further heat treatment at temperatures lower than

conventional sintering temperatures augmented the relative density and ionic conductivity of NASICON. A comparison of energy consumption between CSP and conventional sintering processes reveals that CSP consumed 0.28 kW·h, which is significantly lower than that consumed by conventional sintering (4.70 kW·h), underscoring its potential to drastically reduce sintering energy consumption.

Grady *et al.* [234] prepared $\text{Na}_3\text{V}_2(\text{PO}_4)_3$ samples with a relative density of $89\% \pm 2\%$ and a room temperature conductivity of 3.98×10^{-8} S/cm through CSP (10 wt% NaOH, 375 °C, 360 MPa, 3 h). The same CSP was extended to biphasic composites of $\text{Na}_3\text{V}_2(\text{PO}_4)_3$ with nanocarbon fibers (CNFs) or $\text{Na}_3\text{V}_2(\text{PO}_4)_3$ with NASICON solid electrolyte ($\text{Na}_3\text{Zr}_2\text{Si}_2\text{PO}_{12}$). These composites show high densities (90%) and significantly enhanced bulk conductivities. The composite containing 60 wt% $\text{Na}_3\text{Zr}_2\text{Si}_2\text{PO}_{12}$ had a total conductivity of 5.81×10^{-6} S/cm, while the composite with 5 wt% CNF achieved a total conductivity of 1.31×10^{-5} S/cm. The increases in conductivity demonstrate the ability to enhance the conductive properties of electrodes through simple compositional variation via low-temperature densification.

(6) TE materials

TE materials, capable of converting waste heat into electricity, offer avenues for mitigating the energy crisis. They have attracted significant attention owing to their widespread applications in thermoelectric refrigeration, waste heat recovery, aerospace, and various other fields [235]. The preparation of thermoelectric materials typically involves hot pressing (HP) and spark plasma sintering (SPS) methods [236,237]. These technologies are high-temperature processes that may deteriorate the micro/nanostructures of materials, leading to chemical uniformity issues. The aforementioned challenges can be addressed through the adoption of low-temperature processing in the manufacturing of thermoelectric materials. It is demonstrated that a number of thermoelectric materials with better figure of merit (ZT) can be densified at lower sintering temperatures and shorter times using the cold sintering process, as shown in Table 9, revealing that CSP is promising for the development of thermoelectric materials.

Guo *et al.* [155] integrated the 2D material $\text{Ti}_3\text{C}_2\text{T}_x$ into ZnO ceramics by a cold sintering process at 300 °C (Fig. 46). With the incorporation of up to 5 wt% $\text{Ti}_3\text{C}_2\text{T}_x$, the electrical conductivity increased by 1–2 orders of magnitude, leading to a significant improvement in the overall power factor. The 99ZnO–1 $\text{Ti}_3\text{C}_2\text{T}_x$ nanocomposite demonstrated a metallic conductivity of 312 S/cm, a Seebeck coefficient of -120 $\mu\text{V/K}$, and a power factor of approximately 4.5×10^{-4} W/mK² at 750 °C, indicating that the CSP could serve as an effective method for fabricating functional nanocomposites with the addition of 2D materials.

CSP was employed by dos Santos *et al.* [238] to fabricate $\text{Ca}_3\text{Co}_4\text{O}_9$ ceramics at a low temperature of approximately 150 °C. The cold-sintered $\text{Ca}_3\text{Co}_4\text{O}_9$ ceramics exhibited a higher relative density than conventionally sintered ceramics (approximately 58%), achieving 78.2% of the theoretical density. The grain growth of CSP samples is limited, which reduces the thermal conductivity. Compared to conventionally sintered ceramics, the cold-sintered ceramics exhibited greater electrical conductivity (6459 S/m) and

Table 9 Comparisons of cold-sintered materials in thermoelectric applications

Material	Cold sintering transient liquid phase	Key parameters compared with those prepared by TS	Performance
ZnO–MXene	Acetic acid (annealing may or may be not required)	TS is not available	Good electrical conductivity and mechanical properties
Ca ₃ Co ₄ O ₉	Acetic acid (annealing at higher temperatures is required)	Lower ST and shorter <i>T</i>	Higher electrical conductivity and smaller thermal conductivity than those prepared by TS
Bi ₂ Te ₃ -based compounds	DI water (annealing may be not required)	Lower ST and shorter <i>T</i>	Ultralow thermal conductivity and comparable ZT value, compared with commercial ingot
PbTe _{0.94} Se _{0.06}	Solvothermal precursor (annealing is not required)	Lower ST and shorter <i>T</i>	ZT is better than that prepared by warm pressing and on par with that prepared by SPS
Cu ₂ Se	Thiol-amine solution (annealing may or may be not required)	Lower ST and shorter <i>T</i>	With reduced thermal conductivity, ZT is better than that prepared by hot-press
Ag ₂ Se	Thiol amine or aqua regia (annealing may be not required)	Lower ST and shorter <i>T</i>	Low thermal conductivity and high ZT value

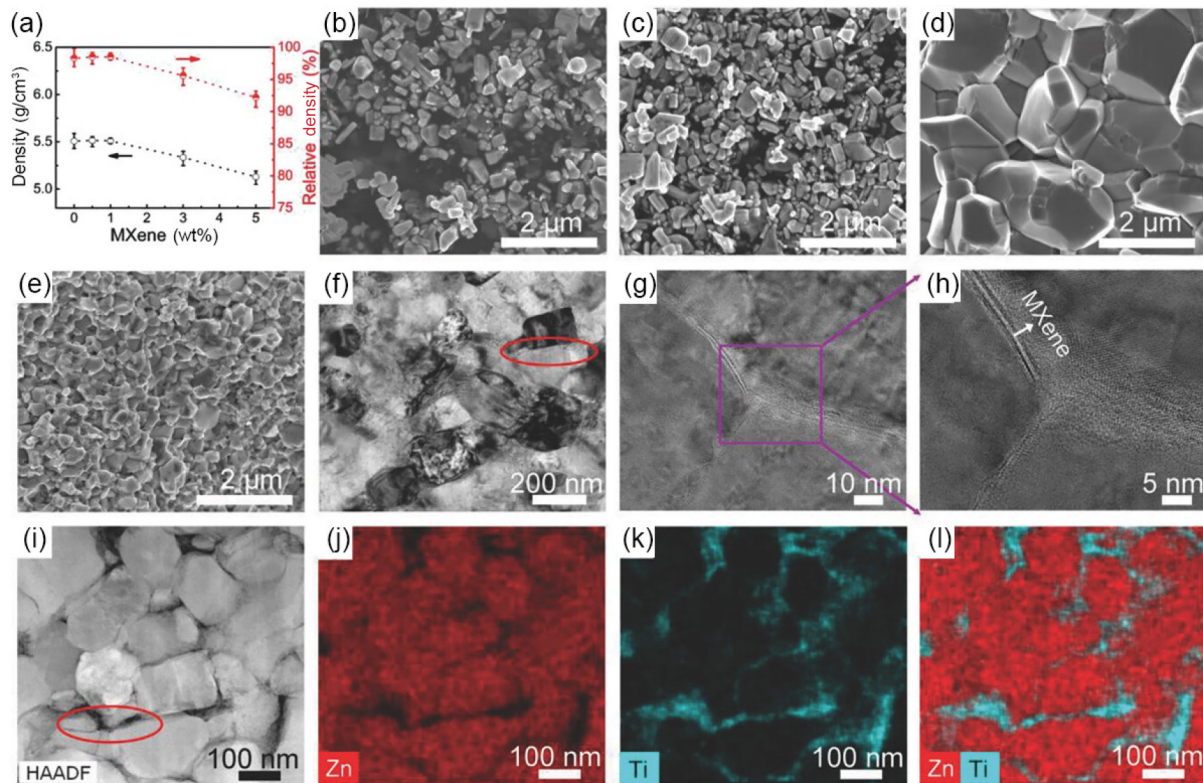


Fig. 37 Density and microstructure of ZnO–Ti₃C₂T_x nanocomposites cold sintered at 300 °C for 1 h [155]. (a) Densities of cold sintered ZnO–Ti₃C₂T_x nanocomposites. SEM images of (b) ZnO and (c) 99ZnO–1Ti₃C₂T_x raw powders and the cross sections of cold sintered (d) ZnO and (e) 99ZnO–1Ti₃C₂T_x ceramics. (f–h) TEM, (i) HAADF-STEM images, and (j–l) energy dispersive spectroscopy (EDS) elemental mapping of cold sintered 99ZnO–1Ti₃C₂T_x nanocomposite. The red circles in the TEM and HAADF-STEM images show one example of the Ti₃C₂T_x region. In the HAADF image, the bright areas belong to ZnO, and the dark areas belong to Ti₃C₂T_x. EDS maps, where elemental Zn is shown in red and Ti is shown in cyan, show the presence of Ti₃C₂T_x at the ZnO grain boundaries. Reproduced with permission from Ref. [155], © WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim 2018.

reduced thermal conductivity (0.672 W/(m K)) at 600 °C. The highest figure of merit value obtained was 0.12 for the cold-sintered ceramics, which is three times that of conventionally sintered ceramics.

Bi₂Te₃-based compounds currently stand as the most commercially relevant thermoelectric materials at temperatures close to room temperature. Zhu *et al.* [239] prepared p-type Bi_{0.5}Sb_{1.5}Te₃ bulk materials using CSP at 110–150 °C. Facilitated by the liquid phase of deionized water and pressure, the Bi_{0.5}Sb_{1.5}Te₃ sample approaches a state of complete densification, exceeding 97%. Furthermore, the CSP can result in a decrease in grain size and an increase in twin boundaries. Phonon scattering diminishes the thermal conductivity of the lattice, ultimately yielding an

ultralow lattice thermal conductivity of 0.57 W/(m·K) at 380 K. A ZT value of up to 0.56 at 450 K can be attained, competitive with that of a commercial Bi_{0.5}Sb_{1.5}Te₃ ingot (ZT = 0.8–1.0).

Lu *et al.* [240] achieved densification of PbTe_{0.94}Se_{0.06} material at 300 °C using CSP with a solvothermal precursor as the transient liquid phase, including NaOH, DI water, Te powder, PbCl₂, ethylene diamine tetraacetic acid disodium salt, ethylene glycol, Se powder, and NaBH₄. The conductivity of cold-sintered PbTe_{0.94}Se_{0.06} is notably higher than that of samples prepared using SPS and WP (warm pressing) methods. Additionally, the bipolar effect and lattice thermal conductivity due to enhanced scattering are effectively suppressed, as the dissolution–precipitation process significantly inhibits the coarsening of the precipitated phase. The

final figure-of-merit values are 56% and 140% higher than those of the densified samples by the SPS and WP methods, respectively.

Cu_2Se , a superionic conductor aligning with the “phonon-liquid electron-crystal” concept, is recognized as a p-type thermoelectric material with exceptional TE properties at elevated temperatures. However, a notable drawback of Cu_2Se materials is their limited high-temperature stability, which often leads to compositional alterations and property deterioration. Low-temperature sintering techniques are thus sought to address these issues. Recently, Piyasin *et al.* [241] used CSP to prepare Cu_2Se bulk with a relative density approaching 90% by introducing a thiol-amine solution as a transient liquid phase at sintering temperatures as low as 473 K. This CSP approach not only preserves the morphology of the Cu_2Se precursor powders but also effectively mitigates grain growth. As a result, the lattice thermal conductivity of the CSP samples is significantly reduced, owing to the augmented grain boundary phonon scattering. This reduction leads to a considerable enhancement in the figure-of-merit, which increases from 0.65 at 800 K in the hot-pressed sample to 2.13 at 800 K in the CSP sample.

Ag_2Se is a promising material for thermoelectric applications near room temperature. Palaporn *et al.* [242] proposed a modified cold sintering process (mCSP) for preparing bulk Ag_2Se to enhance its ZT. Unlike conventional CSP, mCSP uses thiol amine (TA) as the liquid medium, which does not dissolve Ag_2Se , resulting in the free flow of TA solution in the interparticle channels. By sintering at a temperature higher than the boiling point of TA, the liquid evaporates, leaving behind large chunks of Ag_2Se material with a tubular porous structure. These induced defects have a significant impact on the thermoelectric properties of Ag_2Se , resulting in a notable increase in the Seebeck coefficient and a decrease in conductivity. The overall power factor (PF) of the mCSP sample exceeded that of the control sample without TA. In addition, the micropores formed during the mCSP process effectively suppressed thermal conductivity, resulting in an optimized ZT value of over 1.0 for the mCSP sample at 400 K. The average ZT in the range of 300–400 K is approximately 1.0, which is one of the highest reported values. Further optimization of the mCSP parameters can make Ag_2Se more competitive with state-of-the-art materials near room temperature.

(7) Optical materials

In addition to its widespread applications in various material systems mentioned above, the cold sintering process has also emerged in the realm of optical materials. Cold-sintered fluorides show comparable in-line transmittance, while the performances of cold-sintered zeolite need to be further enhanced, as shown in Table 10.

Gao *et al.* [243] utilized highly reactive raw powders derived from hexagonal faujasite (EMT)-type zeolites, with NaOH as a transient liquid phase, to fabricate translucent glasses with high relative density via the cold sintering process. The chemical reaction between NaOH and zeolite-derived powders facilitated the dissolution-precipitation process during sintering, characterized by an extremely low activation energy of 27.13 kJ/mol. Furthermore, it was noted that both excessively high

and low concentrations of NaOH solution led to a decrease in light transmittance, highlighting the crucial role of the transient liquid phase in the CSP. The glass prepared through CSP exhibited high hardness, reaching 4.3 GPa, which is sufficient for common optical applications.

CaF_2 , renowned for its exceptional transmittance, broad transmittance range, high chemical resistivity, and low phonon energy, has found extensive utilization in infrared windows, lenses, and prisms. A dense CaF_2 nanoceramic with visible transmittance up to 75% can be prepared using a cold sintering process at 350 °C [244]. The in-line transmittance of cold sintered CaF_2 with $\approx 50\%$ – 80% in the wavelength range of 400–800 nm is comparable to that prepared by conventional hot pressing at 800 °C.

There are also a number of cold sintered materials for other applications, such as bioceramics for medical applications, ceramic targets, magnetic materials, refractory ceramics, and flame-retardant materials [245–247]. It is anticipated that with further study of the sintering mechanism and a comprehensive understanding of process–microstructure–property relationships, an increasing number of materials with better performances will be developed using the cold sintering process in the future.

2.2.4 Challenges and prospects of the cold sintering process

In summary, the emergence of the cold sintering process has significantly propelled the development of low-temperature sintering technology, offering a new route for energy conservation and environmental protection. This breakthrough of ultralow-temperature sintering with the cold sintering process makes it feasible for the design of novel material systems. Ceramics can be co-fired with organics, inorganics, metals, fibers, and two-dimensional materials, thereby enhancing the electrical, thermal, magnetic, optical, and mechanical properties of ceramics. Furthermore, the utilization of lower processing temperatures mitigates the volatilization of elements during sintering and facilitates the design of grain sizes and interfacial structures. However, the cold sintering process is in the early stage, which needs further investigation:

(1) Sintering mechanism of CSP. The sintering mechanism is still incomplete and requires further exploration. Future work may combine experimental results with simulations to explain the thermodynamics and kinetics of CSP, as shown in Fig. 38. Molecular dynamics, the finite element method, field phase simulation, and other mathematical approaches are promising methods to study the sintering mechanism. In addition, *in situ* experiments may also be employed to further understand the detailed process of CSP.

(2) Comprehensive understanding of process-microstructure-property relationships. CSP is an ultralow-temperature sintering technique, making it promising for the manipulation of grain sizes and grain boundary structures, which further tailors the performances of ceramics. Once an amorphous phase is formed at grain boundaries during the cold sintering process under specific processing conditions, which affects the physical and chemical properties of ceramics, its thickness and composition should be

Table 10 Comparisons of cold-sintered materials in optical applications

Material	Cold sintering transient liquid phase	Key parameters compared with those prepared by TS	Performance
Zeolite	NaOH solution (Annealing may be not required)	Lower ST and shorter T	In-line transmittance and mechanical properties are lower than that prepared by SPS
Fluoride	HCl or LiOH solution (Annealing may be not required)	Lower ST and shorter T	In-line transmittance are similar to that prepared by TS

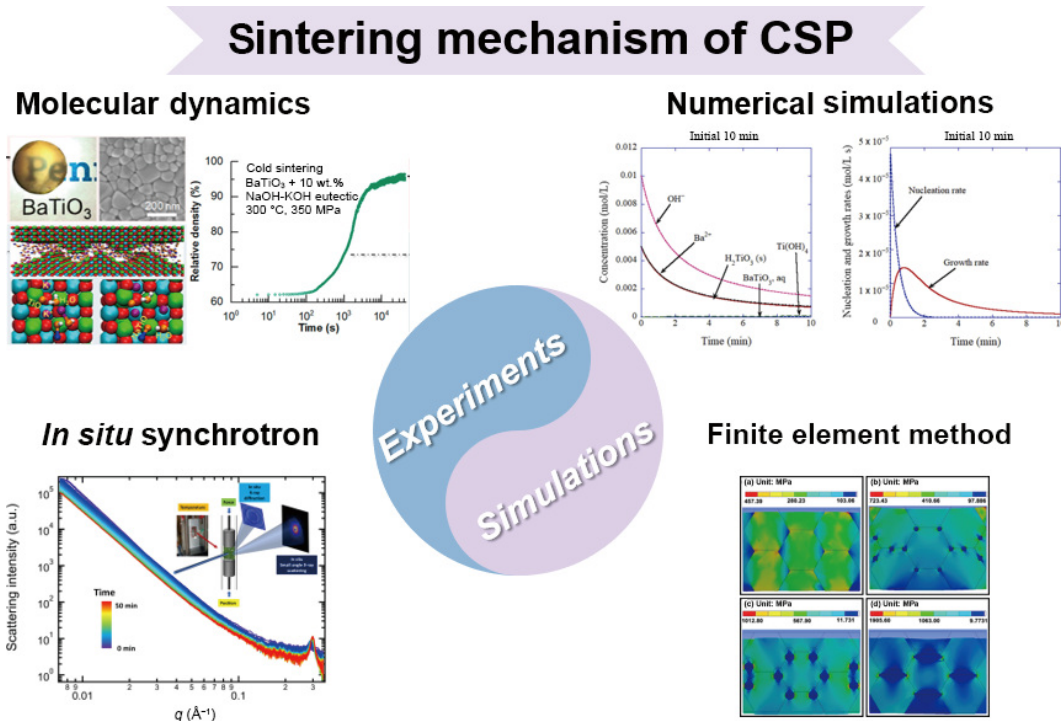


Fig. 38 Sintering mechanism based on combinations of experiments and simulations. Reproduced with permission from Ref. [143], © Acta Materialia Inc. 2018; from Ref. [508], © American Chemical Society 2021; from Ref. [509], © Elsevier Ltd. on behalf of Acta Materialia Inc. 2023; from Ref. [510], © Elsevier Ltd. 2023.

optimized based on the understanding of the process–microstructure–property relationship. Elaborated experimental skills and a strong theoretical background are required for better insight. The chemical stability of the grain boundaries after CSP is also important for functional applications, which needs further study in the future.

(3) Unstable and composite material designs. Since the proposal of the cold sintering process in 2016, it has been widely utilized to densify unstable materials and integrate ceramics with other materials with low melting points, e.g., ceramic–ceramic, ceramic–polymer, ceramic–metal, ceramic–glass, ceramic–2D material, and ceramic–fiber, due to the ultralow temperature of CSP, as shown in Fig. 39. A number of thermally unstable materials, such as aluminum hydroxide [248], SnO [249], and ZrW_2O_8 [250], have been successfully developed based on the cold sintering process. New unstable or composite designs are essential to take full advantage of the cold sintering process, and promoting the applications of CSP in the production of new materials and devices will be the focus of future research.

2.3 High-pressure and ultra-high-pressure sintering

2.3.1 Introduction to high pressure and ultrahigh pressure

As an important component of engineering materials, advanced ceramics have been widely used in many fields, including machinery, aerospace, nuclear energy, biomedicine, and modern communication industries, and they are playing an increasingly important role in the national economy and national defense construction. In conventional sintering technologies, including atmosphere sintering, vacuum sintering, hot pressing, and hot isostatic pressing, fully dense ceramics are mainly obtained by atomic diffusion, eliminating interparticle pores at high temperatures and long soaking times. However, atomic diffusion may cause unavoidable grain growth, which will decrease the mechanical properties of materials. In addition, long holding time for hours or even days is an extremely great consumption, which

is unfavorable to industrial production. Advanced ceramic materials, especially for boride and carbide ceramics, mostly have strong covalent bonds and low atomic self-diffusion coefficients, which make their sintering process difficult.

Relative density and grain size are two important indicators for structural ceramic materials because they have a direct impact on their mechanical properties. To obtain high-performance ceramics, high-pressure sintering has become a promising method that can simultaneously increase density and restrain grain growth. Applying high pressure can reduce the distance between particles and strengthen the contact between individual particles, resulting in plastic deformation. In recent years, high-pressure sintering has been regarded as a promising method to obtain high-performance ceramics, and plastic deformation has been proven to be the dominant mechanism in high-pressure sintering.

As a novel fast sintering technology, SPS achieves rapid heating and cooling of the samples by utilizing pulsed current passing through molds and samples. Consequently, it could effectively restrict grain growth during the heating process, retaining the required high-temperature structures and phase compositions, thereby realizing flexible regulation of the microstructure of materials. The heating rate is within the range of 100–3000 °C/min, and the holding time usually does not exceed 10 min. Compared with conventional sintering technologies, SPS exhibits higher thermal efficiency. A series of new rapid sintering technologies (SPS plus) based on SPS offer numerous possibilities for the fabrication of advanced ceramics. Ultra-high pressure sintering (UHPS) technology is considered one of the most promising and efficient techniques for fabricating nanosized ceramics. Compared to conventional high-temperature sintering methods, UHPS technology showed two primary advantages. First, it could achieve full density at relatively low temperatures while simultaneously inhibiting rapid grain boundary migration and preventing the occurrence of intragranular pores, thus restraining grain growth. Second, new grain boundaries and subgrains were effectively created by applying ultra-high pressure,

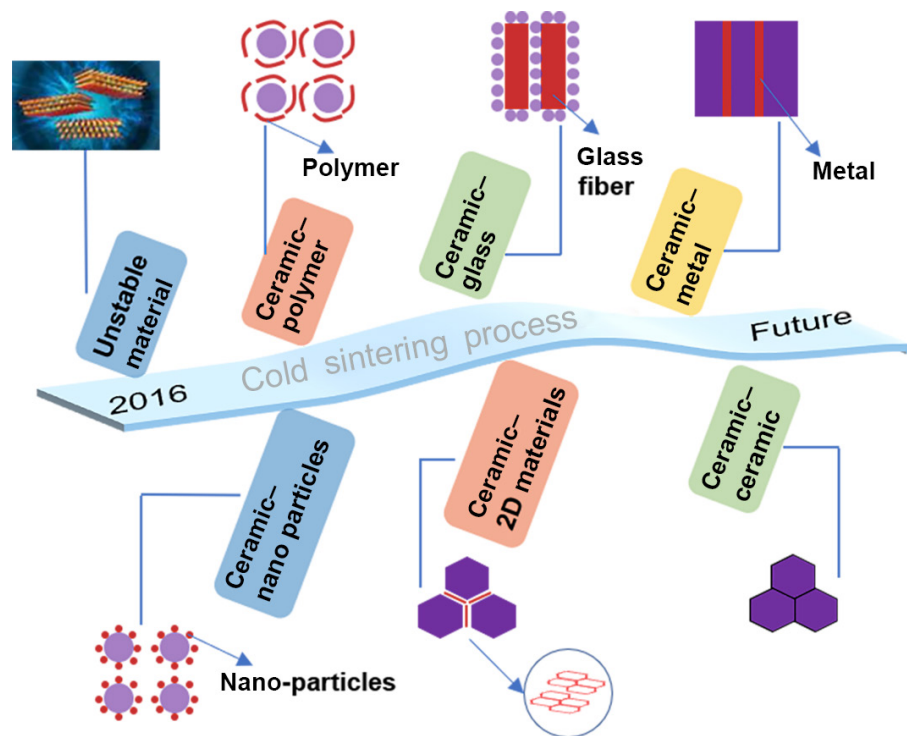


Fig. 39 Unstable materials and composite designs via the cold sintering process.

thereby leading to grain refinement. The unique microstructure may contribute to enhanced mechanical properties, oxidation resistance, and other properties. Consequently, the strategy of improving pressure and reducing sintering temperature is beneficial to fabricated nanocrystalline ceramic materials without intragranular pores with exceptional properties.

2.3.2 Ceramic material systems

(1) Difficult-to-sinter ceramics

Carbide ceramics have been widely applied in engineering fields because of their excellent properties, such as high hardness, excellent high-temperature resistance, and good stability. However, it is difficult to achieve high density owing to the low self-diffusion coefficient and strong covalent bonds [251–254]. Pressureless sintering of pure boron carbide is extremely difficult, and only 93% of the density can be obtained at a high temperature of 2375 °C [255]. The addition of additives is helpful to reduce the temperature of B_4C for pressureless sintering, but it still requires sintering temperatures of 2075–2250 °C [256,257]. Ji *et al.* [258] obtained fully dense B_4C ceramics without grain growth at lower temperatures with SPS. The dense boron carbide ceramics possessed excellent mechanical properties, including a Vickers hardness of 37.8 GPa, flexural strength of 445.3 MPa, and fracture toughness of 4.7 $MPa \cdot m^{1/2}$. Compared with the conventional pressureless sintering method, the sintering temperature could be reduced nearly 400 °C by SPS, and the sintering time was also dramatically shortened. Such a rapid sintering method with low energy consumption also improves the cost-effectiveness of other advanced ceramics for practical applications.

It is difficult to densify WC by conventional sintering methods. Adding additives is an efficient way to reduce the sintering temperature but weakens the properties of materials, including hardness, wear resistance, and oxidation resistance [259–261]. The mechanical properties of materials could be enhanced by using nanosized powder as the original powder, but the surface energy of nanopowder could lead to abnormal grain growth [262].

Therefore, binderless WC with higher hardness, wear resistance, and better high-temperature oxidation resistance is urgently needed. Investigations of binderless WC have focused on SPS from nanopowder in recent years. Zhao *et al.* [263] fabricated fully dense WC without obvious grain growth at 1550 °C and 200 MPa and reported that high pressure reduced the sintering temperature and facilitated the densification process. The as-sintered sample showed ultra-high hardness and excellent tribological properties.

(2) Ultrahigh temperature ceramics

Many ultrahigh temperature materials are difficult to densify through conventional methods due to low diffusion coefficients and strong covalent bonding. Generally, the sintering temperature should be raised to 2200 °C, and the soaking time is as long as 20–60 min [251,264,265]. A high sintering temperature and long soaking time will inevitably lead to severe grain growth, thereby weakening the mechanical properties of materials. Wang *et al.* [266] used HP to sinter ZrC with MC (M = V, Nb, and Ta) additions. The relative density of ZrC sintered without MC reached only 80% and 92% under a pressure of 30 MPa at 1900 and 2000 °C, respectively. Additives could facilitate the densification process; the relative density of ZrC sintered together with MC reached 98% under conditions of 1900 °C and 30 MPa, but the as-sintered sample exhibited obvious grain growth, from 1–3 μm to 8.20 μm . Ke *et al.* [267] used SPS to sinter zirconium carbide (ZrC) without any additives; the relative density of the sample could reach 98%, and there was no obvious grain growth under the conditions of 1900 °C and 200 MPa. Subgrains and high-density dislocations formed in the ceramics sintered under high pressures. The hardness and fracture toughness of the ZrC ceramics reached 20.53 GPa and 2.70 $MPa \cdot m^{1/2}$, respectively. Zheng *et al.* [268] studied the densification behaviors, grain growth, and mechanical properties of tantalum carbide (TaC) ceramics obtained by SPS at different temperatures and pressures. The results showed that nearly fully dense (98.6%) TaC ceramics with fine grains (1.42 μm) could be obtained at 1700 °C under 200 MPa, and the as-sintered specimen possessed a high dislocation density of

$1.9 \times 10^{14} \text{ m}^{-2}$, which contributed to its high hardness of 16.26 GPa. Zhu *et al.* [269] obtained fully dense $\text{Hf}_{0.95}\text{Ta}_{0.05}\text{B}_2$ ceramics with ultrafine grains by SPS, and its average grain size was only 0.6 μm . The fully dense ceramics displayed superior properties, including a high Vickers hardness of 24.8 GPa and a fracture toughness of 4.2 $\text{MPa}\cdot\text{m}^{1/2}$.

(3) Nanocrystalline ceramics

Nanocrystalline materials possess extreme physical and mechanical properties, including enhanced superplasticity and ductility, superior strength, and optical transparency in usually opaque ceramics [270]. Polycrystalline ceramics with higher performance could be achieved by refining the grain size and increasing the density. However, conventional sintering technologies inevitably lead to grain growth due to long soaking time and high sintering temperatures [271]. Chen and Wang [12] fabricated fully dense cubic nanocrystalline Y_2O_3 by a “two-step sintering method”. The sample was heated to a temperature of 1250–1350 °C to achieve an intermediate density of nearly 80% and then cooled down and held at a lower temperature of 1150 °C until it was fully dense. Grain growth in the second stage was suppressed by exploiting the difference in kinetics between grain-boundary diffusion and grain-boundary migration. The finest grain size of fully dense Y_2O_3 is only 60 nm, but it has increased 4–6 times from the original powder. Lee *et al.* [272] obtained 5 mol% TiO_2 -excessive BaTiO_3 at a relatively lower temperature of 1290 °C, with long soaking time of 100 h; the sample showed high density and limited grain growth. However, this sintering method is not suitable for industrial applications because of its long soaking time.

Ji and coworkers [273] densified nanocrystalline 3 mol% yttria stabilized zirconia (3YSZ) ceramics without grain growth under conditions of 900 °C and 1.5 GPa. The Vickers hardness of the as-sintered sample was above 14 GPa and 12.5% higher than that of the 3YSZ sintered by conventional methods. Xu *et al.* [274] obtained fully dense nanocrystalline alumina ceramics with limited grain growth at relatively low temperatures by using the high-pressure sintering method. Two kinds of commercially pure $\alpha\text{-Al}_2\text{O}_3$ powder with average particle sizes of 220 nm and 3 μm were sintered at relatively low temperatures and under a high pressure of 200 MPa. The sintering densification temperatures and the starting threshold temperatures of grain growth were determined by the applied pressure, and the surface energy was related to grain size, as they both increased with grain size and decreased with applied pressure. Wu *et al.* [275] synthesized $(\text{La}_{0.2}\text{Nd}_{0.2}\text{Sm}_{0.2}\text{Eu}_{0.2}\text{Gd}_{0.2})_2\text{Zr}_2\text{O}_7$ raw powder by a solid-phase reaction. Then, fully dense and ultrafine nanocrystalline ceramics were fabricated under 10 GPa at a low temperature of 800 °C, and the average grain size of the sample was only 151 nm, which was 40% smaller than that of the raw powder (252 nm). Compared with the samples fabricated by the pressureless sintering method, the UHPS-ed sample exhibited higher hardness and better aqueous durability. In later research work, Lin and coworkers [276] synthesized nanocrystalline powder with an average particle size of 11 nm by the combustion method and then fabricated fully dense high-entropy nanocrystalline $(\text{La}_{0.2}\text{Nd}_{0.2}\text{Sm}_{0.2}\text{Eu}_{0.2}\text{Gd}_{0.2})_2\text{Zr}_2\text{O}_7$ ceramics under an ultra-high pressure of 10 GPa at a lower temperature of 600 °C; the hardness of the as-prepared specimen was 11.9 GPa.

2.3.3 Sintering mechanism

(1) SPS

High pressure-assisted sintering has been proven to be an efficient method to fabricate high-performance ceramics. Fu *et al.* [258] proposed a new technology principle of high-pressure, low-

temperature, and rapid sintering, which is dominated by quasiplastic deformation. First, the sintering temperature is controlled below the temperature that leads to grain growth (T_g) to inhibit grain boundary migration and grain growth. Second, the densification process is dominated by plastic deformation under conditions of high pressure and relatively high temperature. This perspective challenges the conventional sintering densification mechanism that is controlled by atomic diffusion. Investigators have used micro-sized B_4C as the research object, which is difficult to sinter but widespread and is in urgent demand for various industries. Figure 40(a) shows that there existed a temperature range “M” in T_d – T_g , and the optimal range might be 1675–1700 °C for the as-sintered micro-sized B_4C , in which fully dense ceramics with excellent mechanical properties could be obtained under a high pressure of 80–100 MPa. The morphologies of the original powder and polished and etched surfaces of the B_4C specimen sintered at 1700 °C under 80 MPa are shown in Figs. 40(b) and 40(c), respectively. The grain size of B_4C was 2.36 μm , and almost no grain growth occurred.

Researchers proposed and validated a two-stage model based on a rapid sintering process with quasiplastic deformation. The schematic sintering route by quasiplastic deformation as the dominant mechanism is displayed in Fig. 40(d). Based on the analysis of the experimental results and theoretical calculations, the whole efficient low-temperature and high-temperature sintering process could be divided into two stages. In the first stage, an ultra-high contact force emerged for ceramic particles due to the contact area being so small; then, quasiplastic deformation occurred in the ceramics and resulted in a relatively high densification degree (~80%). The contact force decreased with increasing contact area, and quasiplastic deformation occurred until the contact force was lower than the ultimate yield stress of the materials under high-temperature conditions. In the final stage, a high density can be obtained rapidly depending on grain boundary migration and the creep mechanism. Fully dense (99.7%) micro-sized B_4C with limited grain growth was fabricated at 1700 °C under 80 MPa in 5 min; the sample exhibited significant mechanical properties. Plastic deformation under high pressure and low temperature conditions was successfully proven to be the dominant mechanism during the densification process.

(2) UHPS

In recent years, UHPS technology has become well known for its efficient fabrication of high-performance ceramics with unique microstructures. Tian *et al.* [277] fabricated ultrahard nanotwinned cubic boron nitride (c-BN) from onion-like BN (o-BN) at 1800 °C under an ultra-high pressure of 15 GPa. The resulting c-BN bulks retained a striking combination of physical properties, including an ultra-high Vickers hardness of 100 GPa (the optimal hardness of synthetic diamond), a high oxidation temperature of 1294 °C, and a superior fracture toughness of above 12 $\text{MPa}\cdot\text{m}^{1/2}$ (beyond the fracture toughness of commercial cemented tungsten carbide, 10 $\text{MPa}\cdot\text{m}^{1/2}$). The same researcher group then synthesized nanotwinned diamond from onion carbon nanoparticles at 2000 °C and 20 GPa. The as-sintered sample exhibited unprecedented hardness and thermal stability, with a remarkably high hardness of 200 GPa and an in-air oxidation temperature more than 200 °C higher than that of natural diamond [278].

Xu and coworkers [279] fabricated fully dense ZrB_2 with grains refined under an ultra-high pressure of 15 GPa at a relatively lower temperature of 1450 °C. ZrB_2 obtained significant mechanical and oxidation-resistant properties, and its grain size decreased 56% from that of the raw powder. Compared with high-temperature sintered specimens at temperatures over 2000 °C, the

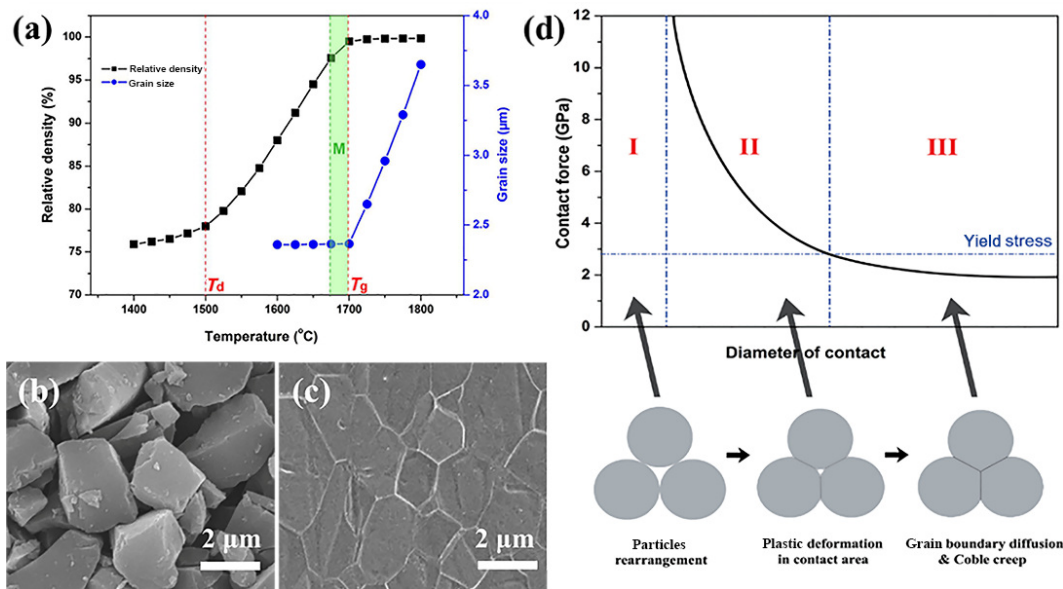


Fig. 40 (a) Relative density and grain size of B₄C ceramic as a function of temperature during sintering with quasiplastic deformation as the dominant densification mechanism; (b, c) SEM images of B₄C raw powder and dense B₄C ceramic sample, respectively; (d) schematic of the sintering route by quasiplastic deformation as the dominant densification mechanism. Reproduced with permission from Ref. [258], © The Author(s) 2015.

hardness and fracture toughness increased nearly 46% and 69%, respectively. The dislocation density was up to 3 orders of magnitude, and the grain size decreased by 96%. According to Hall–Petch and Taylor dislocation hardening and the work-hardening effect, the mechanical properties of materials could be enhanced by the refined grains, substructure, and high dislocation density that occur due to plastic deformation during the sintering process. According to the unique structure, its threshold oxidation temperature was approximately 250 °C higher than that of ZrB₂ sintered at high temperature, which had achieved one of the highest values (1100 °C) among the reported monolithic ultrahigh-temperature ceramics. A densification mechanism of dislocation multiplication and grain refining was proposed and demonstrated as the dominant mechanism during sintering, while both mechanical and oxidation resistance properties were enhanced simultaneously.

Figure 41 displays the EBSD analysis probe characterizations of ZrB₂ ceramics sintered at 1450 °C, forming a high density of low-angle grain boundaries because most grains are broken to create subgrains. These grain boundaries were not only distributed uniformly in the grains but also along the original high-angle grain boundaries. The as-sintered ceramic showed refined grains, and its average grain size was only 0.7 μm, which decreased 56% from the raw powder.

2.3.4 Excellent properties

(1) Hardness and wear resistance

Generally, carbide ceramics possess excellent physical, chemical, and mechanical properties, including high hardness, high melting point, high thermal conductivity, and excellent wear resistance. With the above advantages, they are widely applied in the manufacture of cutting tools, wear-resistant parts, and product components used in extreme environments. Ji *et al.* [258] fabricated fully dense and micro-sized B₄C without grain growth with a high hardness of 37.8 GPa by SPS technology. Zhao *et al.* [263] prepared high-density and limited-grain-growth tungsten carbide ceramics under a high pressure of 200 MPa at a relatively low temperature of 1550 °C without any additives. The average grain size of the WC ceramics was 210 nm, which was only 5%

larger than that of the raw powder (200 nm). The densification behavior, microstructure evolution, mechanical properties, and tribological properties of as-sintered nanocrystalline WC ceramics sintered under different pressures were studied. The experimental results illustrated that increasing pressure could promote the densification process, accelerate the densification rate, and hinder grain growth. Meanwhile, the sample sintered under high pressure obtained high-density dislocations, better mechanical properties, and better tribological properties. The sample sintered at 1550 °C under 200 MPa exhibited outstanding properties, such as an ultra-high hardness of 32.19 GPa and good wear resistance. Additionally, the hardness increased by 23%, the friction coefficient decreased by 30%, and the wear resistance improved by 63%.

(2) Lower-temperature plasticity

B₄C ceramics possess many outstanding features and properties, including high hardness (second only to diamond and cubic boron nitride), high melting point, low density (the lightest ceramic), high neutron absorption cross section, excellent chemical stability, and wear resistance. However, its intrinsic brittleness and relatively lower plasticity limit its application scope [280]. Xu *et al.* [281] fabricated boron carbide with lower-temperature plasticity under conditions of 1800 °C and 80 MPa. The plastic deformation temperature of the as-produced specimen decreases by 200 °C compared to the ceramics obtained by conventional high-temperature sintering. The excellent plasticity of fully dense B₄C is greatly improved by the boundary nonstoichiometry, which is induced by the applied high pressure. Grain boundary diffusion promotes grain boundary sliding, which dominates the lower-temperature plasticity. The mechanical properties of B₄C sintered under high-pressure low-temperature (HPLT) and low-pressure high-temperature (LPHT) conditions are studied. Figure 42 shows that the flexural strengths of both HPLT-B₄C and LPHT-B₄C first increased with increasing temperature prior to decreasing as plasticity began to occur. The plastic deformation of B₄C occurred at 1700 and 1900 °C. The flexural strength of HPLT-B₄C (320 MPa) is far higher than that of LPHT-B₄C (143 MPa), and they exhibit different fracture modes.

(3) Fracture toughness

In the past hundred years, the prestress strategy has been

utilized in glass and concrete to improve mechanical properties. However, this strategy is hard to apply in ceramic materials

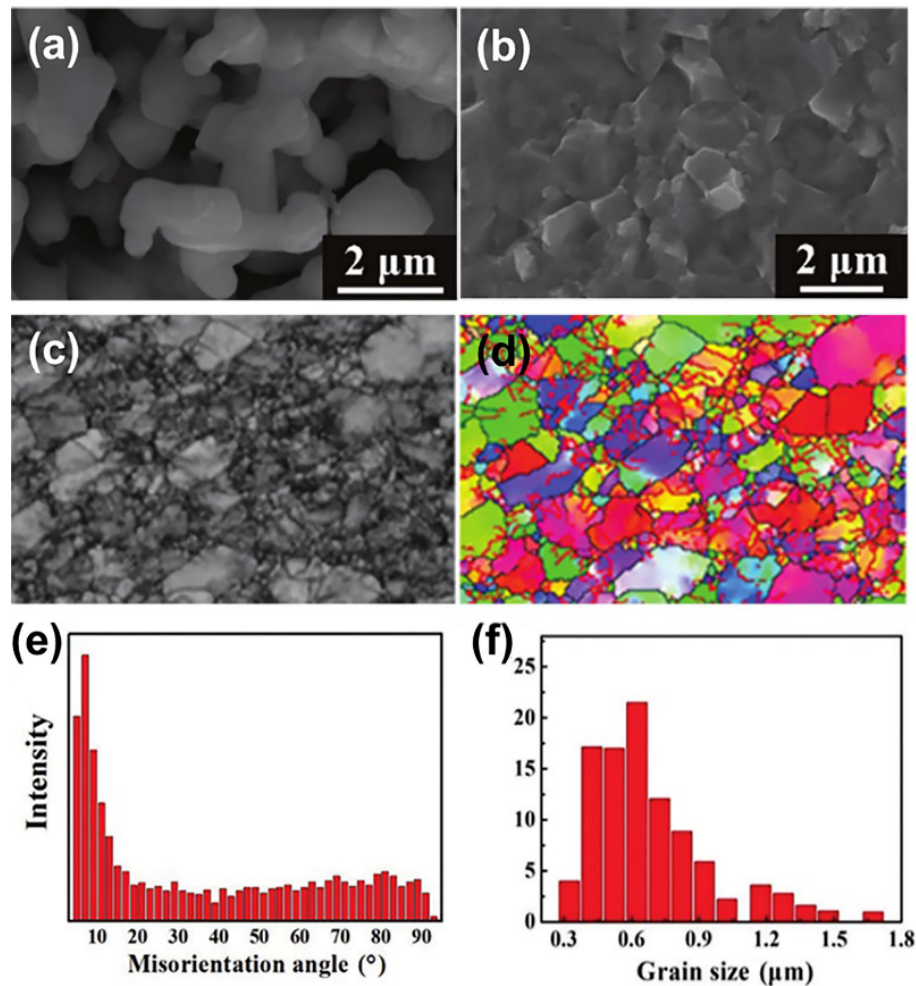


Fig. 41 SEM images of (a) raw powder and the fracture surface of UHPS-ed ZrB₂ under 15 GPa at (b) 1450 °C. EBSD results including (c) the band contrast figures, (d) inverse pole figures, (e) misorientation angle distributions, and (f) grain size distributions of ZrB₂ ceramics sintered under 15 GPa at 1450 °C. Reproduced with permission from Ref. [279], © The Authors 2022.

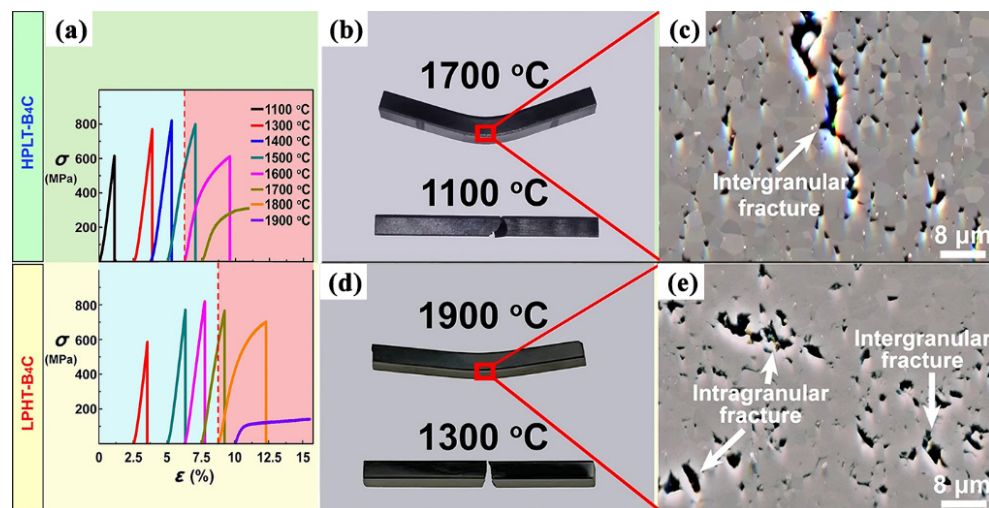


Fig. 42 High-temperature flexural test with B₄C ceramics at a constant strain rate of $5.7 \times 10^{-4} \text{ s}^{-1}$. (a) The upper and lower parts depict the high-temperature flexural stress-strain behavior of HPLT-B₄C and LPHT-B₄C, respectively. (b, d) Photos of HPLT-B₄C and LPHT-B₄C after high-temperature flexure at 1100, 1700, 1300, and 1900 °C, respectively. (c, e) Morphologies of regions under maximum strain during the test shown in (b) 1700 °C specimen and (d) 1900 °C specimen. Reproduced with permission from Ref. [281], © The Author(s) 2023.

because the consolidation process is solid-state. High-pressure sintering technology is a useful way to fabricate fully dense ceramics with suppressed grain growth and unique residual stress. Xu and coworkers [282] prepared nanocrystalline alumina ceramic with high compressive prestress and coherently aligned nanograins by UHPS technology, and the grain size of the ceramic was only 115 nm. The as-sintered specimen showed remarkably excellent mechanical properties, including a high hardness of 25.9 GPa and fracture toughness of 3.2 MPa·m^{1/2}, which were 36% and 51% higher than the corresponding parameters of sapphire, respectively [283]. The results showed that crack propagation could be restrained by compressive prestress, thereby enhancing the mechanical properties. Raman analysis showed a uniform residual stress distribution in the Al₂O₃ ceramics, and the compressive residual stress reached 264 MPa when the sintering temperature was 600 °C (Fig. 43(b)). Nanograins of 10–15 nm with coherently aligned grain boundaries were created by plastic deformation under applied ultra-high pressure, which remarkably contributed to the improvement in toughness and hardness. Researchers proposed a novel mechanism in which both hardness and toughness were simultaneously enhanced by compressive prestress and coherently aligned nanograins. The diagram briefly and clearly illustrates the mechanism, as shown in Fig. 44.

(4) Enhanced mechanical properties and oxidation resistance

Zirconium diboride (ZrB₂) has been widely applied in extreme environments because of its advantages, which include high hardness, high stiffness, high strength, excellent oxidation resistance at high temperatures, and high electrical and thermal conductivities. However, it is difficult to obtain fully dense ZrB₂ with fine grains because of its low diffusion coefficient. Xu *et al.* [279] prepared fully dense and grain-refined (decreased 56% from the raw powder) ZrB₂ at a relatively low temperature of 1450 °C

under an ultra-high pressure of 15 GPa. The as-produced ZrB₂ possessed significantly excellent mechanical properties, including high hardness (25.4 GPa) and fracture toughness (3.25 MPa·m^{1/2}), which were 46% and 69% higher than those of the high-temperature sintered ceramics above 2000 °C, respectively. The UHPS-ed ZrB₂ ceramics exist at a high dislocation density (1.7×10¹⁵ m⁻²), which is 3 times that of the SPS-ed specimen (Table 11). A developed densification mechanism of dislocation multiplication with grain refinement is proposed and proved to play a dominant role in the sintering process, which simultaneously enhances the mechanical and oxidation-resistance properties. In addition, the temperature at which UHPS-ZrB₂ started to exhibit obvious oxidation was 1100 °C (Fig. 45(b)), which was much higher than those of SPS-ZrB₂ (850 °C) and other reported ultrahigh temperature ceramics (TaB₂ (930 °C), ZrB₂ (800 °C), and HfC (700–800 °C)) [284–286]. The ZrO₂ layer of UHPS-ZrB₂ oxidized at 1100 °C and SPS-ZrB₂ oxidized at 850 °C show thicknesses of 15 and 40 μm, respectively. The results indicated much better oxidation-resistance properties for the ZrB₂ ceramics sintered by UHPS technology (Figs. 45(c) and 45(g)). The morphology characterization and elemental mapping showed that the nanosized structures of UHPS-ZrB₂ were recreated into fine ZrO₂ grains (Figs. 45(d)–45(f), and 45(h)–45(j)), and the densified ZrO₂ could enhance oxidation resistance by preventing further oxygen diffusion into the interior structure.

2.3.5 Conclusions

Advanced ceramics have wide applications in many fields due to their excellent properties, but their use is also constrained to some extent by their sintering method. Conventional sintering methods can no longer meet the demand for high-performance ceramics today, so developing a new and efficient sintering method is

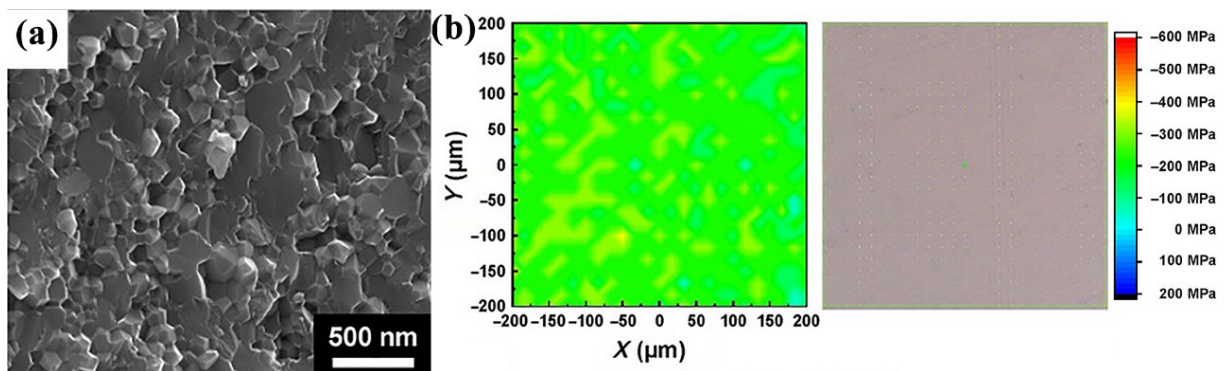


Fig. 43 (a) Fracture surface and (b) residual stress distribution calculated by micro-Raman mapping analysis of alumina sintered under 15 GPa at 600 °C. Reproduced with permission from Ref. [282], © The American Ceramic Society 2024.

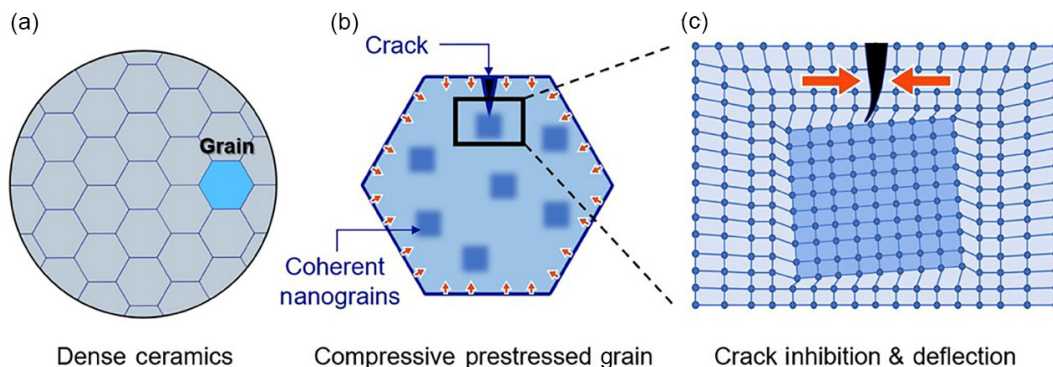


Fig. 44 Diagram of nanocrystalline ceramics with coherently aligned nanograins. (a) Fully dense ceramics. (b) Coherently aligned nanograins and (c) compressive prestress. Reproduced with permission from Ref. [282], © The American Ceramic Society 2024.

Table 11 The hardness, fracture toughness, grain size, and dislocation density of ZrB₂ ceramics fabricated under different conditions

T (°C)	p (MPa)	ρ (%)	HV (GPa)	K_{IC} (MPa·m ^{1/2})	d (μm)	ρ_d (m ⁻²)	Ref.
2000	25	97.5	14.85±0.7	—	20±4.9	—	[287]
1850	35	99.8	23.91±1.5	3.31±0.2	~5	—	[288]
2200	50	~98.2	14.4±1.33	2.52±0.13	8	—	[289]
2100	30	86	13±3	—	—	—	[290]
2630	—	~90–93	—	—	19.0±8.4	—	[291]
2100	32	99	15.3	3.9	5.9	—	[292]
1450	15×10 ³	100	25.4±1.2	3.25±0.15	0.7±0.09	1.7×10 ¹⁵	[279]
2000	200	99	17.4±0.8	1.92±0.14	19±1.33	2.78×10 ¹²	[279]

Note: T —temperature, p —pressure, ρ —relative density, HV—hardness, K_{IC} —fracture toughness, d —grain size, and ρ_d —dislocation density.

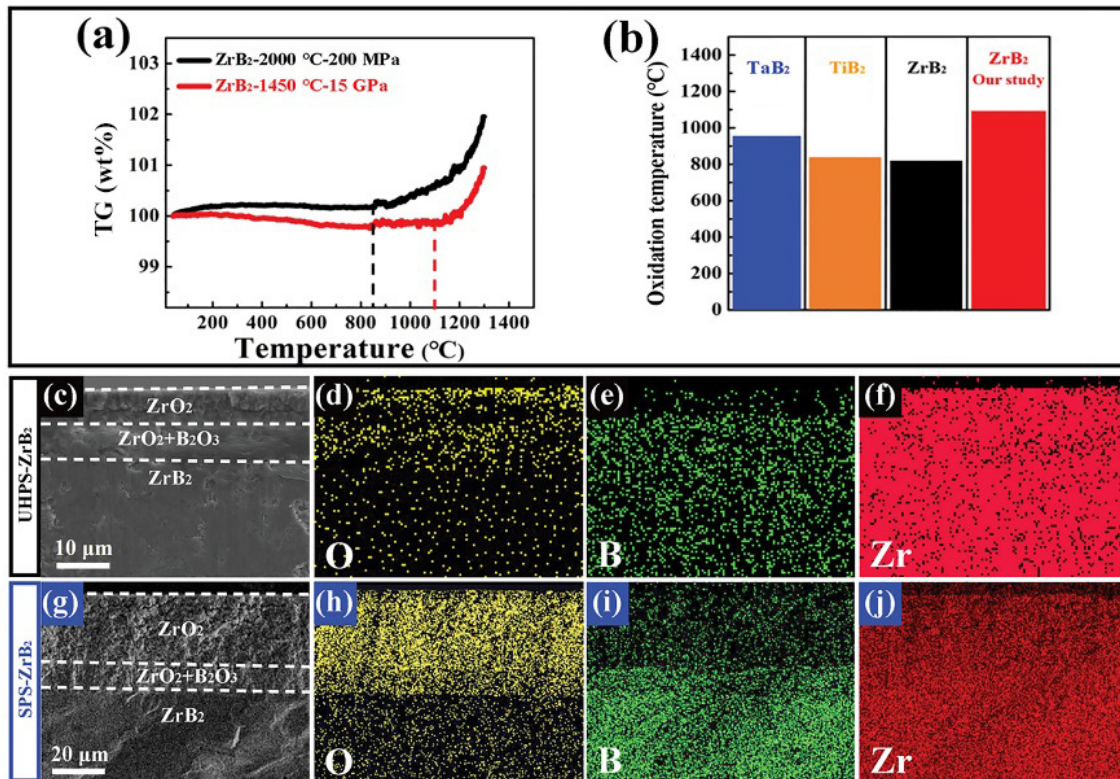


Fig. 45 Thermal analysis of UHPS-ZrB₂ and SPS-ZrB₂ in an air atmosphere: (a) thermogravimetric (TG) curves, (b) comparison of the onset oxidation temperatures of UHPS-ZrB₂ with SPS-ZrB₂ and other reported ultrahigh temperature ceramics, including TaB₂, TiB₂, and ZrB₂. Cross-section morphologies and EDS mapping images of as-sintered ZrB₂ ceramics after oxidation in air for 0.5 h at (c–f) 1100 °C for UHPS-ZrB₂ and (g–j) 850 °C for SPS-ZrB₂. Reproduced with permission from Ref. [279], © The Authors 2022.

particularly important. The conventional pressureless sintering method can obtain high density while inevitably causing abnormal grain growth, which will greatly reduce the mechanical properties of the material. Compared with conventional pressureless sintering, high-pressure sintering retains some very prominent advantages, which can greatly shorten the sintering time and obtain ceramics with better properties. Based on the large subdivision of the pressure applied, we reviewed two high-pressure sintering technologies for advanced ceramics: SPS and UHPS technology. The discussion is conducted from the following three aspects:

First, the influence of high pressure on material densification is discussed for different material systems. For some hard-to-sinter and ultrahigh-temperature ceramics with strong covalent bonds and low atomic self-diffusion coefficients, it is difficult for conventional sintering methods to densify them and generally require very high sintering temperatures and long soaking time,

which will greatly weaken the mechanical properties of materials. Under the condition of applying high pressure, high-performance ceramics with high density and without obvious grain growth can be quickly prepared at lower temperatures, and ultrafine nanocrystalline ceramics can be fabricated by accurately controlling the sintering temperature and pressure. Second, the sintering mechanism of ceramics under high pressure is explained in detail. The main densification mechanism of SPS and UHPS is still dislocation increment and grain boundary slip caused by plastic deformation, which can obtain high-performance ceramic materials with high density and fine grains. Compared with SPS, UHPS can effectively refine the grains, thus obtaining fully dense and ultrafine ceramics with a grain size smaller than the original powder. Finally, we presented a series of examples to illustrate the importance of high-pressure sintering for the optimization of ceramic properties.

3 Electric field-assisted sintering techniques

3.1 Spark plasma sintering

3.1.1 Brief history of spark plasma sintering

The word “ceramic”, coming from the Greek word “keramikos”, means “burnt stuff”. It indicates that desirable properties of the materials are normally achieved by high-temperature treatment processes of sintering. Sintering is a technique of consolidating powder compacts using thermal energy. As one of the oldest human technologies, it originated in pottery firing in the prehistoric era. In conventional sintering processes, powders and/or green compacts are placed and heated by external radiant heating in furnaces. This situation persisted until the end of the 19th century. In 1906, Bloxam [293] conducted direct current (DC) through metal powders and consolidated the compacts using the Joule heat generated from the resistance inside the samples. The technique is referred to as the first resistance sintering (RS) [294]. Weintraub [295,296] combined pressure into RS in the 1910s. It is considered the precursor of modern electric current activated/assisted sintering (ECAS) techniques that involve simultaneous applications of pressure, temperature, and electric fields for sintering. In the following decades, RS was further developed by researchers worldwide. In the 1960s, Kiyoshi Inoue [297,298] in Japax Inc. (Japan) innovatively introduced pulsed electric current for sintering and discovered that pulsed currents combined with pressure promoted interparticle bridging by spark discharges. The technique was patented as electric discharge sintering (EDS), which is also known as impulsive spark discharge sintering. Inoue also introduced the basic principle that discrete sparking at particle interfaces could act as an effective means of promoting local heating and thus interparticle bonding. Based on the patents, Inoue Japax Research Inc. commercialized the first generation of spark sintering and the second generation of plasma-activated sintering (PAS) [11], but they were not widely used. In 1989, Sumitomo Coal Mining Co., Ltd. (Japan) commercialized the present-day SPS apparatus (Model: Dr. Sinter) as the third generation of large DC pulse applied sintering. The birth of SPSs has gradually attracted widespread attention from both academia and industries worldwide. The first SPS equipment installed outside Japan was at California University Davis in 1996, in Europe at Stockholm University in 1998, and in China at Wuhan University of Technology in 2000. Since the 1990s, the term “SPS” has become popular in scientific literature. In many cases, it refers to the prevalent belief that plasma is generated between powder particles and accelerates sintering kinetics by cleaning particle surfaces [299]. However, until today, the formation of plasmas has never been proven without doubt [300,301], especially in the cases of insulating ceramics. Hence, the terms pulsed electric current sintering (PECS) [302] and field-assisted sintering technique (FAST, by FCT Systeme GmbH, Germany) [133] are also used for replacement. Although the names are different, the basic working principles are rather similar [303]. At present, SPS technology has been widely applied in the sintering of metallic materials [304], ceramics, polymers, and polymer-based composites [305].

3.1.2 Modifications of spark plasma sintering

The scheme of regular SPS is shown in Fig. 46(a). Similar to conventional HP, the powders or green bodies in the SPS are loaded in a die and pressed under uniaxial pressure. However, instead of using irradiating heating sources, a pulsed DC is allowed to pass through the electrical conducting die, in

appropriate cases, and through the samples. Such a die setup enables much higher heating/cooling rates and thermal efficiency during the SPS process. It has attracted great attention in ceramic fabrication for its remarkable accomplishments [306–309], including enhanced sintering, improved or unique properties, and the achievement of new microstructures and/or phases.

The regular SPS setup can be modified and combined with established sintering techniques to generate further variations. For instance, by adopting the conception of a matryoshka doll, a high uniaxial pressure, up to 1 GPa, can be applied, referred to as the high-pressure SPS (HPSPS) scheme in Fig. 46(b). The pressure that graphite molds can withstand does not exceed 150 MPa. To survive higher pressure, several materials are used as the inner molds: WC [310] and types of hot work tool steels [311] are used for sintering temperatures lower than 1000 °C, while SiC and Si₃N₄ can be sustained up to 1400 °C. A dedicated ultrahigh temperature, high-pressure mold material, called SL-400H [312], has also been reported. The utilization of high pressure effectively decreases the sintering temperature and consequently suppresses grain growth during the densification thermal histories. The HPSPS was first reported by Prof. Munir’s group [313,314] in the preparations of zirconia nanoceramics. Under the brutal force, the densification temperature can be greatly reduced, and consequently, grain growth can be effectively suppressed. The inventors speculated that plastic (or superplastic) deformation played a very important role in the HPSPS technique. The later work by Ji *et al.* [273] evidenced the coexistence of two types of plastic deformation, dislocations and sliding, in nanocrystalline 3 mol% yttria-stabilized tetragonal polycrystal (3Y-TZP) ceramics prepared under 1.5 GPa pressure. The results confirmed for the first time the dominant role of plastic deformation in the HPSPS. The technique has been successfully applied in the fabrication of a variety of fine-grained bulk ceramics [315].

In addition, the external pressure during the regular SPS process can be totally removed by the mold design. In 2012, Bradbury and his colleague [316] reported the production of SiC–C composites using the SPS apparatus, as shown in the schematic in Fig. 46(c). In their work, regular cylindrical punches were replaced by T-shaped punches to realize pressureless conditions during the SPS process. They named the novel technique the free-pressureless SPS (FPSPS). Wang *et al.* [317] reported the synthesis of nano HfB₂ powder using a graphite crucible in SPS. The authors also used punches of larger diameters against the die [318] for FPSPS, as shown in the schematic in Fig. 46(d). In the FPSPS condition, the sample is not subjected to any external pressure but only to fast heating. It was reported that the contact resistance was vital for obtaining high temperatures and Joule heating rates during PFSPS, and the resistance must be sufficient to provide resistance between the punch exteriors and die [319]. The utilizations of the FPSPS technique mainly include but are not limited to powder synthesis, preparations of porous materials, and consolidations of bulk ceramics with complex geometries.

The pressure in the common mechanical field-assisted sintering technique is static. Professor Xie’s group [1] developed a novel hot oscillatory pressing technique by coupling oscillatory pressure into conventional HP. The static pressure during the regular SPS can also be transformed to be dynamic. The samples are heated by converting alternating current (AC) into DC pulses through filters during the SPS process. Shen’s group [320] reported that oscillatory pressure could be introduced into the SPS process by adjusting the DC pulse ratios. Moreover, it can be programmed to achieve a pressure output similar to that of hot forging in SPS [321].

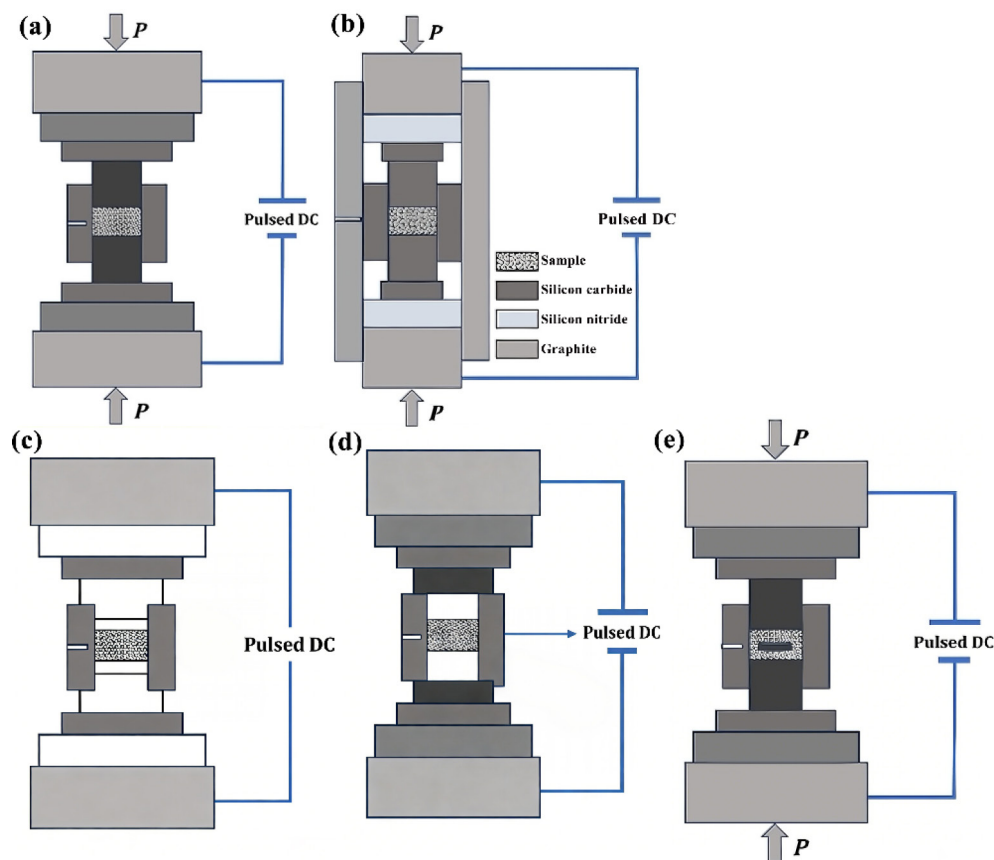


Fig. 46 Schemes of (a) regular SPS, (b) HPSPS, (c) and (d) FPSPS, and (e) pseudoisostatic SPS.

The external force during regular SPS is commonly uniaxial pressure. It can be transformed to quasi-isostatic pressure during the sintering process. Inoue's patent issued in 1972 was viewed as the first pseudoisostatic ECAS [303]. Fu's group [322] recently reported the preparation of KNTeO_6 transparent ceramics. In their work, the samples were embedded in magnesium aluminate powders loaded in the SPS graphite die, as shown in the schematic in Fig. 46(e). The setup was proposed to take advantage of the particle rearrangement and plastic deformation by shear stress compared with the regular SPS process.

3.1.3 Strategies in spark plasma sintering

The sintering of SPS, in most cases, is processed at a constant heating rate, and the output current gradually increases as the sintering temperature increases. Alternatively, the sintering profile in SPS can be controlled by the output of current, known as the current control mode (CCM). In this mode, even faster heating rates can be achieved during the early and intermediate sintering stages. Prof. Sakka's group [323,324] investigated the temperature and temperature distribution of CCM and prepared ultrafine-grained WC ceramics. Niu *et al.* [325] also reported the fast densification of pure B_4C ceramics in this mode.

The heating rate of SPS can be more than 1000 °C/min. The fast heating rate of SPS is beneficial for obtaining refined microstructures. On the other hand, it may be adverse to the homogeneity of the final products. Kim *et al.* [326] and Morita *et al.* [327] reported that a lower heating rate was actually beneficial for the preparation of transparent Al_2O_3 and MgAl_2O_4 ceramics with uniform color and good optical properties. They interpreted that lower-speed heating was beneficial for grain boundary diffusion and defect concentration reduction. In

addition, a lower heating rate was recommended for the sintering process involving exothermic chemical reactions [328,329].

The pressure is commonly loaded from the beginning and kept constant until the end of the SPS cycles. Wang *et al.* [330] reported that the homogeneity and transparency of MgAl_2O_4 ceramics prepared by SPS decreased with increasing preloaded pressure. A similar phenomenon was also observed in the case of Al_2O_3 [331]. Santanach *et al.* [332] reported that step-by-step pressure loading could enhance grain sliding and rotation at high temperatures to achieve homogenous microstructures in final products.

Chen and his colleague [12] proposed a novel two-step sintering method for nanoceramic preparations in 2000. Schwarz *et al.* [333] adopted the strategy in cubic yttria-stabilized zirconia in SPS. The latter works by Reddy *et al.* [334,335] developed the multistage SPS technique in the preparation of fine-grained oxide ceramics. Moreover, a strategy in which a partially SPS-densified sample is further subjected to hot isostatic pressing treatment (SPS+HIP) was also reported in the preparation of transparent ceramics [336]. Grasso *et al.* [337] developed flash SPS (FSPS), which combined the advantages of both techniques.

The graphite die is an important heating source in SPS. In addition to the cylindrically shaped mold, the cone-shaped mold is also used to generate a temperature gradient during sintering, especially for the preparation of laminated ceramic composites [338]. Through mold design, it is also possible to prepare samples with curved surfaces [11].

Carbon contamination is a common issue arising from the use of graphite dies in SPS. When the sintering temperature is not higher than 1000 °C, other materials, such as WC-Co, Mo alloys, quartz, and alumina, can also be used as molds.

3.1.4 Mechanistic explorations of spark plasma sintering

Great attention has been given to exploring the mechanisms underlying the fast sintering of SPS. Possible contributions from plasma, electric field, heating rate, temperature distribution, and pressure have been widely investigated. However, simultaneous influences from those factors during the SPS process make the investigation rather complicated. A universal explanation for the rapid densification achievable by SPS is still unavailable, especially in the cases of nonconductive ceramics.

Plasma excitation and/or spark discharge were observed in the spark plasma sintering of conductive materials, mainly metals and metal-based composites. The observation of pulsed current flow through a semiconductive specimen, for instance, ZnO [339], was also reported. The formation of “discharge necks” and net-like “bridging” between metal powder particles during SPS sintering was experimentally reported by some researchers. It was proposed that electric fields induced positive and negative poles in the microregions of the particles. Under the action of the pulse current, discharge occurs between particles, exciting plasma. This plasma promotes sintering by purifying the particle surfaces. It is suggested that the plasma generated between metal particles by the pulse current plays a role in the sintering process and the formation of oriented grains. Simultaneously, the pulsed electric field reduces the diffusion free energy of metal atoms, accelerating atomic diffusion, while Joule heating (self-resistance heating) also promotes plastic deformation.

The explanation for the rapid densification of metallic materials via SPS technology was “borrowed” in explaining the fast sintering of nonconductive ceramic materials by many researchers. Some works attributed the rapid densification mechanism in SPS primarily to interparticle discharge and plasma excitation during the sintering process. However, it is absent in the process of nonconductive ceramics. It was reported that the current passing through the alumina was less than 100 nA in SPS when the sintering current was 1000 A, and the change in pulse current made a negligible change in the current passing through the nonconductive samples [340]. Hulbert *et al.* [341] employed three separate experimental methods, including *in situ* atomic emission spectroscopy, direct visual observation, and ultrafast *in situ* voltage measurements, to determine the presence or absence of plasma during SPS and reported that no evidence of plasma was found in the electrical insulating specimens.

During SPS, the sample is heated by a large current (several thousand amperes) and several volts. The contribution from the electric field is another issue of controversy. The relevant research of the supporters mostly pertains to ceramics that have at least some degree of ionic character so that the anions and cations and

the vacancies and interstitials arising from them can form electrical dipoles with some measure of electrostatic force. Based on theoretical calculations, Raj *et al.* [342] claimed that the DC voltage (3–10 V) of the SPS system was enough to produce significant fields across a thin insulating sample, for instance, yttria-stabilized zirconia (YSZ). Chen *et al.* [343] attributed the formation of wave-like grain boundaries in 8YSZ ceramics to the ion-wind effect under the electric current of SPS. Ceramics that are highly covalent may behave in a different way. A similar effect was not experimentally observed in other oxides, such as Al₂O₃ [344] and ZnO [345]. Until now, quantitative methods have been lacking to benchmark the contribution of electric fields in the SPS process.

One of the possible reasons for the rapid sintering of SPS is the higher actual sample temperature than the measured value [346]. The temperature during SPS is monitored either by a pyrometer focused on the outer wall or by a thermocouple inserted into the mold. Therefore, there is a difference between the actual temperature of the sample and the measured value. Vanmessnesl *et al.* [347] conducted finite element simulations on the temperature fields of SPS-sintered conductive TiN and nonconductive ZrO₂ ceramic materials. The comparative results showed that the temperature gradient in conductive TiN gradually decreased from the center of the sample to the wall of the mold, while the situation was inverse for insulating ZrO₂. In addition, the contact resistance has an important influence on the temperature distribution during SPS. However, a higher temperature was observed at the place with higher contact resistance [318]. Moreover, sample heating in SPS is regulated by the output of current and voltage. A higher heating rate would be expected to result in a larger temperature difference. The correction between the measured temperatures vs. the actual values was also studied. In addition, under fast heating conditions, the process of SPS deviates far from the equilibrium state of conventional sintering. In such a nonequilibrium state, novel sintering mechanisms, for example, dynamic ripening [348], were discovered.

3.1.5 Intensive particle rearrangement in spark plasma sintering

Intensive particle arrangement was reported in the case of partially densified 3Y-TZP nanoceramics with a relative density of ~93% prepared by SPS, as seen in Fig. 47 [349]. Residual pores in the samples were dramatically amplified by more than 10 times, accompanied by rather limited grain growth during prolonged soaking in pressureless sintering conditions. This phenomenon was attributed to grain movement, such as sliding and rotation, even in the high-density sintering stage.

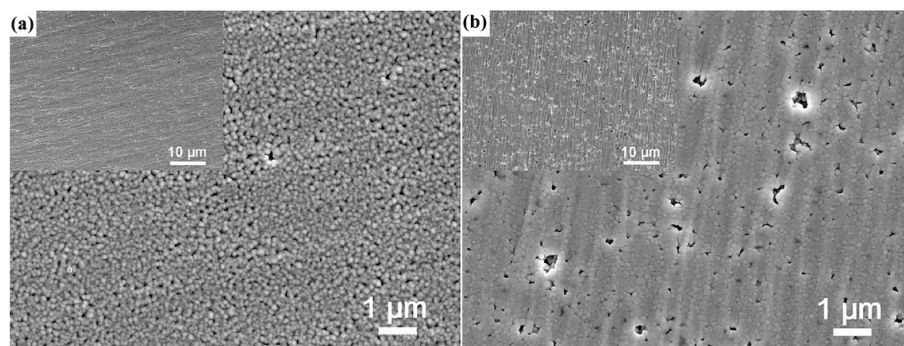


Fig. 47 (a) SEM images revealing the microstructure in 3Y-TZP ceramic samples with ~93% relative density prepared by SPS at 1100 °C for 6 min under 100 MPa pressure. (b) After 30 h soaking in pressureless sintering conditions, dynamic pore amplification was observed with rather limited grain growth. Reproduced with permission from Ref. [349], © The American Ceramic Society 2011.

In classical sintering theory, it suggests that particle rearrangement only occurs in the initial stage. The movement of grains ceases, accompanied by the formation of rigid necks between particles, and sintering enters the intermediate stage. The empirical relative density values for the final sintering stage are ~93% and ~95% for pressureless sintering and hot pressing, respectively. However, the situation may be different as the grain size enters the nanometric range. The previous work by Chen and his colleague [350,351] showed that particle rearrangement took effect in the pressureless sintering of nanoceramics with a relative density higher than 80%. Harris *et al.* [352] reported *in situ* observations of grain rotation in nanometric Au films, and their simulation indicated that the rotation rate increased by a factor of 16 when the grain size was halved. The discovery of intensive particle rearrangement challenges the classic sintering theory, which is based on the sintering behaviors of coarse powder compacts with grain sizes mostly in micrometers.

The intensive particle arrangement mechanism may offer possible explanations for some SPS phenomena. In 1998, Penn and Banfield [353] discovered a novel oriented attachment/aggregation (OA) mechanism for crystal growth, where larger grains were formed by self-assembly of smaller grains. As an important complement to classic Oswald ripening, the OA process essentially involves grain rotation for crystal orientation adjustment. Grain growth by the OA mechanism, which is common in liquid environments, was also reported in cases of Si_3N_4 [354], SrTiO_3 [355], Al_2O_3 [356], and BaTiO_3 [357] in SPS conditions (Fig. 48).

The ideal two-sphere model is conventionally used to describe the grain-growth behaviors during sintering. The movement of grains/particles ceases with the formation of rigid necks between particles. The fast heating of SPS can effectively suppress the growth of necks. Chaim *et al.* [93] proposed the formation of a softened surface in spark plasma sintering nanoparticles. Although such a softened layer in ceramics had not been experimentally confirmed as that in metals [358], the speculated analogy may be plausible since atomic migration-related phenomena, such as grain-boundary diffusion and migration, always initiate from the particle surface. One consequent impact of such surficial flux is to decrease friction between particles. Furthermore, the rapid heating rate of SPS can effectively suppress the growth of grains as well as the necks between grains. These facts thereby favor intensive particle rearrangement through grain rotation and sliding during spark plasma sintering, as shown in the schematic in Fig. 49.

One of the typical experimental observations is the unusually high densification rate. The relative density rapidly increases from ~50% to ~90%, accompanied by rather limited grain growth. The maximum speed can be in the 10^{-3} s^{-1} order in solid-state sintering and even in the 10^{-2} s^{-1} order in liquid-phase sintering [359], as shown in Fig. 50(a). The value in conventional mechanical field-assisted sintering, such as HP and HIP, is commonly on the order of 10^{-4} s^{-1} or even less. A grain and an atom are the only two basic units during sintering. In classic theory, sintering is predominantly governed by atomic diffusion-related processes, and the contribution to density increase from particle packing is suggested to be rather limited. Bernard-Granger *et al.* [69] reported that the

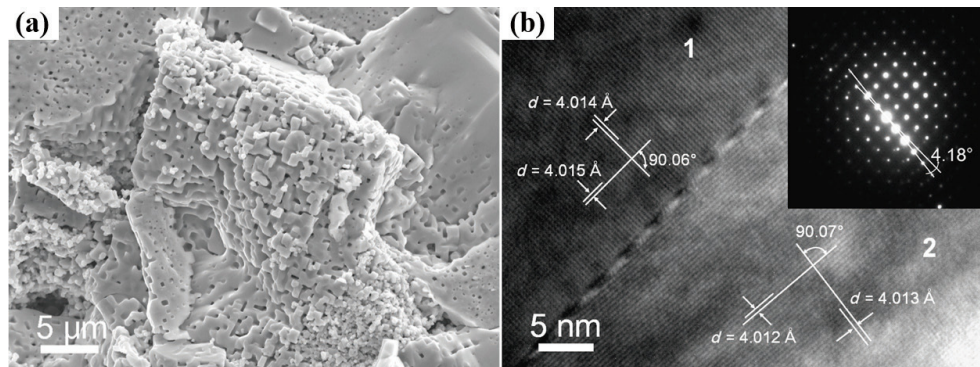


Fig. 48 (a) SEM and (b) TEM images of abnormal grain growth during spark plasma sintering of BaTiO_3 ceramics. Reproduced with permission from Ref. [357], © Elsevier Ltd and Techna Group S.r.l. 2022.

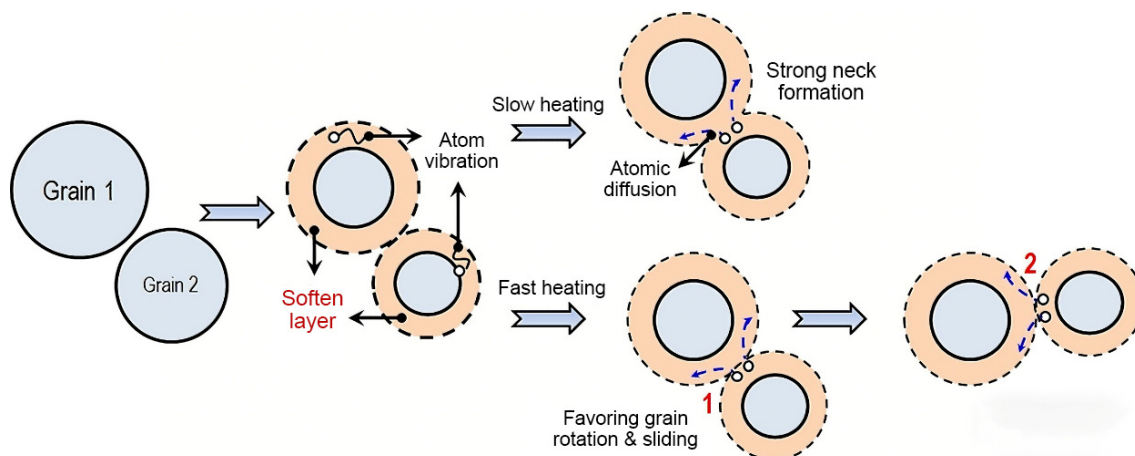


Fig. 49 Modified two-sphere model during spark plasma sintering, during which the formation of a softened layer was proposed to enhance the intensive particle rearrangement. Reproduced with permission from Ref. [357], © Elsevier Ltd and Techna Group S.r.l. 2022.

maximum densification rate was always observed in the 64%–68% relative density range when the heating rate ranged from 100 to 900 °C/h in pressureless sintering of 3Y-TZP ceramics, while the grains hardly grew, as seen in Fig. 50(b). Comparatively, under SPS conditions, the maximum densification rate was always observed at a relative density of ~78% with no grain growth when the heating rate was 100 °C/min. However, the maximum densification rate shifted to the ~85% relative density region with observable grain growth when the heating rate decreased to 10 °C/min [359]. For a certain powder, the maximum density achieved by physical packing is relatively constant. The fast heating of SPS greatly constrains the density increase from atomic diffusion, especially during the early and intermediate sintering stages. The grain, as the basic building unit, is much larger than the atom. Combined with the intensive particle rearrangement, the contribution from powder packing to the rapid densification of SPS is worthy of reconsideration.

3.1.6 Conclusions

Ceramics can be viewed as composites of pores and crystalline grains. Densification is then the process through which pores are replaced by solid phases. One of the major obstacles for fine-grained ceramic preparations is rapid grain growth during the final stage of sintering. Ceramic materials have a relatively high elastic modulus, and their plastic deformation is very limited. During the final sintering stage, residual pores can only be eliminated by interfacial atomic diffusion since the microstructure has been frozen due to physical constraints and chemical bonding with neighboring grains. Hence, the state of pores, such as pore size and distribution in the sintered body, has a decisive impact on the densification behaviors. Compared with the dynamic pore amplifications, the microstructures in SPS conditions experienced spontaneous refinement such that the pores were small and homogeneously distributed in the sintered bodies [336]. These pores could be easily removed at relatively lower temperatures. By SPS die-upset design, each possible influence from the electric field, heating rate, and pressure was peeled to identify its contribution to the spontaneous microstructure refinement in SPS. It was found that neither electric current nor fast heating had any decisive contributions, while pressure was a necessity [360]. The intensive particle rearrangement could be further enhanced under external force. The full advantages of this mechanism can be taken in rapid heating conditions. Combined with the application of higher pressures, this makes the family of fast sintering techniques advanced in the preparation of fine-grained ceramics over their conventional counterparts characterized by slow heating features.

Although the intensive particle rearrangement (IPR) mechanism was discovered during the SPS process, it would be expected to have significance in both general sintering theory and product industrialization. A typical example is the fast sintering of 3–4Y-TZP ceramics currently used in denture restoration, where the densification of a single zirconia crown and 3-unit bridge can be finished within 15 min. The routine highlights the particle packing homogeneity in green nanoceramic bodies, which is preferred to be obtained via wet-chemical processing to suppress pore dynamic enlargement during sintering. Careful *in situ* observations at high temperature in SEM and/or HRTEM will help reveal the microstructural evolution of this mechanism in greater detail.

3.2 Ultrafast high-temperature sintering

3.2.1 Introduction and overview of the processed materials

The paradox of sintering is that the process is exothermic as it consumes surface area [361]; however, most of the heat is irreversibly lost. In fact, during sintering, the heat, instead of being accumulated within the ceramic workpiece, is irreversibly lost toward the surroundings via a combination of convection, radiation, and conduction-based mechanisms. As an innovative attempt to minimize energy waste and to decarbonize the firing process, Wang *et al.* [362] in 2020 developed ultrafast high-temperature sintering (UHS). Despite the centuries of evolution in the design of ceramic kilns, clearly, UHS represents a disruptive innovation, as it surpasses multiple challenges using a low-cost and effective approach. The technique suits the consolidation of various structural and functional materials, as shown in Fig. 51.

3.2.2 Sintering processing setups

The UHS technique is not material specific, and it can be employed to consolidate particles with sintering temperatures even above 3000 °C. The core concept is to employ a thermally insulating heater energized by the Joule effect. The thermal energy is directed to the workpiece, minimizing heat losses. In other words, the heater also serves as a thermal insulator, with thermal conductivity on the order of 10^{-1} W/(m·K), greatly simplifying the construction of a furnace. The techniques feature a heating rate of the sintering parts on the order of 10^3 – 10^4 °C/min. Under such conditions, both the heater and the sintering parts are subjected to a significant thermal shock. As a result, graphite-based heaters, having a layered van der Waals bonded structure, are preferred to sustain the enormous thermal shocks during repeated firing cycles.

The original configuration employed either graphite felt (30 V,

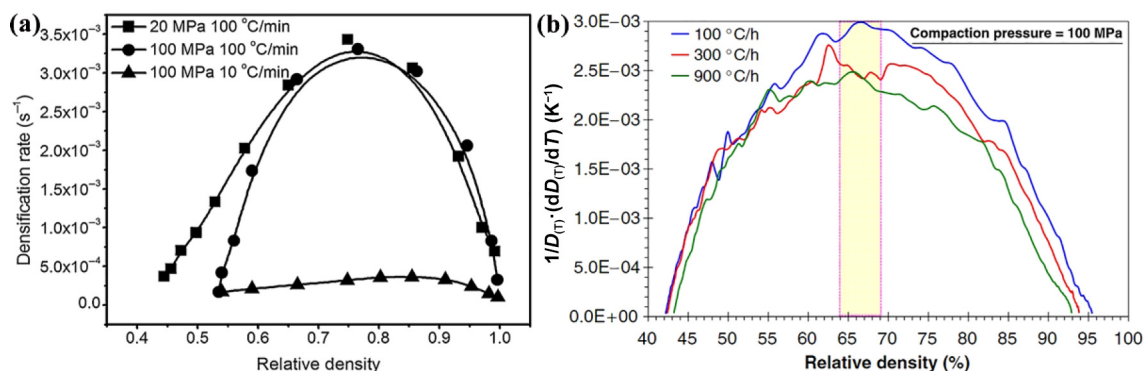


Fig. 50 Relationship of relative density vs. densification rate during (a) spark plasma sintering and (b) pressureless sintering of 3Y-TZP nanoceramics. Reproduced with permission from Ref. [359] for (a), © The Ceramic Society of Japan and the Korean Ceramic Society 2015; from Ref. [69] for (b), © The American Ceramic Society 2007.

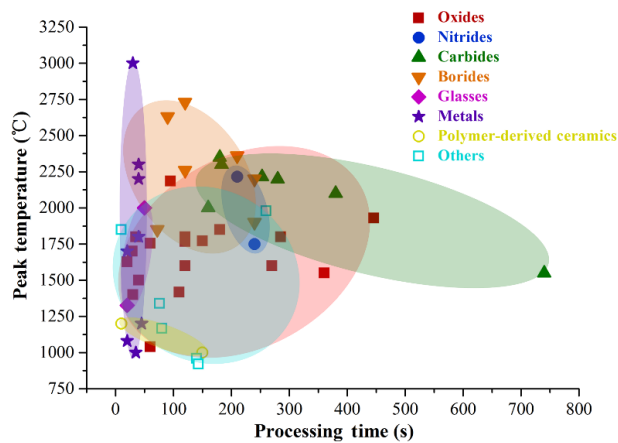


Fig. 51 Overview of the inorganic materials processed using UHS grouped with respect to their composition. The plot correlates the peak processing temperature with sintering time. In comparison with other pressure-less sintering techniques, UHS typically features increased sintering temperature and shortened processing time. Oxides [362–377], nitrides [378,379], carbides [380–386], borides [387–391], glasses [392,393], metals [394–396], polymer-derived ceramics [362,397], others [398–403].

50 A) or graphite paper (10 V, 150 A [368]) as a heater or used a graphite powder bed (30 V, 100 A [366]). As shown in Fig. 52, the sample is located in close proximity to the heater energized by Joule heating. The heat transfer mechanism between the heater and the sample to be fired is rather complex, and the heat exchanges depend on the employed configuration. Assuming nondirect contact with the sample under vacuum, heat transfer, as modeled in Ref. [362], is mostly achieved through radiation. However, when the sample is in direct contact with the heater,

heat transfer by convection and conduction should not be neglected [363].

The typical processing UHS run consists of several consecutive steps, including the selection of starting powders, powder pretreatment, green body preparation, and firing, followed by posttreatment if needed. Unlike conventional firing, powders resulting in minimal outgassing upon heating are preferred; in fact, the ultrafast heating rate causes a surge in the vaporization and/or sublimation of volatile species, eventually causing premature cracking during firing. As-received powders are commonly compacted using uniaxial pressing; however, better results are typically obtained when using isostatic pressing [404]. The green density gradients, induced by uniaxial pressing, for example, are exacerbated during firing. Alternatively, other techniques, such as slip casting, tape casting, or gel casting, contributing to even density distribution throughout the green body, are also available options. While additives and residual organic fraction associated with the green body preparation can be removed by lowering the heating rate down to 1 °C/min during conventional firing, in the case of UHS, the use of binder-free compaction is the preferred option to produce dense parts without the need to lower the heating rate. Alternatively, the debinding step is typically carried out using a conventional benefit of improved homogeneity of the sintered pellets [402]. UHS, being a green firing technology with minimal energy consumption, ideally still requires a debinding step to consume much less energy than the firing step.

Despite the effort of Bhandari *et al.* [371] in developing a protocol for ultrarapid debinding followed by ultrafast high-temperature, there is a need to identify binders compatible with fast heating rates. As a result, recently, the combination of cold sintering followed by UHS has been effectively demonstrated to

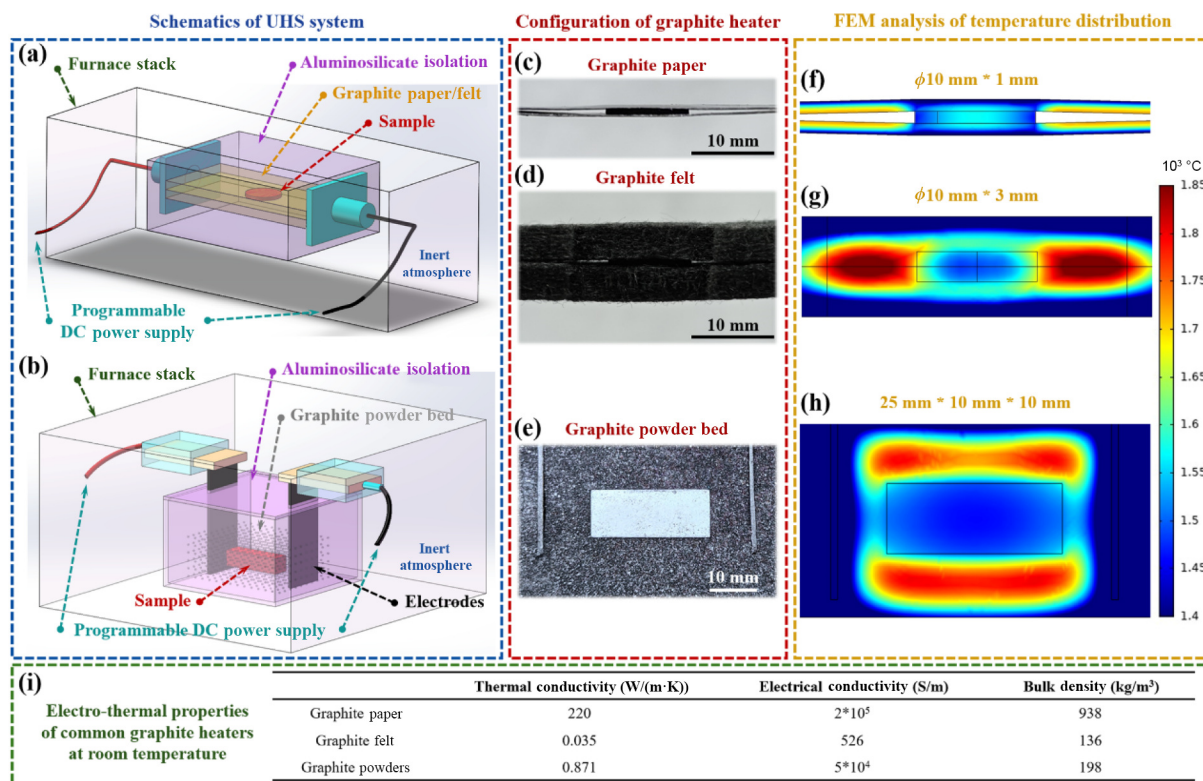


Fig. 52 (a, b) Schematic diagram of existing typical UHS experimental devices. Based on (c–e) their different heating configurations and (i) electrothermal properties of different graphite heaters, (f–h) the ultrafast heating process (without temperature holding) exhibits different features for applicable sample size and temperature distribution. Reproduced with permission from Ref. [378] for (a), © Elsevier Ltd. 2021; from Ref. [366] for (b), © Acta Materialia Inc. 2022.

offset the need for a binder during processing [374]. Additionally, the combination of these two emerging sintering techniques is aimed at maximizing the green density of the compact, facilitating their high-temperature consolidation [405]. In particular, chemically or physically absorbed water or other species formed during wet processing can potentially result in cracking of the specimens under ultrafast heating as a result of their volumetric expansion during heating. Additional research efforts are required to engineer particles in terms of purity, green density, and storage, which are better compatible with ultrarapid heating without causing cracking of the parts during heating. For example, free carbon is commonly added to compensate for the oxidic impurities in carbide ceramics; however, in ultrafast firing, the accelerated kinetics of sintering lasting a few tens of seconds might not allow a complete reaction.

Inevitably, during ultrafast heating, surface-to-core thermal gradients develop. Due to radiative heat transfer, the surface is hotter than the sample core upon heating and has an inverted trend during cooling. It has been shown that the greatest temperature gradient (ΔT) for an “infinite slab” geometry between the surface and the core can be approximated using (Eq. (9)):

$$\Delta T = \frac{l^2}{2\lambda} \phi \quad (9)$$

where l is the half thickness of the slab; λ is the thermal diffusivity; ϕ is the heating rate on the surface of the sample. It is worth noting that the thermal diffusivity for sintering particles is a function of both the temperature and relative density prior to the first stage of sintering. In the case of oxides, green bodies can be as low as 0.1 W/(m·K) for ZnO and 0.6 W/(m·K) for alumina [406]. As a result, a modulated heat profile with a stepwise increase in power is typically required to homogenize the temperature and prevent cracking. Apart from cracking, abrupt heating of a green body with ultralow thermal conductivity seems to cause a hot-to-cold condensation effect, resulting in blackening of the samples. An alternative approach to mitigate these effects is to employ suitable sintering additives capable of enhancing the mass transport, necking, and, in turn, thermal conductivity of the sintering particles. Another effective approach is a multistep heating profile to allow temperature equilibration.

As in most ultrafast firing techniques, temperature measurements are rather challenging, especially when the heater serves as thermal insulation, blocking the point of observation when using noncontact pyrometers. Additional undesired effects compromising the quality of the temperature measurements include (i) the uneven temperature distribution causing large spatial thermal gradients [400] and (ii) the mismatch in thermal mass of the heater of a few grams, the sample, and the thermocouple of tens of grams [407]. According to our experience, shielded thermocouples suffer a delay in the temperature readings, while pyrometers allowing a direct measurement of the temperature on the sample surface might cause undesired cracking due to radiative heat losses. Likewise, unshielded bare thermocouples with diameters down to 0.1 mm should not be in contact with the conductive felts. Initial work by Dong *et al.* [370] suggested the use of metal wires to reliably calibrate the temperature against their melting point. However, the geometry and placement of metal wires or ceramic pieces have a relevant impact on the temperature evaluation. Therefore, combining the technical characteristics and practical limitations of different temperature measurement methods, it is recommended to integrate the experimental measurements with the finite element model. A temperature evaluation method that combines

may be a solution. This approach may provide a relatively more accurate and comprehensive determination of the sample temperature.

3.2.3 Advantages and opportunities

The advantages of UHS have been widely summarized in a recent review [4]. The processing advantages of the UHS technique can be divided into three categories: (i) multilateral integration, (ii) fine microstructural tuning, and (iii) accelerated kinetics under ultra-high temperature conditions. These aspects feature unique advantages of the technique, as summarized below:

(1) Multimaterial integration

As a pressureless fast heating process controlled by conventional heat transfer mechanisms between the heater and the sample (thermal radiation, conduction, and convection), the UHS is universally applicable for bulk materials in the form of pellets or even more complex three-dimensional (3D) shapes [362,366,371,408]. Thus, the technique offers distinct advantages in achieving a multiscale design and manufacturing of sample components as finished parts. As a proof of concept, this strategy has been successfully applied in multilateral integration for various applications ranging from ceramic solid-state electrolyte films [409], drill bits with high-entropy boride composite coatings [389], and TiB₂/SiCNO thin-film-based sensors [397]. The ultrafast high-temperature sintering process can not only minimize the interlayer cross-contamination/reaction between multilayer materials/structures but also aid the formation of new and controlled interfaces with improved bonding characteristics.

In general, the UHS route may also preserve the desired phase composition. This control can be generally supported by two physical effects. First, it is the reduction in losses of volatile elements due to reduced time exposure above a critical temperature. The high-efficiency heat transfer with exceptionally high heating and cooling rates allows for a remarkable shortening in the sintering cycle, thus preventing undesired volatilization/decomposition of light elements [367,377,402] and/or solid-state reactions between phases [392,409]. Second, heating rates on the order of 10³–10⁴ °C/min combined with high temperature may promote the sintering system evolving to a nonequilibrium state, forming transient compositions without reaching thermodynamic equilibrium, as in the case of conventional firing [378,381]. The effect of a nonequilibrium state on composition is generally attributed to defect chemistry [373,410,411] and kinetically incomplete reactions, which might also provide an additional chemical driving force for densification.

(2) Fine microstructural tuning

Grain refinement is one of the most frequently observed microstructural features induced by UHS [363,370,372]. In general, the sintering process involves competition between densification and grain growth. Assuming that the activation energy for densification is greater than that for grain growth, it is then possible to achieve a denser microstructure as well as finer grains via fast heating. In this sense, the UHS process greatly reduces the time spent in the early stage of sintering, thereby maintaining fine grains and a high densification driving force up to high temperatures, where densification-relevant diffusion mechanisms can occur. Apart from the grain size, the grain shape can also be affected by ultrarapid heating, although this process also requires being combined with additional conditions, such as the existence of phase transformation [378], liquid phase precursors [365], and chemical reactions [412].

The effects of UHS on the pore size and its distribution [366,398,400] should not be neglected. It has been observed that

the materials prepared by UHS might possess finer pores with a reduced coordination number [372], and the proportion of intragranular pores seems significantly higher than that observed in conventionally sintered counterparts [366,400]. Ultrafast heating was considered the key contributor to these microstructural features [372], and it is unlikely that the weak electric field present in UHS might play a significant role. In any case, UHS engineering of pores and interfaces seems to contribute to an improvement in the material performance [391].

(3) Accelerated kinetics at ultra-high temperature conditions

Both wetting and chemical reactions are key elements that control the sintering process. For wetting, ultrafast heating may enhance liquid flow between particles. The liquid phase formation is justified by the increased processing temperature. The rapid homogenization of the liquid phase within the matrix will then accelerate mass transfer, in turn affecting the densification, reaction, and microstructure evolution [378].

Regarding the reactivity, during the shortened thermal cycle, the solid-state reaction would be kinetically activated only assuming a significantly increased temperature. The existence of a liquid-phase environment would promote more uniform reaction processes and direct morphological phase control over the sintered samples [413]. Accordingly, the ultrafast heating and high temperature in the UHS technique enable higher reaction efficiency [394] and better control over the reaction process [414]. UHS provides a novel strategy to more effectively balance wetting, reaction, and consolidation, which should be challenging to achieve with conventional approaches and hence leads to high selectivity of desired final products and improved performance.

3.2.4 Challenges and future trends

Table 12 highlights the aspects of differentiation with respect to UHS compared with conventional sintering. Some of the key features, advantages, and challenges are summarized in the table below.

The UHS key feature is represented by an ultrafast heating rate requiring readaptation of the ceramic processing flow layout. The analysis proposed in Fig. 53 suggests future development trends. The roadmaps cover the fundamental research aspects relevant to the fundamental mechanisms theory of sintering along with different engineering challenges using *in situ* techniques [415]. These effects, including the formation of defects, are particularly difficult to capture due to their ultrarapid evolution during the firing step. Ultrafast heating also has profound implications for the thermokinetic effects, both at the interface level in the case of

multimaterial integration or phase transformations and reactions during sintering.

Assuming that the research on the fundamental aspects along with thermodynamic analysis points in the right direction, there is plenty of effort required for suitable instrumentation. Despite the significant research efforts, the UHS technique is currently at deployment technological readiness level (TRL 7–9). Among the engineering challenges, to the best of our knowledge, UHS equipment has not been commercialized outside China, and lab-built equipment is commonly employed for carrying out this research. The latter suggests a need for the standardization of the UHS equipment to obtain more reproducible and controllable results. Assuming that a large system is capable of sintering parts with diameters well above 10 cm in size, there is still a need to improve our understanding of the heat transfer mechanism between the heater and the sintering parts. The latter could guide the development of UHS furnaces, resulting in homogeneously sintered parts with halved energy consumption compared to conventional firing. In the long term, the expectation is to build ultra-high energy-efficient furnaces with continuous throughput.

3.3 Flash sintering

3.3.1 Introduction to flash sintering

The phenomenon known as “flash sintering” was first reported by Cologna *et al.* [13] in 2010. In their pioneering investigation, an electric field was applied across a YSZ specimen held at 850 °C, resulting in rapid densification of the material within a matter of seconds. This experimental methodology has since been designated flash sintering.

The typical experimental setup for flash sintering is shown in Fig. 54(a). A ceramic specimen is positioned inside a furnace held at a constant temperature, and its ends are electrically contacted via electrodes connected to an external circuit. An ammeter, a voltmeter, and related instrumentation record the evolution of current and voltage throughout the flash event, while a high-speed camera documents concomitant changes in the specimen's appearance. The flash sintering process itself can be divided into three successive stages—incubation, runaway, and steady-state—as illustrated in Fig. 54(b). During the incubation stage, a gradual increase in applied voltage produces only a slow rise in current; power dissipation remains negligible, and thermal effects are minimal, so the specimen temperature stays essentially constant. The runaway stage is characterized by a sudden surge in circuit current coupled with an abrupt drop in sample voltage;

Table 12 Summarizing key features and challenges concerning UHS research

Aspect of differentiation respect of UHS compared with conventional sintering	Widely accept mechanisms or explanation	Demonstrated advantage	Open challenge
All material inclusive processing with heating rate 10^3 – 10^4 °C/min up to 3000 °C	Accelerated kinetics driven by ultrahigh temperature	Pressure less sintering of difficult to sinter materials	Narrow processing window suited for optimal microstructural design
Ultrafast heating to decouple kinetics, for example densification vs grain growth	High temperature speeds-up process with high activation energy	Decoupling of concurring kinetics with the purpose of making better performing material	Scalability. Achieve uniform temperature distribution in thick specimens ($\phi > 20$ mm; $h > 5$ mm)
Pressure less and compatible with thin-walled specimens suited for thermodynamic fragile materials	Enhanced sinter activity as surface area is preserved to high temperature	Compatible with thermodynamically fragile compounds (i.e., Li based compounds)	Develop mechanical field assisted sintering techniques while maintaining an ultralow energy consumption
Low cost and highly energy efficient	Uses low-cost heaters and does not require high vacuum	Highly desirable for materials research as 50 to 100 samples can be produced daily	More robust approached to mitigate carbon contamination based on carbon free heaters

Note: The proposed analysis put forward the significance of ultrafast heating rates in combination with high temperatures to explain the additional benefits not observed in conventional firing cycles under a rate of 10 °C/min. Inevitably, because of its novelty, UHS brings many challenges.

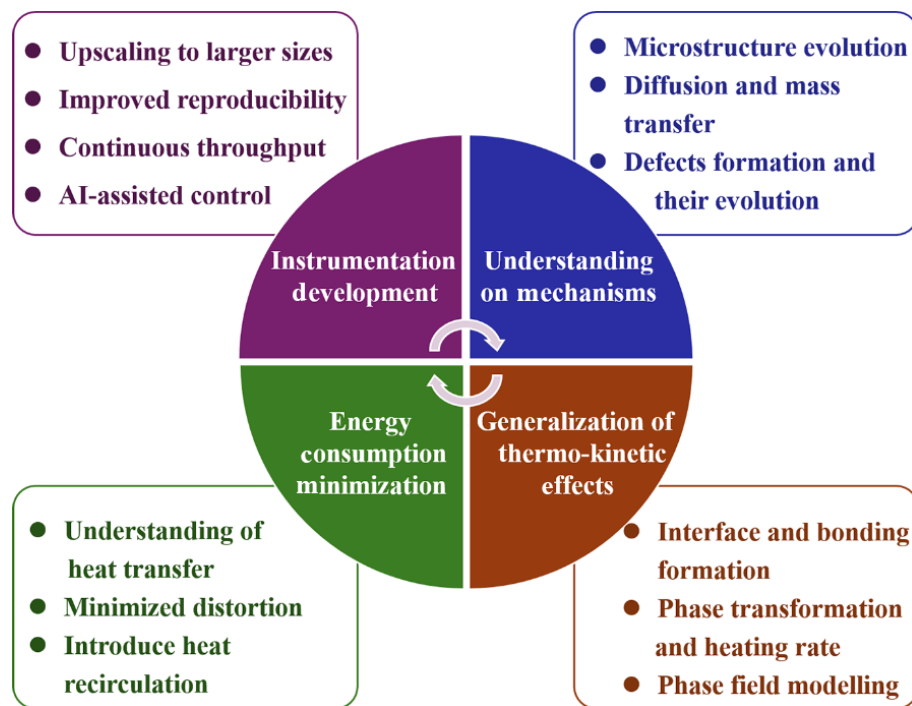


Fig. 53 Outline of UHS future development trends encompassing fundamental understanding of sintering under ultrafast heating rates, generalization of thermokinetic effects, instrumentation development, and heat management.

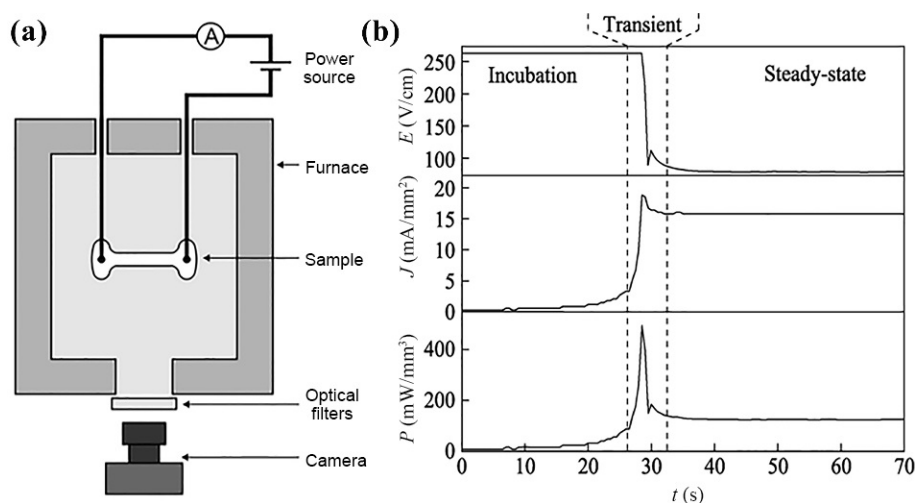


Fig. 54 Introduction to the flash sintering experiment: (a) typical schematic diagram of the flash sintering apparatus [417], (b) the three stages of flash sintering process [418]. Reproduced with permission from Ref. [417] for (a), © Elsevier Ltd. 2013; from Ref. [418] for (b), © Shanghai Institute of Ceramics, Chinese Academy of Sciences 2022.

power dissipation spikes rapidly and then falls just as quickly. This stage is very brief, and the specimen temperature again remains effectively unchanged. In the ensuing steady-state stage, both voltage and current attain relatively stable values, triggering rapid sample heating and achieving full densification of the green body within a few tens of seconds. Since the advent of flash sintering, the variety of ceramic systems processed by this method has grown, experimental protocols have been continuously refined, and the critical onset voltage and temperature for the flash event have steadily decreased—some materials now undergo flash sintering even at ambient temperature [416]. However, to date, no unified mechanism has been established to explain the experimental behaviors observed across different material systems. Room-temperature flash sintering can eliminate the high-temperature furnace conventionally required in ceramic

processing. By connecting the specimen in series with a current-limiting resistor directly to the power supply, the energy consumption is further reduced, and the overall apparatus is significantly simplified.

3.3.2 Flash sintering setup

A typical flash sintering setup is based on the work reported by Cologna *et al.* [13]. The green compact with a dog bone shape is suspended in a furnace by two metal wires with high conductivity, high melting temperature, and high oxidation resistance (platinum is usually used), one attached to each end of the green compact, as shown in Fig. 55(a). A DC or AC power supply is connected to the green compact through the metal wires to apply the electric field [419–422]. To achieve good electrical contact between the green compact and the metal wires, conductive paste

or suspension (typically containing platinum or silver) is always employed. A voltmeter and an ammeter are used to measure the voltage and current of the sample, respectively. In addition, charge coupled device (CCD) cameras and pyrometers are sometimes used to measure sample shrinkage and sample temperature, respectively [423,424].

The dog-bone-shaped green compact is usually used in flash sintering. However, the dog bone shape of the green compact is complex, which limits the application of flash sintering. Therefore, cylindrical pellets and rectangular-shaped green compacts are developed [425–429], as shown in Fig. 55(b). In this case, the green compact is sandwiched between metallic disks or grid electrodes. To ensure close contact between the green compact and the electrodes, a load (P) is usually applied to the electrodes.

3.3.3 Flash sintering process

A flash sintering experiment can be carried out at a constant heating rate or isothermal furnace temperature, as shown in Fig. 56. For the flash sintering experiment at a constant heating rate (Fig. 56(a)), a constant electric field (E) is applied to the sample, and the sample is heated in a furnace at a constant heating rate. Owing to the negative temperature coefficient of resistivity, the current (J) flowing through the sample gradually increases with the furnace temperature. This stage is commonly called stage I or the incubation stage [423,430]. When the furnace temperature rises to a specific value, the current increases rapidly, accompanied by a sudden increase in power dissipation [431], which indicates

the onset of a flash event (rapid densification and intense luminescence). This stage is called stage II. To avoid the thermal runaway of Joule heating, the power supply is quickly switched from voltage control to current control upon reaching a preset current value. Subsequently, the current is kept constant, and flash sintering enters stage III.

Another configuration for flash sintering experiments is carried out at an isothermal furnace temperature, as shown in Fig. 56(b). A constant electric field (E) is applied to the sample, and the furnace is kept at a constant temperature (T). The current (J) progressively increased with time. This stage is also called stage I or the incubation stage [432,433]. After a specific time, the current increases abruptly, and a flash event occurs (stage II). Subsequently, the current is kept constant under current control, flash sintering enters stage III, and the system reaches a new equilibrium.

Recently, two-step flash sintering has been proposed to achieve fast densification with suppressed grain growth [434,435]. In two-step flash sintering, a constant electric field (E) is applied to the sample, and the furnace is kept at a constant temperature (T). When the flash event occurs, the sample is first kept at a higher current (J_1) for a short duration and then kept at a lower current (J_2) for a longer duration, as shown in Fig. 56(c).

3.3.4 Touch-free flash sintering

Normal flash sintering should attach the electrodes to the surface of the green compact. Consequently, the requirement for electrodes limits the geometry of sintered parts. These issues restrict or hinder the commercial application of flash sintering. To address this challenging issue, some touch-free flash sintering techniques have been developed recently. Saunders *et al.* [436] used two arc plasma electrodes to achieve touch-free flash sintering. The plasma flash experimental setup is shown in Fig. 57. Two isolated transformer welders were used to generate two electric arcs, which transfer heat into the sample. The sample was preheated for several seconds, and then the flash power supply was activated to pass current through the sample for a period of time. Pure SiC and B₄C could be rapidly sintered by this touch-free flash sintering technique.

Dong *et al.* [437] reported dielectric barrier discharge atmospheric cold plasma to achieve touch-free flash sintering. The cold plasma could operate at room pressure, and its temperature was much lower than that of the arc plasma, which allowed flash sintering without external preheating. Figure 58 shows a schematic of the cold plasma flash experimental setup.

Jalali *et al.* [438] developed a touch-free flash sintering system with magnetic induction within a reactor activated by the usual flash method. As shown in Fig. 59, the flash event of the dog bone-shaped sample was first induced, and the surfaces of the sample

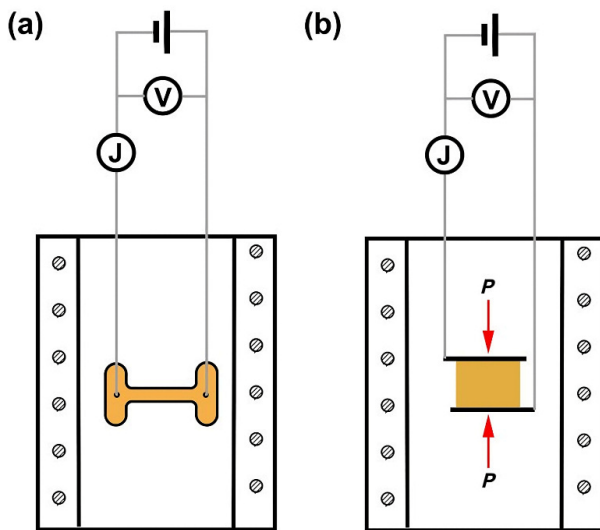


Fig. 55 Schematic illustrations of the setup for flash sintering of a green compact with (a) a dog bone shape and (b) cylindrical pellet.

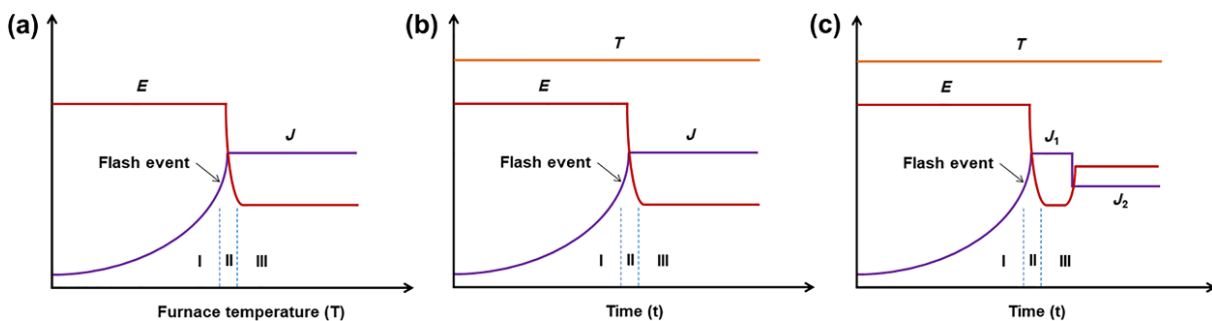


Fig. 56 (a) Electric field (E) and current (J) evolution during flash sintering at a constant heating rate, (b) flash sintering at an isothermal furnace temperature, and (c) two-step flash sintering.

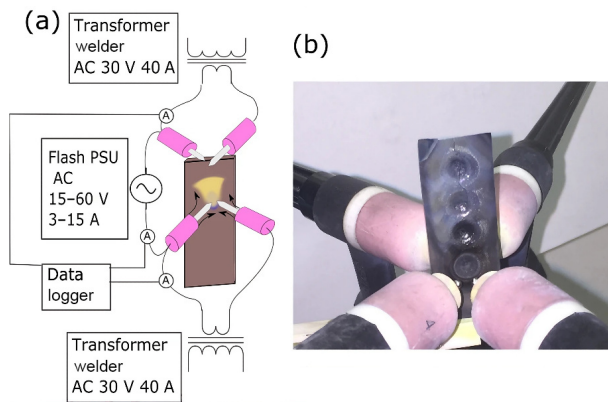


Fig. 57 (a) Plasma flash experimental setup and (b) photo of two twin torches and a processed sample. Reproduced with permission from Ref. [436], © The Author(s) 2016.

acted as the source of the plasma. Then, the plasma was concentrated into a free-standing sample using a magnetic field. Using this touch-free flash sintering system, samples of arbitrary shapes could be flash-sintered.

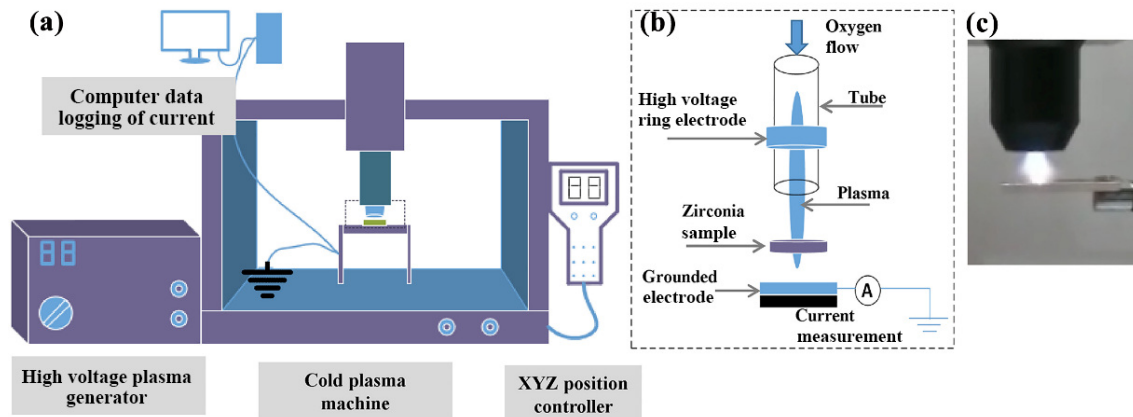


Fig. 58 Schematics of (a) cold plasma setup for touch-free flash sintering and (b) high-voltage plasma electrode and ammeter to measure the current flowing across the sample; (c) photograph of plasma plume during electrical discharge. Reproduced with permission from Ref. [437], © Acta Materialia Inc. 2019.

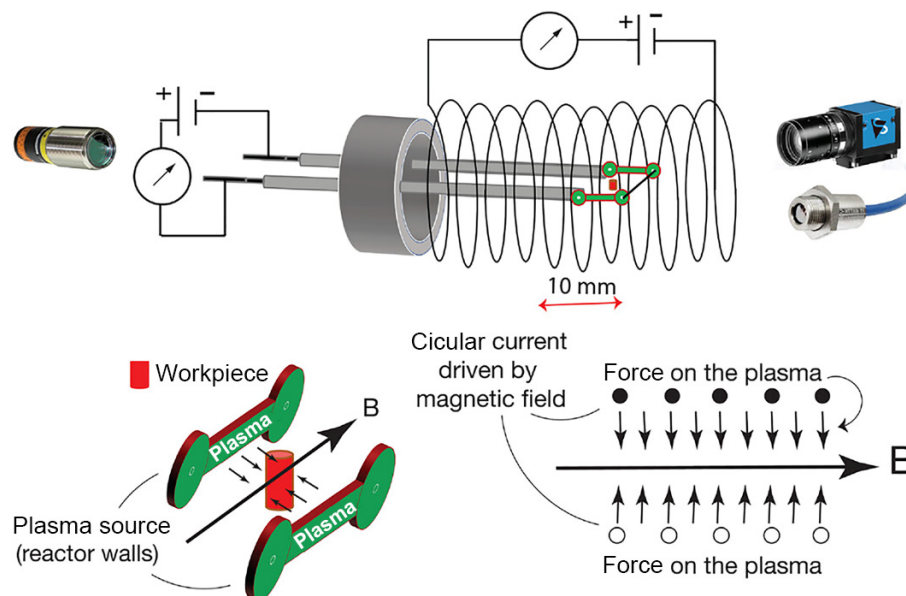


Fig. 59 System configuration for magnetic field-induced flash sintering of a free-standing specimen. Reproduced with permission from Ref. [438], © The American Ceramic Society 2022.

3.3.5 Current status of flash sintering research

(1) Selection of the material system

The electrical conductivity of a material constitutes a critical parameter that governs the ease of flash sintering and dictates the functional relationship between the onset field strength and the initial sintering temperature. To date, flash sintering studies have focused on three broad classes of materials: ionic conductors, semiconductors, and insulators. In addition, reactive flash sintering (RFS) has been demonstrated in a variety of material systems.

(i) Ionic conductors

YSZ, as the first material system subjected to flash sintering, remains the most extensively studied. In YSZ flash-sintering experiments, the effects of AC vs. DC electric fields, sample conductivity, and defect concentrations on the sintering kinetics and final properties were investigated in depth. The surface temperature distribution of 8YSZ specimens during flash sintering under both AC and DC fields was monitored by infrared thermography. It was found that DC fields produced significant thermal inhomogeneity—leading to uneven grain growth—and that electrochemical reactions at the electrodes under DC bias were the root cause of this temperature nonuniformity [439].

Through comparative experiments conducted with and without an applied electric field—and by employing different methods to achieve rapid heating rates—3YSZ ceramics were successfully fabricated, demonstrating that rapid heating is the primary driver of accelerated densification and that the electric field itself does not directly influence this process [440].

The defect concentration within YSZ exerts a strong influence on its electrical conductivity, which is a key determinant of the onset field strength for flash sintering in 3YSZ. Studies have shown that high current densities increase the concentration of oxygen vacancies in YSZ, thereby enhancing its conductivity. This conductivity enhancement is manifested as improved electrical transport behavior during the steady-state stage of flash sintering [440–442]. The increase in defect concentration during flash sintering is not solely attributable to the electric field; *in situ* mechanical analyses reveal that electrothermo-mechanical coupling also generates defects within the green YSZ compact [443]. Consequently, compared to conventional sintering, flash-sintered 8YSZ ceramics exhibit superior ionic conductivity [444]. Room-temperature flash sintering of 3YSZ materials requires an enhancement of the specimen's electrical conductivity, which can be achieved by doping with carbon nanotubes [445] or coating the surface of 3YSZ with iron powder [446], which can facilitate room-temperature flash sintering.

Environmental parameters—including atmosphere composition, the onset temperature for flash sintering, and experimental settings such as current density—also exert a significant influence on the flash-sintering behavior of YSZ. By varying the electric field strength and the gas-pressure conditions, the oxygen-vacancy concentration during the YSZ flash-sintering process was monitored. Higher electric-field intensities promote the formation of oxygen-vacancy defects, whereas elevated oxygen partial pressures inhibit their formation, thereby making flash sintering increasingly difficult to achieve [447,448]. Ong *et al.* [449] investigated the relationship between current density and the densification of sintered YSZ ceramics. Contrary to expectations, the densification increased initially with rising current density but then declined at higher current densities, rather than continuously improving with greater current. It can be seen that an alternating electric field promotes more uniform sintering of 3YSZ. The onset temperature for flash sintering is negatively correlated with the applied field strength. Moreover, the electrical conductivity of the sample depends on its internal defect concentration, which gradually increases during the flash-sintering process.

(ii) Semiconductor

One of the primary research directions in semiconductor ZnO flash sintering is how to lower the furnace temperature and realize room-temperature flash sintering. Low-temperature flash sintering helps reduce energy consumption, refine the grain size of the sintered ceramic, and enhance its properties. Strategies for reducing the flash-sintering onset temperature include increasing the electrical conductivity of the sample and raising the applied electric field. In 2014, Dargatz *et al.* [450] lowered the furnace temperature for ZnO flash sintering from 700 to 625 °C by increasing the DC field from 40 to 160 V/cm. Zhang *et al.* [451] lowered the ZnO flash-sintering furnace temperature to 550 °C by applying a 300 V/cm electric field and further reduced it to below 120 °C using a 500 V/cm field under an Ar atmosphere doped with 5% H₂. The reducing atmosphere enhances the electrical conductivity of the ZnO samples [452]. Nie *et al.* [453] passed an Ar flow

doped with 5% H₂ through a conical flask containing deionized water and directed the resulting humidified gas over ZnO samples, increasing their electrical conductivity by four orders of magnitude. Using only a 200 V/cm electric field, they then achieved room-temperature flash sintering of ZnO.

Ni *et al.* [454] and Mishra *et al.* [455] argued that the Debye temperature constituted the lower bound of the flash-sintering ambient temperature and that the Debye temperature of ZnO at room temperature was approximately 243 °C. The occurrence of room-temperature flash sintering in ZnO contradicts this theoretical framework, as Mishra *et al.* [455] neglected the influence of the sintering atmosphere. Even when the slight reduction in the Debye temperature of ZnO with increasing ambient temperature is taken into account, achieving flash sintering at room temperature by merely increasing the DC field strength remains impracticable without purposeful modification of the sintering atmosphere.

Alternating-current flash sintering is a novel flash-sintering technique in which no electrochemical reactions occur at the electrodes, resulting in uniform densification. In 2019, Wang *et al.* [456] reduced the ZnO flash-sintering furnace temperature to 50 °C by applying a 2800 V/cm AC high electric field and employing a ZnO powder insulation layer. Zhu *et al.* [457] utilized a 3530 V/cm AC electric field to achieve room-temperature flash sintering of ZnO. The excessively high electric field strength required for room-temperature flash sintering hinders its wider adoption. Li *et al.* [458] reduced the field strength needed for ZnO room-temperature flash sintering to 1765 V/cm by employing tip-shaped electrodes. Liu *et al.* [459] applied water droplets to the ZnO surface to enhance its electrical conductivity, thereby reducing the onset field strength to 1628 V/cm. Zhou *et al.* [416] reduced the AC field strength required for room-temperature flash sintering of ZnO to as low as 700 V/cm by employing an atmosphere of 20% air and 80% argon at a total pressure of 20 MPa. Unlike Nie *et al.* [453], who created a humid atmosphere, Liu *et al.* [459] directly hydrated the specimens themselves. Schwarz *et al.* [460] noted that residual moisture within ZnO green bodies during flash sintering promoted densification by enhancing grain rearrangement and mass diffusion processes. Dargatz *et al.* [461] found that during field-assisted sintering, hydrated ZnO green bodies underwent hydroxide-ion diffusion, which lowered the activation energy for densification and generated protonic defects that enhanced electrical conductivity. Based on the promotional effect of water on ZnO densification, Kermani *et al.* [462] proposed a technique termed flash cold sintering (FCP). In their sample preparation, 23 wt% distilled water was added, and the mixture was uniaxially pressed into disks 5 mm in diameter and 3.1–3.4 mm thick. ZnO room-temperature flash sintering was then achieved under a DC electric field as low as 60 V/cm.

Current research on room-temperature flash sintering of ZnO exhibits several limitations: the apparatus developed by Nie *et al.* [453] for generating a humid reducing atmosphere is cumbersome, requires stringent atmospheric control, and involves lengthy preparation time; the methods of Li *et al.* [458] and Liu *et al.* [459] demand high AC field strengths, which hinder scalability and risk sample fracture due to high-temperature arcing and current surges; although tailoring the atmosphere can lower the onset field strength, it imposes specialized equipment and atmospheric requirements; the flash cold sintering technique proposed by Kermani *et al.* [462] depends on a structurally complex apparatus, limiting its broader implementation.

(iii) Insulator

The alumina (Al_2O_3) system is a widely used structural-ceramic material family in which the grain size directly governs the performance of the finished ceramic. Owing to alumina's extremely low electrical conductivity, doping Al_2O_3 ceramics with other metal oxides facilitates flash sintering; moreover, the flash-sintered ceramics exhibit finer grains, thereby enhancing their final properties. Compared with conventional sintering, Al_2O_3 - $\text{Y}_3\text{Al}_5\text{O}_{12}$ eutectic ceramics produced via flash sintering display superior mechanical performance [463]. Al_2O_3 ceramics were fabricated by flash sintering with Co-Mn oxide doping. It was observed that the flash-sintering onset temperature gradually decreased as the electric field strength increased, consistent with the behavior reported for other materials [464]. The team led by Yang at Zhengzhou University of Aeronautical Industry Management conducted a series of studies on the flash sintering of alumina ceramics [465]. By incorporating MgO, they synthesized Al_2O_3 ceramics at 1300 °C and demonstrated that the flash-sintered alumina exhibited a more uniform microstructure [466]. They further investigated the influence of various dopant cations on the flash sintering behavior of Al_2O_3 and found that cation doping introduced additional internal defects during sintering, thereby enhancing the electrical conductivity of the samples [467].

Flash-sintering studies on pure alumina have focused on the onset flash-sintering temperature and the corresponding field strength. Investigations of the Al_2O_3 system indicate that increasing the applied field strength is an effective means of flash-sintering pure alumina. Under a furnace temperature of 1300 °C and an electric field of 4000 V/cm, high-purity alumina ceramics with a relative density exceeding 99% were fabricated [468]. Moreover, by regulating the current rise rate, the onset flash-sintering temperature of alumina was lowered to 1200 °C [469]. The purity of the Al_2O_3 powder dictates its flash-sintering parameters, suggesting that the onset flash-sintering temperature and field strength of alumina ceramics may depend on impurity levels, while both the initial sintering temperature and the nature of the applied field substantially affect the properties of the final product. In one study [470], 99.8%-pure Al_2O_3 ceramics were fabricated at 900 °C under a 1500 V/cm electric field, and it was proposed that the choice of electrode material exerted a significant influence on the onset sintering temperature. Another investigation [471] into the roles of AC vs. DC fields in Al_2O_3 flash sintering revealed that ceramics flash-sintered under an AC field exhibited the highest fracture toughness and hardness; these improvements were attributed to the more uniform temperature distribution under AC conditions and to the generation of defects (e.g., dislocations) during the flash-sintering process.

It is evident that alumina ceramics produced by flash sintering exhibit superior mechanical properties. Consistent with other material systems, the onset flash-sintering temperature of Al_2O_3 decreases as the applied electric field strength increases. Moreover, owing to alumina's intrinsically low electrical conductivity, doping to enhance the overall conductivity or increase the external field strength are commonly adopted strategies for flash sintering. In addition, the choice of electrode material and the concentration of internal oxygen-vacancy defects also critically influence the flash-sintering behavior of alumina.

(iv) Reactive flash sintering

RFS is a novel ceramic sintering process developed from flash sintering technology, first proposed by Perez-Maqueda *et al.* [472] in 2018. They found that BiFeO_3 ceramics can be flash-sintered not only from presynthesized BiFeO_3 powder but also directly from compacts of the precursor oxides Bi_2O_3 and Fe_2O_3 , yielding

dense, single-phase BiFeO_3 . RFS enables synthesis and densification in a single step—eliminating a separate powder-preparation stage—so that multiphase precursors are converted into a single-phase ceramic during sintering. This one-step approach dramatically reduces energy consumption and lowers production costs. It has been reported that RFS of BiFeO_3 ceramics can cut energy use by approximately 84% compared to the conventional route of powder synthesis via solid-state reaction followed by sintering.

At present, RFS is predominantly applied to the synthesis of oxide-based compounds, ceramic composites, and high-entropy oxide ceramic systems. Although RFS has been demonstrated in a variety of ceramic materials, most oxide ceramics still require auxiliary thermal energy and cannot be densified at room temperature, which complicates the sintering apparatus and increases production costs. To date, the only reported instances of room-temperature RFS involve high-entropy oxide ceramics, limiting its broader applicability. Identifying a paradigm for room-temperature RFS of oxide-compound ceramics would therefore represent a paradigm shift in the field. Moreover, existing RFS research has focused almost exclusively on DC electric fields; the behavior of ceramic precursors under high-field AC conditions remains virtually unexplored and merits systematic investigation. Additionally, Jha *et al.* [511] have shown—by studying phase transformations during RFS of the Al_2O_3 - TiO_2 system—that RFS can achieve ultra-rapid densification and may supply energy beyond Joule heating, thereby accelerating reaction kinetics. These phenomena warrant further experimental exploration.

(2) Investigation into the mechanisms of flash sintering

In conventional ceramic sintering, elevated temperature drives sustained mass transport within the green body, expelling pores and ultimately yielding a dense ceramic. In contrast, flash sintering applies a strong electric field directly across the sample, accelerating mass transport and enabling rapid densification. To explain the two hallmark features of flash sintering—ultrafast densification and the precipitous rise in electrical conductivity—researchers have proposed the following mechanisms:

(i) Joule heating effect

This represents the simplest physical mechanism for explaining flash sintering. The ceramic compact can be modeled as a resistor with variable resistance; the passage of current generates Joule heating within the compact, while heat is simultaneously lost to the surroundings. The principal mechanisms proposed for flash sintering are thermal runaway, dielectric breakdown, and the Frenkel defect-pair mechanism. Under a constant current density regime, at the instant of flash initiation, the power input from Joule heating greatly exceeds the power dissipated, inducing thermal runaway. The specimen's temperature then rises rapidly until, at a sufficiently elevated temperature, the input and output powers achieve equilibrium.

(ii) Dielectric breakdown

Dielectric breakdown refers to the phenomenon whereby the application of a strong electric field to a ceramic induces electrical failure, causing the specimen to exhibit conductor-like behavior. Liu *et al.* [459] explained the initiation of ZnO flash sintering under a strong electric field by invoking discharge and dielectric breakdown mechanisms. It has also been observed that during α - Al_2O_3 flash sintering, an originally insulating specimen becomes conductive; based on this phenomenon, the researchers inferred that dielectric breakdown occurred within the ceramic under the applied high electric field [473]. Additionally, Fagnard *et al.* [474] employed a partial-discharge technique to evaluate the electrode/3YSZ specimen contact quality during flash sintering. Their results show that in the 250–500 °C range, under a 50 Hz

AC field ramped to 3200 V/cm, the partial discharge amplitude abruptly increases at the moment of flash onset. Although the applied field remains well below the intrinsic dielectric breakdown strength of 3YSZ, the surface microstructure generated during partial discharges closely mirrors that produced under full dielectric breakdown.

(iii) Frenkel defect pair

Electric-field-induced generation of abundant Frenkel defect pairs accelerates sintering kinetics, resulting in rapid densification during flash sintering at reduced temperatures [2]. Studies have shown that during flash sintering, field-induced point-defect pairs play a crucial role in achieving high densification, lattice expansion, and microstructural gradients in ceramics. The applied electric field promotes ionic lattice disorder, thereby increasing the population of defect pairs (interstitials and vacancies). When the field exceeds a critical threshold, these defect pairs further evolve to generate large concentrations of electrons and holes, resulting in an abrupt rise in electrical conductivity. Under DC bias, neutral defects migrate: interstitials drift toward pore regions, while vacancies become concentrated at grain boundaries [475]. The large atomic displacements measured *in situ* during X-ray scattering flash-sintering experiments indicate that the directionality of lattice vibrations may serve as a potential origin of defect formation [476]. Raj *et al.* [477] further investigated that defect generation may originate from lattice vibrations during power spikes, with the power peak reflecting the phonon energy. Phonon-diffusion molecular dynamics simulations under adiabatic conditions for Frenkel pair formation demonstrate that phonon energy injection drives the system temperature toward a steady-state equal to the Debye temperature. Phonon injection near this Debye temperature is concomitant with the creation of Frenkel defects in the form of vacancy–interstitial pairs, and the attainment of thermal equilibrium is attributed to the absorption of phonon energy into the formation enthalpy of these Frenkel pairs. Naik *et al.* [478] pointed out that following flash-sintering onset, the activation energy for grain growth remained essentially unchanged; however, localized regions exhibited anomalous lattice expansion, likely attributable to an increased internal defect concentration. Schmerbauch *et al.* [479] observed that an applied electric field enhanced grain coarsening in ZnO—an effect that became increasingly pronounced with higher field strength. Muccillo *et al.* [480] similarly reported abnormal grain growth during field-assisted sintering of SnO₂. Su *et al.* [481] further demonstrated that ZrO₂-doped K_{1-x}Na_xNbO₃ (KNN) ceramics generated a high density of defects during flash sintering and that these defects remained in the specimen after the flash treatment.

When the electric field intensity is high, the Joule heating effect, dielectric breakdown, and Frenkel defects do not exist independently during the flash sintering process. Instead, they interact through a positive feedback loop to jointly promote the rapid densification of ceramics. Frenkel defects are the “microstructural foundation” of flash sintering. Their concentration is closely related to the temperature and electric field intensity (the higher the temperature and the stronger the electric field, the higher the defect concentration). The existence of defects alters the electrical and mechanical properties of the material, lowers the breakdown threshold, directly affects the rate and quality of flash sintering, and promotes the occurrence of dielectric breakdown. Dielectric breakdown is the “trigger point” of flash sintering. Whether it occurs depends on the breakdown field strength of the material (related to the material composition and structure) and the electric field intensity. Once breakdown occurs, the local high temperature and current concentration

generated by dielectric breakdown further enhance the Joule heating effect and accelerate atomic diffusion, and the flash sintering process will enter the “rapid densification” stage from the “slow heating” stage. The Joule heating effect is the “energy source” of flash sintering. Its effect directly depends on the current size and duration and is the basis of all flash sintering mechanisms. The Joule heating effect, in turn, promotes the movement and proliferation of Frenkel defects, forming a “defect–breakdown–heat effect” positive feedback loop.

(3) Powder selection

The characteristics of the powder not only exert a critical influence on the properties of the sintered ceramic but also affect the flash-sintering process. To date, flash-sintering studies have scarcely undertaken a systematic investigation of powder properties. The powder preparation route governs defect concentrations, while powder purity, particle size, and morphology control its electrical and thermal conductivities, which in turn modulate flash-sintering behavior—manifested in the varying ease of specimen densification. Therefore, the characterization of powder-system properties should be conducted in parallel with flash-sintering research.

(i) Doping modification

Most ceramic powders exhibit low electrical conductivity. By incorporating conductive phases through mixing, the electrical conductivity of the ceramic can be increased, thereby optimizing its flash-sintering behavior.

Guan *et al.* [482] introduced graphite particles into Sm₂O₃-doped CeO₂ powders and applied a 30 V/cm DC electric field at 25 °C to trigger flash sintering. During subsequent heating, the graphite was gradually oxidized to CO₂ via Joule heating, resulting in a continuous rise in the green body's electrical resistance; concurrently, the Joule heat power generated by the increasing current intensified, thereby driving the densification of the compact. Yang [465] at the Zhengzhou Institute of Aeronautical Industry Management conducted a systematic study of the flash-sintering process in alumina ceramics. Using MgO as a sintering aid, they fabricated alumina samples exhibiting high densification and a more uniform microstructural distribution. They further evaluated the effects of different cation dopants on the field-assisted sintering process and found that doping introduced additional defect sites within the ceramic, thereby significantly enhancing its electrical conductivity [467]. Zapata-Solvas *et al.* [483] introduced 10 wt% of a 1:1 molar-ratio Al₂O₃–Y₂O₃ sintering aid into cylindrical SiC green bodies (5.8 mm diameter, 2.5–3.0 mm height). Under an electric field of 100 V/cm, a current of 15 A, and a furnace temperature of 1170 °C, flash sintering yielded Al₂O₃–Y₂O₃–SiC composite ceramics with a relative density of 88%. Under identical conditions, samples prepared from pure SiC powder reached only 70% relative density. Peng *et al.* [484] employed flash sintering at 850 °C to fabricate SiO₂-doped ZnO-based varistor ceramics and observed that a 2 wt% SiO₂ addition yielded the most favorable overall electrical performance. In the work of Jiang *et al.* [485] on ZnO–Bi₂O₃-based varistors prepared by flash sintering, the authors systematically varied the MnO₂ content in the starting powders. They found that an optimal MnO₂ addition significantly lowered the flash-sintering onset temperature and effectively improved the electrical properties of the resulting varistor ceramics. Wu [486] systematically reviewed the flash-sintering synthesis of ZnO varistor ceramics with different formulations, encompassing both the ZnO–Bi₂O₃–MnO₂ ternary system and multicomponent doped systems. Through side-by-side comparison of doped samples with pure ZnO, they demonstrated that the inclusion of

sintering aids significantly optimized the flash-sintering response and enhanced both the densification and the electrical properties of the ceramics.

(ii) Powder particle size

The reduction in powder particle size increases the internal surface area of the specimen, thereby enhancing surface diffusion rates, lowering the environmental temperature required to initiate flash sintering, and accelerating densification. Francis *et al.* [487] employed tetragonal 3YSZ for flash sintering using powders of four different particle sizes (1, 2, 5, and 10 μm). They observed that as the particle size increased, the onset temperature for flash sintering rose and the final density of the flash-sintered ceramics decreased with larger initial particle diameters, while the power consumption remained essentially constant across all particle sizes.

(iii) Defect engineering

Different defect concentrations can influence the flash-sintering regime: increasing the defect concentration enhances the material's surface conductivity and significantly lowers the critical field strength required to initiate flash sintering.

Wu [486] investigated two postthermal-treatment methods—liquid-nitrogen quenching and hydrogen annealing—for elevating the defect concentration in ZnO green bodies and assessed their effects on flash-sintering behavior. They found that liquid-nitrogen quenching after thermal treatment reduced the band-gap energy of the ZnO specimens and increased their conductivity. The resulting increase in oxygen-vacancy concentration markedly decreases the flash-sintering onset field, owing to a synergistic interaction between oxygen-vacancy migration and internal discharge development that induces thermal runaway and thereby triggers flash sintering.

(4) Electrode selection

(i) Electrode materials

The types and configurations of electrode materials have a profound impact on the flash-sintering process. Electrode materials for flash sintering must maintain both electrical and mechanical stability at elevated temperatures to ensure reliable electrical contact and structural integrity throughout the process. Furthermore, electrode geometry can modify the uniformity of the electric-field distribution, thereby influencing the incubation time for flash initiation and, in room-temperature flash sintering, the discharge behavior. For practical applications, the cost and wear of electrode materials must also be considered.

Most studies still adopt the conventional “metal-wire electrode+dog-bone sample” setup (Fig. 60), in which electrode wires are threaded through end holes in the specimen and suspended within the furnace. Platinum wire is overwhelmingly used: pure Pt offers an extremely high melting point, excellent electrical and thermal conductivity, and moderate hardness and

ductility, making it an ideal flexible connector in this context. There are a few exceptions—for example, Akbari-Fakhrabadi *et al.* [488] employed copper wire as the electrode connection in the flash sintering of Gd-doped ceria electrolytes—but the specific assembly configuration and implementation details were not disclosed.

During flash -sintering experiments on silicon carbide (SiC), Zapata-Solvas *et al.* [483] employed molybdenum wire as the electrode material; however, their apparatus more closely approximated the coaxial graphite-die assembly used in SPS (Fig. 61). In this configuration, the molybdenum wire serves solely to establish electrical continuity between the graphite dies and does not make direct contact with the SiC specimen.

In most flash-sintering studies, the ceramic specimen is placed in direct contact with metal-wire electrodes. A number of investigations, however, have applied conductive pastes at the metal-wire/ceramic interface—forming a coated-electrode layer—to bridge the specimen and electrode wires, thereby improving contact stability and reducing interfacial resistance. Conductive pastes reported in the literature include platinum, silver, carbon, and stainless-steel formulations. Although these experiments differ in sample geometry and electrode configuration, their results remain highly informative given the limited overall dataset.

In the coaxial SPS assembly shown in Fig. 61, pressure is typically exerted on the specimen via a graphite punch. In their SPS investigation of ZrB_2 , González-Julian *et al.* [489] found that allowing the ceramic to make direct contact with the electrically connected graphite electrode plate yielded the highest current density, thereby achieving maximum densification and the coarsest grain growth. However, this configuration also increases the risk of adhesion between the specimen and the graphite punch. To prevent sticking, a thin graphite foil can be inserted between the sample and the punch; although this interlayer effectively inhibits adhesion, it concurrently reduces the current flowing through the specimen. When fabricating solid oxide fuel cell (SOFC) electrolytes in an SPS-type apparatus, the material's extreme sensitivity to carbon and CO_2 contamination precludes the use of graphite interlayers; instead, high-temperature metal foils such as tantalum, molybdenum, or tungsten are employed [490].

A composite layered carbon electrode structure was developed to conduct large-diameter sample flash sintering at room temperature in an air atmosphere under a DC voltage below 100 V (Fig. 62) [491]. The graphite paper is 0.1 mm thick, and the graphite felt is 8.0 mm thick with a porosity of approximately 90%. The graphite paper was precut to match the dimensions of the green bodies, while the graphite felt was cut slightly larger to fully cover the green body. For the sintered sample of ZnO discs

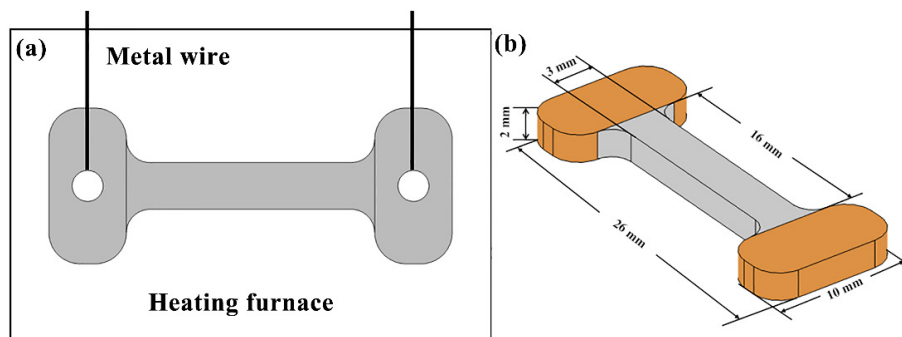


Fig. 60 Schematic diagram of dog-bone-shaped specimen geometry: (a) perforated type; (b) electrode-coated type.

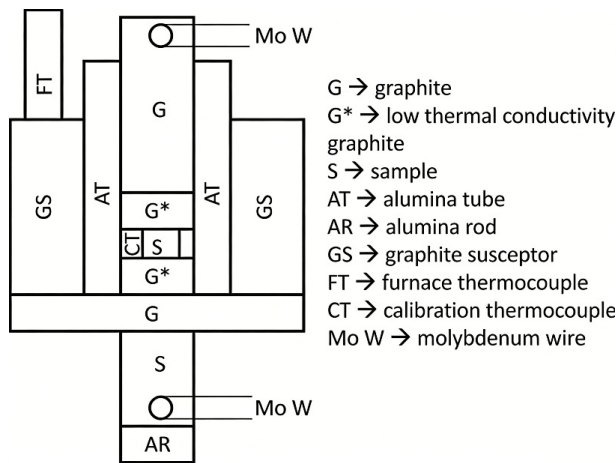


Fig. 61 Apparatus used by Zapata-Solvas *et al.* [483] for SiC flash sintering. Reproduced with permission from Ref. [483], © Elsevier Ltd. 2013.

with a green body diameter of 40.0 mm and a thickness of 1.80 mm (hereinafter referred to as ZnO-d40 discs), upon powering the flash sintering device, intense light was emitted from one edge of the sample. The voltage quickly dropped from 100 V to 50–60 V, and the current increased to the first-level limit of 1 A. Subsequently, the voltage dropped further, and at 6 s, the current reached its final stable value, with the voltage then decreasing to approximately 20 V. The initial intense glow lasted only a few seconds before fading rapidly, leaving only the main body of the sample exhibiting high temperature and dimmer red light.

For disc-shaped specimens, conductive pastes (e.g., platinum or silver pastes) are typically applied to the surface to form coated electrodes. Zhang *et al.* [452] employed magnetron sputtering to deposit platinum thin-film electrodes, whereas Muccillo *et al.* [492] opted for discrete platinum foils or platinum wire meshes and ensured reliable electrical contact by applying mechanical pressure.

Biesuz and Sglavo [470] performed flash-sintering experiments on alumina green compacts under an electric field of 1500 V/cm using platinum paste, silver paste, and carbon paste coatings to establish electrical contact with platinum-wire electrodes. Compared to the platinum paste, the carbon paste reduced the flash-initiation temperature by approximately 100 °C, while the silver paste achieved a reduction of approximately 250 °C. Such pronounced temperature differences cannot be explained solely by enhanced electrode adhesion or lowered contact resistance. A more plausible hypothesis is that carbon and silver catalyze reactions at the alumina–metal interface, thereby increasing the conductivity of the system, whereas chemically inert platinum does not engage in such interactions.

Li *et al.* [458] experimentally compared the performance of different wire electrodes (copper, platinum, molybdenum) in room-temperature flash sintering and found that the copper wire—with the lowest melting point—was most prone to fuse during the flash event. Simultaneously, they evaluated coated electrodes (copper, nickel, silver, and graphite felt) under identical conditions and observed significant differences. Notably, during the power-spike phase at flash initiation, copper-coated electrodes suffered localized discharges and overheating, leading to defect formation and current localization that degraded sample uniformity. In contrast, silver-coated electrodes maintained the most stable current distribution and were identified as the optimal choice for room-temperature flash sintering, whereas graphite-felt coatings failed to trigger the flash-sintering process.

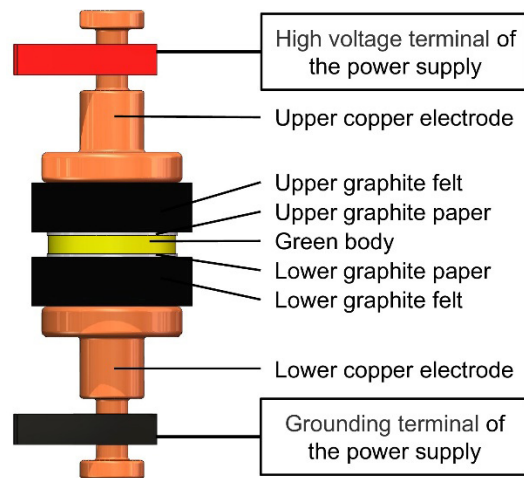


Fig. 62 Diagram of the flash sintering device. Reproduced with permission from Ref. [491], © The Author(s) 2025.

(ii) Electrode configuration

Since Cologna *et al.* [13] first demonstrated successful flash sintering of 3YSZ in 2010, the so-called “dog-bone” specimen geometry—featuring enlarged end sections and a narrowed gauge region—and its corresponding electrode configuration have been widely adopted. This arrangement not only facilitates sample mounting during setup (e.g., by wrapping or suspension) but also establishes a relatively uniform electric field between the enlarged-end electrodes. A wealth of experimental results indicates that this specimen-geometry/electrode pairing markedly enhances both the stability of the flash-sintering process and the postsintering homogeneity of the material. In flash-sintering trials on certain ceramics (e.g., ZnO), specimens frequently fracture extensively as the furnace temperature falls and the applied voltage rises. To prevent such failure, precise control of the flash-onset voltage and initial current density is critical. Once a stable voltage plateau is reached, further elevation of the effective field can be achieved by reducing the interelectrode spacing or deliberately introducing field distortions to create localized high-field zones. Through judicious electrode-structure design, one can lower the onset voltage and temperature thresholds for flash sintering and substantially mitigate sample damage arising from stress concentrations during the process.

Li *et al.* [458] designed two novel electrode architectures—a two-stage configuration and a needle-tip configuration—for room-temperature flash sintering. These designs reduced the flash-trigger voltage from over 6 to 2.77 and 2.53 kV, respectively, and correspondingly lowered the initial current density from above 100 to 55.0 and 53.4 mA/mm². Under both configurations, specimens attained relative densities exceeding 90%. Based on experimental observations, they proposed a surface-tracking flashover-induced flash-sintering mechanism to explain platinum-wire fusing, the transition of light emission from localized spots to the entire specimen, and flash-sintering temperatures that are significantly below the Debye temperature.

(5) An exclusively ambient-temperature process

At present, room-temperature flash sintering has been applied to the fabrication of a wide range of ceramic materials; however, binder burnout still necessitates the use of a conventional sintering furnace. Under the dual-carbon objectives of carbon peaking and neutrality, binder removal techniques—such as dielectric barrier discharge—can enable a completely ambient-temperature ceramic manufacturing process.

Binder burnout aims to remove residual organic binders (e.g.,

polyvinyl alcohol, PVA) and moisture prior to sintering, thereby preventing quality defects—such as deformation, pore formation, surface spalling, and cracking—during subsequent firing. The conventional long-dwell thermal debinding process is simple and well suited to large-scale production, and it remains in widespread use. It relies on the volatilization and thermal decomposition of organic molecules, often employing a two-step protocol to minimize residual carbon within the green body. However, this method involves an inherent trade-off between the debinding rate and final product quality: rapid heating can elevate internal gas pressure in the unsintered body, causing cracks or interlayer delamination, whereas extended dwell time leads to sharply increased energy consumption and reduced throughput. In the face of ever-more stringent environmental regulations and energy conservation imperatives, the conventional debinding process can no longer meet the industry's sustainability requirements.

The application of dielectric barrier discharge (DBD) technology offers a novel solution to the challenges described above. DBD can generate low-temperature plasma directly at ambient temperature; its apparatus is simple and easy to operate, and under atmospheric pressure, it efficiently produces reactive oxidizing species such as atomic oxygen (O), ozone (O₃), and hydroxyl radicals (·OH). These active oxidants not only improve both the surface and bulk properties of the material but also rapidly decompose residual organic binders during debinding, drastically reducing the required thermal input and minimizing volatile organic emissions. In this way, DBD enables a genuinely all-stage, room-temperature flash-sintering process.

Early investigators have employed plasma technologies for cotton fabric desizing, PVA degradation in wastewater, and surface functionalization of PVA films while probing the underlying chemical reaction mechanisms. Since the 1960s, owing to its excellent tensile strength, PVA has been used as a warp-sizing agent in textile manufacturing, forming a durable film on yarn surfaces to ensure superior weaving performance. Cai *et al.* [493] systematically studied the desizing efficacy of atmospheric-pressure plasmas generated from air/O₂/He mixtures on PVA-sized cotton and compared it with conventional H₂O₂ desizing. Their findings showed a marked decrease in C–C and C–H bond populations within the PVA matrix and a concomitant increase in C–O–, C=O, and O–C–O functionalities, leading them to propose a plausible reaction pathway. Wang *et al.* [494] constructed a multi-pin-plate pulsed-discharge plasma reactor and systematically evaluated the influences of initial PVA concentration, pulse peak voltage, air flow rate, solution pH, and iron additive content on PVA degradation efficiency. Zhao *et al.* [495] demonstrated that a 3 min oxygen plasma treatment of vinyl-alcohol (VA) nanofibers produced nanoscale pores of varying depths, yielding a pronounced rough surface; however, extending the treatment to 10 min resulted in diminished interfacial adhesion in the corresponding fiber composites. Cataldo *et al.* [496] elucidated the ozone–PVA reaction mechanism, revealing the introduction of stable trioxatrimethylene-dihydroxy moieties onto the PVA backbone. Finally, Ojah *et al.* [497] investigated atmospheric-pressure DBD plasma modification and assessed its effects on the surface properties and biocompatibility of silk/PVA nanofibers.

DBD technology offers a highly feasible approach for removing organic binders from ceramic green bodies by harnessing the synergistic effects of localized heating and *in situ* generation of reactive species. Moreover, the cold plasma produced by DBD can substantially lower the temperature required for binder burnout, thereby mitigating microstructural damage and providing clear advantages over conventional thermal debinding.

Huang *et al.* [498] established a DBD-based experimental platform for ceramic green-body debinding and employed microscopic analyses to compare specimens treated by DBD with those subjected to conventional thermal debinding, validating the efficacy of the plasma approach. The team then systematically investigated the effects of key process parameters—peak discharge voltage, power-supply frequency, electrode gap width, ceramic/binder formulation, forming method, and sample geometry—on DBD treatment efficiency. Finally, they examined how DBD pretreatment influences subsequent conventional sintering and flash-sintering stages, thereby introducing new strategies and methodologies for ceramic fabrication and performance optimization.

3.3.6 Challenges and prospects

(1) Size effects

Currently, flash sintering remains at the laboratory research stage, and the dimensions of the ceramics produced to date are insufficient for practical applications. Furthermore, increasing the specimen size poses new challenges for the flash-sintering process. Larger parts require higher onset voltages and greater currents, placing more stringent demands on the power supply. Nonuniformities in the current distribution are magnified as the dimensions grow, necessitating precise electric field control to maintain a uniform grain size in the final ceramic. Moreover, when sintering larger green bodies, variations in shrinkage rate during the flash event must be considered; dynamically regulating the external power input throughout the process is a promising approach to mitigate these effects.

(2) Neural network prediction

Since its proposal in 2010, flash sintering technology has undergone more than a decade of development, and experimental results for a wide range of materials have now been compiled into a comprehensive database. Future work can leverage neural network methods to predict, for each material class, the relationships between flash-sintering onset temperature and critical field strength, as well as the required sintering durations, and to evaluate the relative ease or difficulty of flash sintering. By integrating experimental data with machine-learning models, this database can be continuously expanded and refined, thereby providing informed guidance for the continued advancement of flash sintering.

(3) Practical application

Reports on flash sintering applied to actual products are relatively rare. Most focus on the research of mechanisms, but these mechanisms are limited to models such as rapid Joule heating, and there is a lack of exploration of new models. There are also no simulation or mechanism models that can cover the entire flash sintering process. Research on the regulatory influence of electrical/thermal parameters on the microstructure during the flash sintering process is also limited. The density of flash sintering samples is usually not very high, which also limits their progress to the stage of practical application.

4 Challenge and outlook

4.1 Mechanisms of sintering

The study of field-assisted sintering technology faces three major challenges in mechanistic research. First, the mass transport mechanisms under the coupling effects of thermo-electro-mechanical fields remain unclear, particularly the competitive/synergistic relationships between field-induced effects

and conventional diffusion or flow mechanisms, which lack quantitative description. Second, research on nonequilibrium thermodynamics under extreme conditions (e.g., ultrafast high-temperature sintering with heating rates on the order of seconds) lags behind, and existing models struggle to accurately predict microstructural evolution. Third, the complexity of interfacial behavior poses significant challenges, as the response of grain boundaries and phase boundaries under external fields requires further clarification. For example, how can the direct effect of the electric field on grain boundary migration in SPS be quantitatively decoupled?

Future research should focus on multiple simulation methods that are coordinated for use [499–502], such as molecular dynamics, finite elements, and phase fields, to establish quantitative structure-property relationships between field strength, microstructure, and property; advancing *in situ* characterization techniques (e.g., synchrotron X-ray tomography) to enable real-time analysis of defect evolution during multi-field coupling; and building intelligent computational platforms that integrate multisource data through machine learning to achieve accurate prediction and inverse design of sintering processes. These breakthroughs will facilitate a paradigm shift in multi-field coupled sintering from empirical trial-and-error to a model-driven approach, providing a theoretical foundation for the preparation of a new generation of high-performance materials.

4.2 Microstructure tailoring

The novel field-assisted sintering technology faces three major challenges in microstructural control, which significantly limit its precise application in high-performance material fabrication. First, the complexity of multiphysics coupling mechanisms lies in the difficulty of quantitatively decoupling the nonlinear interactions among temperature, mechanical, electrical, and magnetic fields. For instance, during SPS, intricate synergistic or competitive relationships exist among Joule heating effects, electromigration, and mechanical fields. Currently, no comprehensive theoretical model can accurately describe the evolution of atomic diffusion pathways and interfacial dynamics under such multi-field coupling. Second, the uncontrollable nature of cross-scale structural evolution is prominently reflected in the absence of established mapping relationships between macroscopic process parameters (e.g., temperature gradients, pressure distribution) and microstructural features (e.g., grain morphology, grain boundary structure, pore distribution). This often leads to undesirable phenomena during sintering, such as high grain size dispersion, abnormal grain growth, or elemental segregation bands, which severely compromise the consistency of material properties. Third, the lack of process–structure–property correlation models forces material design to rely heavily on empirical trial-and-error. Existing phase-field models [500] and cellular automata methods [501] struggle to integrate multi-field parameters, resulting in generally high prediction errors for microstructural evolution. Moreover, the absence of a digital design platform combining materials genomics and machine learning prevents accurate reverse regulation of “process parameters → microstructure → service performance”. Addressing these challenges requires urgent interdisciplinary collaboration. By advancing *in situ* characterization techniques, multiscale computational simulations, and artificial intelligence algorithms, a precise theoretical framework for microstructural control spanning atomic to macroscopic scales can be established.

The future development of field-assisted sintering technology in microstructural control focuses on three major breakthrough

directions. First, the establishment of a cross-scale intelligent design platform that integrates first-principles calculations (atomic scale), phase-field simulations (micrometer scale), and machine learning (macroscopic process) to build a closed-loop optimization system for “process parameters → field distribution → microstructure”, thereby enabling a digital transformation in materials design. Second, advancing *in situ* characterization techniques through cutting-edge methods such as synchrotron X-ray nano-computed tomography (CT) and environmental TEM can achieve millisecond-level temporal resolution to dynamically track pore shrinkage and grain boundary migration under multi-field coupling, thus revealing the underlying microstructural evolution mechanisms. Third, precise interface engineering technologies should be advanced by utilizing field-assisted methods to tailor grain boundary structures [503,504] combined with solute segregation control (e.g., grain boundary pinning via rare-earth elements) to achieve customized “grain boundary genome” design. Breakthroughs in these directions will collectively drive the transition of field-assisted sintering from an experience-driven to a model-driven paradigm, providing core technological support for the fabrication of a new generation of high-performance materials.

4.3 Practice

Field-assisted sintering technology holds broad prospects for commercial applications. By integrating the synergistic effects of temperature, mechanical, electrical, magnetic, and other physical fields, this technology can significantly reduce sintering temperatures, shorten process cycles, and enhance product performance, bringing revolutionary changes to high-end manufacturing, as shown in Table 13. During industrial adoption, as equipment modular design matures and the cost of key components decreases, the initially high investment is expected to gradually decline. Meanwhile, the establishment of standardized process systems and the application of artificial intelligence [502,505–507] will accelerate process optimization and quality control, promoting widespread use of the technology in high-end sectors such as aerospace, new energy, and electronic devices. For example, Bourges *et al.* [502] proposed a method for the synergic combination of active machine learning and conventional chemical/theoretical approaches, aiming to determine the most effective sintering process and avoid the degradation of the performance of metal sulfide ceramics, thereby reducing the number of experiments. The results showed that the optimization and control of the densification process led to a nonnegligible 60% increase in the thermoelectric coefficient zT of metal sulfide thermoelectric ceramics. Ultimately, multi-field coupled sintering is expected to become one of the core technologies for advanced materials manufacturing, driving the upgrade and development of related industrial chains while generating substantial economic and social benefits.

5 Summary

Field-assisted sintering technology refers to an advanced class of sintering processes that achieve highly efficient densification and precise microstructural control of materials through the synergistic regulation of multiple physical fields, such as temperature, mechanical, electrical, and magnetic fields. By overcoming the limitations of conventional single-temperature-field sintering, field-assisted sintering technologies leverage synergistic effects such as Joule heating, electromigration, and plastic flow. As a result, they significantly reduce sintering temperatures, shorten process cycles, and enable the fabrication of

Table 13 Comparison of field-assisted sintering technology

Sintering technology	Hot oscillatory pressing	Cold sintering	Ultrahigh pressure sintering	Spark plasma sintering	Ultrafast high-temperature sintering	Flash sintering
Key sintering parameters	Temperature, time, median pressure, amplitude, frequency	Temperature, time, pressure, liquid medium	Temperature, time, pressure	Temperature, time, pressure, pulsed current	Temperature, time, heating rate	Critical flash temperature, time, electric field strength, current density
Main advantages	High densification, strong grain boundaries, synergistic performance enhancement	Ultralow energy consumption, suppressed grain growth	Production of ultrahard/metastable materials	Rapid densification, grain refinement, activated sintering	Extreme energy saving, suppressed grain growth, ultrahigh efficiency	Extreme energy saving, second-scale sintering
Applied pressure	Uniaxial oscillatory pressure (typically 20–100 MPa)	Uniaxial static pressure (typically 100–500 MPa)	Ultrahigh pressure (typically 2–10 GPa)	Uniaxial static pressure (typically 20–100 MPa)	Usually, none or very low (< 10 MPa)	Usually, none or very low
Voltage/current	Usually, none	Usually, none	Usually, none	DC pulsed current (kA level), low voltage	DC or AC, high current density	DC/AC, electric field strength is critical (typically 10–1000 V/cm)
Heating rate	Conventional (5–50 °C/min)	Conventional	Conventional	High (up to 100–500 °C/min)	Extremely high (often > 1000 °C/min)	Controlled by electric field, nonlinear surge (up to 10 ⁴ °C/min)
Typical sample size	Medium to large (diameter up to 200 mm)	Medium to large	Limited by high-pressure chamber, usually small	Limited by mold and current uniformity, small to medium (diameter usually < 100 mm)	Limited by rapid heating uniformity, usually thin sheets or small sizes	Limited by electric field uniformity, usually slender strips or thin sheets
Typical applicable materials	Ceramics, metals	Temperature-sensitive materials, ceramic–polymer composites	Ultrahard materials (diamond, cBN), metastable phase materials, some refractory metals	Fine-grained/nanoceramics, cermets, gradient/composite functional materials	Ultrafine-grained/nanoceramics, materials sensitive to grain growth	Ionically conductive ceramics (ZrO ₂ , YSZ), multicomponent oxides

materials with ultrafine grains, high density, and tailored textures or interface structures. This article introduces several field-assisted sintering technologies, including hot oscillatory pressing, cold sintering, high/ultrahigh pressure sintering, spark plasma sintering, ultrafast high-temperature sintering, and flash sintering. The article covers their definitions, equipment, underlying principles, advantages, typical material applications, recent research advances, and future prospects, with the aim of enhancing the understanding of these innovative sintering methods. These emerging technologies are expected to see widespread use in high-performance ceramics, metal-matrix composites, new energy materials, and other high-end applications, representing a core direction for the next generation of green, precise, and intelligent material manufacturing.

(1) Pursuing ultimate performance

Technologies capable of applying high static or dynamic pressure, such as spark plasma sintering or hot oscillatory pressing, should be chosen to simultaneously achieve high densification and microstructural optimization of materials.

(2) Extreme energy efficiency and high productivity

Electric field-dominated technologies such as flash sintering or ultrafast high-temperature sintering are preferred. By leveraging electromigration and the Joule heating effect, they effectively overcome the rate limitations of conventional thermal diffusion, enabling high efficiency and energy savings.

(3) Processing of special materials or structures

For temperature-sensitive materials, cold sintering is the first choice; for synthesizing ultrahard or metastable materials, ultrahigh pressure sintering is essential; for producing ultrafine or

nanocrystalline materials, spark plasma sintering or ultrafast high-temperature sintering can be selected, as their ultrarapid heating characteristics inhibit grain growth.

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Author contributions

This study was completed by the entire team. Jinling Liu designed the structure of the article. Jinling Liu and Dianguang Liu wrote the Abstract and Sections 4 and 5; Dianguang Liu wrote Section 1; Dianguang Liu, Gang Shao, and Jinling Liu wrote Section 2.1; Xiaoqian Tai, Xinyi Li, Sang-Chae Jeon, and Jing Guo wrote Section 2.2; Mengting Lin and Wei Ji wrote Section 2.3; Yan Xiong and Sha Chen wrote Section 3.1; Fei Zuo, Salvatore Grasso, and Hua-Tay Lin wrote Section 3.2; Xilin Wang, Xinghua Su, Ke Ren, Yifei Liu, and Jinling Liu wrote Section 3.3.

Availability of data and materials

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Competing interests

The authors have no competing interests to declare that are relevant to the content of this article. The author Hua-Tay Lin is the Editor-in-Chief of this journal, and Jinling Liu is the Editorial Committee member of this journal.

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