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Review

Research progress and prospects of colored zirconia ceramics: A review

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Abstract: Owing to their rich color, excellent mechanical properties, and favorable biocompatibility, colored zirconia ceramics have been widely used in intelligent terminals, dental restoration, colored decorations, and other potential fields. This paper starts with the challenges faced by colored zirconia ceramics, followed by a summary of the application market of colored zirconia ceramics. Herein, we review various types of colorants and their mechanism of color development, summarize coloring methods, and analyze their advantages and disadvantages. Finally, the research progress on zirconia ceramics with red, blue, black, and other common colors is summarized, and future development directions are proposed in this review.

Keywords: colored zirconia ceramics; applications; fabrication; color performance; mechanical properties

1 Introduction

As a pivotal ceramic material, zirconia (ZrO_2) boasts exceptional mechanical properties, chemical stability, and biocompatibility, making it extensively utilized in applications ranging from high-temperature structural components to decorative and biomedical materials [1–3]. Despite these advantages, traditional zirconia ceramics are predominantly white or off-white, which imposes limitations on their aesthetic versatility and design flexibility, particularly in light of the growing consumer demand for visually appealing products and the ongoing evolution of lifestyle trends. The emergence of colored zirconia ceramics marks a significant breakthrough in addressing these limitations [4,5]. This new class of ceramics retains all the desirable mechanical properties of traditional zirconia while offering a broader spectrum of vivid colors that are resistant to fading over time. Additionally, the metallic-like luster of these colored zirconia ceramics adds a unique aesthetic appeal, while their non-toxicity and biocompatibility ensure safety for a range of applications [6,7]. These features collectively position colored zirconia ceramics as premium materials for high-end decorative items, mobile phone backplates, smart wearable device casings, and jewelry, among other applications [8,9].

This paper offers a comprehensive review of the advancements in colored zirconia ceramics, focusing on pivotal developments and innovative applications within this domain, and the principal coloring methods and underlying color development mechanisms are summarized. Specifically, the current state of research concerning red, blue, and black zirconia ceramics is scrutinized, encompassing an analysis of ongoing challenges and proposed future research directions aimed at broadening the application scope and enhancing the performance of colored zirconia ceramics. The primary objective of this review is to provide researchers and industry stakeholders with a thorough understanding of the present status and prospective avenues within the realm of colored zirconia ceramics.

1.1 Zirconia materials

Zirconia is an indispensable structural and functional material with excellent physical and chemical properties [10,11]. By virtue of its high mechanical properties, outstanding corrosion resistance and high-temperature resistance, good biocompatibility, and non-toxicity to the human body [12–14], zirconia materials are widely used in various fields, such as decorative materials, grinding media, refractory materials, electronic components, biological materials, and so on [15–21].

Pure ZrO_2 is a polymorphic material that develops in three crystallographic forms at different temperatures: monoclinic (m-ZrO_2) at ambient conditions, and tetragonal (t-ZrO_2) and cubic (c-ZrO_2) at higher temperatures [22–25]. The crystal structures of the three crystallographic forms are presented in Fig. 1 [26]. The red ball represents the zirconium atom, and the other colored balls represent the oxygen atom. The m-ZrO_2 is stable from room temperature to 1170 °C. Between this temperature and 2370 °C, t-ZrO_2 is formed, while c-ZrO_2 is formed at temperatures above 2370 °C up to the melting point of 2680 °C [27–30].

In 1959, McCullough and Trueblood [31] were the first to describe the cell structure of m-ZrO_2 successfully. The crystal structure of monoclinic ZrO_2 is a distorted form of a fluorite structure with Zr in eight-fold coordination. In the monoclinic ZrO_2 molecule, the coordination number of Zr is seven, and all Zr–O bond lengths are not equivalent. The unit-cell dimensions are $a = 0.5169$ nm, $b = 0.5232$ nm, $c = 0.5341$ nm (all ±0.0008 nm), and $\beta = 99°15'\pm10'$.

In 1962, Teufer [32] determined the crystal structure of

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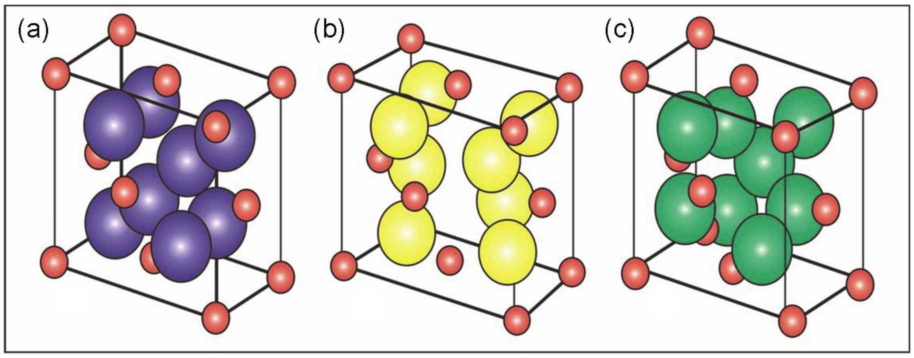

Fig. 1 Crystallographic forms of zirconia: (a) monoclinic, (b) tetragonal, and (c) cubic. Reproduced with permission from Ref. [26], © Academia 2024.

tetragonal ZrO_2 by high-temperature X-ray diffraction analysis, and its single-cell constants are $a = 0.364$ nm and $c = 0.527$ nm. In the tetragonal ZrO_2 molecule, the coordination number of Zr is eight, and each Zr^{4+} ion is surrounded by eight O^{2-} ions: four O^{2-} ions at a distance of 0.2065 nm in a flattened tetrahedron and four other O^{2-} ions at 0.2455 nm in an elongated tetrahedron, which is rotated 90° relative to the former.

In 1962, Smith and Cline [33] first discovered the transition of zirconia from the tetragonal to cubic phase and confirmed the existence of cubic ZrO_2 at high temperatures with a phase transition temperature range of 2285±50 °C via high-temperature X-ray diffraction analysis. Cubic zirconia has a fluorite structure, and in the cubic ZrO_2 molecule, each Zr^{4+} ion is coordinated with eight equidistant O^{2-}ions, and each O^{2-} ion is coordinated with four Zr^{4+} ions in tetrahedral coordination. The single-cell constant is $a = b = c = 0.508$ nm.

Because the phase transformation process is reversible and the density of the three crystallographic forms of zirconia is different, volume change will occur during the phase transformation process. The transition from monoclinic to tetragonal zirconia has a volume contraction of 7%–9%, while the reverse transition is accompanied by a volume expansion of 3%–5% [34]. During the phase transformation between the monoclinic phase and the tetragonal and cubic phases above 1170 °C, the associated volume change will create cracks in the sample structure [35,36]. To solve this problem, t-ZrO_2 and c-ZrO_2 can be stabilized at room temperature by doping ZrO_2 with a certain amount of stabilizing oxides, such as yttrium oxide (Y_2O_3), cerium oxide (CeO_2), calcium oxide (CaO) or magnesium oxide (MgO), called stabilized zirconia [37–42]. The stability properties resulting from different doping stabilizers are not the same, and the different contents of the same stabilizer also affect the performance of doped zirconia ceramics. Li *et al.* [43] summarized the research status of stabilizers for zirconia ceramics in China and abroad. It is suggested that material computation can be used to assist the screening of stabilizers to shorten the experimental period and reduce the cost. In 1975, Garvie *et al.* [44] in Australia prepared stable t-ZrO_2 ceramics using CaO as a stabilizer and first used ZrO_2 to improve the toughness and strength of ceramics by the martensitic phase transition process of quadrilateral monoclinic phase. Among these oxide stabilizers, Y_2O_3 is the most commonly used because yttrium ion (Y^{3+}) has high solid solubility in the zirconia lattice [38]. When Y_2O_3 is added to ZrO_2 as a stabilizer, Y^{3+} replaces Zr^{4+} sites, generating oxygen vacancies to achieve charge balance. A positively charged oxygen vacancy causes a displacement of O^{2-} in the lattice, thereby altering the crystal structure of ZrO_2 [45–49]. Depending on the content of Y_2O_3, different yttria-stabilized zirconia (YSZ) materials can be obtained that maintain phase stability at room temperature [50–52]. When the doping amount of Y_2O_3 is 3 mol%, zirconia ceramics with a crystal structure of t-ZrO_2 can be obtained, called 3YSZ. Owing to phase transformation toughening, the bending strength of t-ZrO_2 ceramics is much higher than that of other ceramic materials, reaching 1100 MPa [53,54]. When the doping amount of Y_2O_3 is 8 mol%, the crystal structure is c-ZrO_2, known as 8YSZ, and polycrystalline ceramics with this crystal structure have no birefringence effect at the grain boundaries, which can be used to prepare transparent ceramics [55–57]. The mechanical properties of c-ZrO_2 ceramics are similar to those of t-ZrO_2 ceramics, except that the fracture toughness is lower than that of t-ZrO_2 ceramics [58].

1.2 Colored zirconia ceramic materials

As color is an important feature in fields of electronics, jewelry, decoration, and other applications, the investigation of color in zirconia materials is important [59,60]. Since the monotonic color of zirconia ceramics has limited their application, many researchers have begun to develop zirconia with various colors to expand the application range of zirconia ceramic materials. With in-depth study of zirconia ceramics, in addition to white ceramics, other colors of ceramics, such as black, red, blue, yellow, green, and other various hues of colored ceramics, have also been developed, greatly enriching high-end decorations, mobile phone backplanes, jewelry and other industries [61–63]. In recent decades, the applications of colored zirconia ceramic materials have extended to smart wearable materials, dental restorations, color decorations, and other potential fields, which are attributed to the brilliant luster, outstanding mechanical and optical properties, and favorable biocompatibility of zirconia ceramics [64–67].

In the study of colored zirconia ceramics, colorants are usually added to the ceramic body or glaze, and the sintering temperature is generally approximately 1300 °C. However, the sintering densification temperature of zirconia ceramics is generally 1550–1650 °C. At high temperatures, colorants may decompose or volatilize due to instability during the sintering process, leading to alterations in the color spectrum of zirconia ceramics. Consequently, it becomes challenging to synthesize colored zirconia ceramics with bright and uniform colors [68]. To solve this problem, on one hand, it is necessary to reduce the sintering temperature by adding an appropriate amount of sintering additives to inhibit the decomposition and volatilization of colorants. On the other hand, the type of colorant and the adding process of colorant will affect the color and mechanical properties of colored zirconia ceramics, so it is also crucial to choose the appropriate colorant and synthesis method, which has important theoretical significance and market application value. At present, colored zirconia ceramics still have some problems, such as poor color saturation, high-temperature stability, uneven coloring, and insufficient mechanical properties, so the development of new colored zirconia ceramic materials still has broad market prospects.

1.3 Applications of colored zirconia ceramics

1.3.1 *Communication field*

In recent years, with the rapid development of 5G communication technology, the shell material of smart wearable electronic devices has become a growing application field of colored zirconia ceramics [69,70]. Compared with traditional engineering plastics, metals, and other shell materials, zirconia ceramics featuring greater wear resistance, better scratch resistance, stronger aesthetic texture, and insulation characteristics are especially useful for providing smaller shielding above the 3 GHz millimeter wave signal used in 5G communication and easy wireless charging [71–73]. The Mohs hardness of zirconia ceramic materials can reach 8.5, which is close to that of sapphire and far exceeds those of metal and tempered glass materials. Moreover, zirconia ceramics have the advantages of electrical signal sensitivity, no electromagnetic interference, no magnetism, and strong signal receiving ability, so they are the best choice for mobile phone backplanes in the 5G era [74,75]. In addition, zirconia ceramic materials have good plasticity and can be modified to produce a variety of shapes of mobile phone backplanes, so through the fine surface polishing process, they can be made to have a glossy mirror texture [76,77]. Therefore, colored zirconia ceramic materials combine excellent mechanical properties and aesthetic characteristics and have great application prospects in the fields of mobile phone backplanes and other smart wearable appearance parts. Today, mass production companies such as Chaozhou Sanhuan (Group) Co., Ltd., Shandong Sinocera Functional Materials Co., Ltd., and Tosoh Corporation have developed more than 30 colors of zirconia ceramic materials. At present, zirconia ceramic backplanes have been successfully applied in various mobile phone models produced by Huawei, Xiaomi, OPPO, LG, Samsung, and other brands.

1.3.2 *Advanced decorations field*

Zirconia ceramics have high wear resistance, corrosion resistance, excellent metallic luster and texture, and no allergic effects on the human body, and can be polished and processed to obtain a variety of exquisite decorations, such as watch dials, watch chains and gem jewelry [78]. Compared with traditional decorative silicate ceramic materials, such as ceramic vases and ceramic floor tiles, zirconia ceramic materials have a brighter luster and better mechanical properties. In the past, single crystals of cubic yttria-stabilized zirconia have become interesting materials for application as diamonds because of their high refractive index of 2.2 and high hardness [24,79]. Owing to the high refractive index of c-ZrO_2, it can be made into transparent and colored zirconia decorative materials, and its Mohs hardness can be as high as 8.5, similar to that of sapphire, with a good luster effect [80,81]. Synthetic c-ZrO_2 has a gem-like luster and low price, and it can replace diamonds and other jewelry. In addition, some smart wearable decorative materials, such as watch dials and chains, are also made of zirconia ceramics [82,83]. For example, international brands such as Chanel and RADO have used zirconia ceramics for their watch shell materials [84].

1.3.3 *Biomaterial field*

Zirconia ceramics are bioinert materials that have stable chemical properties, good biocompatibility, high surface gloss, and natural tooth color and are widely used in human hard tissue repair materials [85–90]. Zirconia dental ceramics exhibit sufficient mechanical strength and toughness to allow their use in low-load areas of the mouth for longer restoration lifetimes [91–94]. Natural-looking dental restorations require appropriate material selection, form, surface, texture, translucency, and color [95–97]. Zirconia-based dental materials are extensively used in clinical practice because of their tooth-like appearance, bio-functionality, bio-compatibility, and affordability [98]. To date, colored zirconia ceramics, including dental crown bridge repair materials, zirconia post core materials, dental implant materials, and bracket materials, have been widely used as dental biomaterials for several decades [99–104]. The use of ZrO_2 for medical purposes was first proposed in 1969 as a new material for hip head replacement instead of titanium or alumina prostheses [105]. At present, a variety of zirconia dental restorative materials, such as Vita In-Ceram, Procera Allceram, IPS·Empress2, and GI·II, have been put into clinical use [106–108].

Currently, biomedical-grade zirconia usually contains 3 mol% Y_2O_3 as a stabilizer called 3Y-TZP or 3YSZ, which has superior mechanical properties, as its flexural strength reaches 900–1200 MPa, and its fracture toughness is approximately 9–10 MPa·m$^{1/2}$ [109–111]. Therefore, it can be used in dentistry for the fabrication of dental crowns and fixed partial dentures. As early as 2002, Ardlin [112] studied the relationship between the strength and surface structure of Denzir™ yellow and white zirconia ceramics and their low-temperature stability and chemical stability and reported that the bending strength of yellow zirconia before and after aging was greater than that of white zirconia ceramics, which can be used as dental inlays, crowns, and bridge materials. In 2007, Studarta *et al.* [113] used 3YSZ, Al_2O_3·ZrO_2 glass complexes, and Li_2O·$2SiO_2$ glass ceramics to conduct rapid fracture tests and fatigue tests on various dental restorations. The results show that the service life of zirconia ceramic materials can reach more than 20 years, and the cyclic fatigue resistance of the two zirconia ceramic materials is greater than that of Li_2O·$2SiO_2$ glass ceramics in the presence of water, which is especially suitable for the preparation of all-porcelain crown bridge prostheses of oral posterior teeth. In 2013, Li *et al.* [114] developed a zirconia ceramic tray that can meet the needs of clinical bonding in orthodontics after being sandblasted and polished and does not easily damage tooth enamel when it is removed. However, natural teeth are not as white as pure 3YSZ ceramics and require slight coloring of dental zirconia ceramics [115]. One way to obtain tooth-colored zirconia ceramics is to simply apply a colored veneering layer, but it often falls off on the ceramic crown and is not conducive to dental restoration [116,117]. Clinically, dental colored zirconia ceramics are prepared mainly by immersion of the zirconia green body with a coloring liquid or by adding a colorant additive or dopant to the powder mixture, such as iron and rare earth cations [118,119].

In 2018, Wang *et al.* [120] prepared colored 3YSZ ceramics by adding different mass fractions of oxide coloring powders of Fe_2O_3, Pr_6O_{11}, Er_2O_3, and CeO_2. To ensure that the ceramic has a certain mechanical strength, the oxide addition range, which is most consistent with the tooth color of Chinese people, was optimized. Figure 2 shows the chromaticity values a^* and b^* of 3YSZ ceramics with different added oxide colorants (the coordinates of the International Commission on Illumination (CIE) 1976 $L^*a^*b^*$ color space, $L^* = 0$ indicates black, and $L^* = 100$ indicates white; $+a^*$ = red, and $-a^*$ = green; $+b^*$ = yellow, and $-b^*$ = blue [121]) and matching color range of the Chinese tooth Vita three-dimensional (3D) color plate. In addition, the flexural

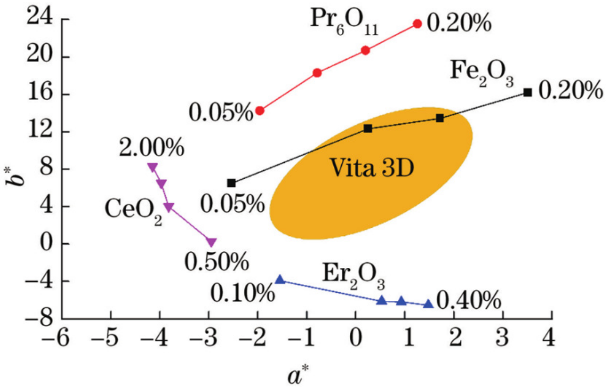

Fig. 2 Effects of different oxide colorants on a^* and b^* values of ceramics and color range of Vita 3D color plates. Reproduced with permission from Ref. [120], © China Academic Journal Electronic Publishing House 1994–2022.

strength of the ceramics decreased with increasing addition of colorant but remained above 800 MPa, meeting the requirements of clinical dental restoration materials.

In 2018, Holz *et al.* [122] reported that the addition of a low concentration of Fe_2O_3 slightly reduced the fracture toughness of 3YSZ ceramics but had no significant influence on the Vickers hardness (> 12 GPa) and biaxial flexural strength (> 1000 MPa). From the color point of view, the Fe:3YSZ ceramics are chromatically stable under different atmospheres and different ranges of temperatures. Kao *et al.* [123] investigated the impact of Fe_2O_3 coloring agent on the sintering behavior of 3YSZ ceramics. The color of sintered 3YSZ ceramics was ivory and became more and more red with increasing Fe content, which fulfilled the requirements for applications in dentistry, as demonstrated in Fig. 3. The size of the grains increased, and more monoclinic phases were found in the ceramics with increasing Fe content. However, the formation of a monoclinic phase was not conducive to the hardness and toughness of ceramics, which limited the amount of colorant added.

In 2019, Willems *et al.* [124] demonstrated that doping small amounts (0.01–0.1 mol%) of Fe_2O_3 as a tooth-colored zirconia ceramic was a promising strategy. The hardness (~13 GPa) and fracture toughness (~3.6 MPa $m^{1/2}$) of the 0.01–0.1 mol% Fe_2O_3-doped 3YSZ were comparable, whereas higher Fe_2O_3 doping amounts (0.5–2 mol%) not only revealed a color that cannot be applied for dental restorations but also led to a strong increase in large-grained cubic phase formation and residual porosity, followed by a decrease in density and mechanical properties, as shown in Fig. 4. In addition, the hydrothermal ageing resistance slightly increased with increasing Fe_2O_3 concentration up to 1 mol%, whereas the translucency slightly decreased with increasing the Fe_2O_3 content.

2 Fabrication and mechanism study of colored zirconia ceramics

2.1 Types of colorants

By adding different types of colorants, zirconia ceramics can exhibit different colors, and the process of adding colorants will also affect the performance of colored zirconia ceramics. At present, zirconia ceramic colorants include mainly transition metal oxides, rare earth ion oxides, and spinel-type oxides [125,126].

Transition metal oxides and rare earth ion oxides colorants have the characteristics of strong coloring ability, stable coloring, simple manufacturing process, and easy operation, and have been widely used in the field of colored zirconia ceramics [127]. Huang *et al.* [128] prepared pink, light yellow, yellow, brown, and black 3YSZ ceramics by adding rare earth ion oxide colorants (Er_2O_3, CeO_2, and Pr_6O_{11}) and transition metal oxides (Fe_2O_3 and MnO_2), respectively. The results show that CeO_2 and Pr_6O_{11} affect mainly the b^* values of the ceramics; Er_2O_3 affects mainly the a^* value; Fe_2O_3 affects the L^*, a^*, and b^* values; MnO_2 affects mainly the

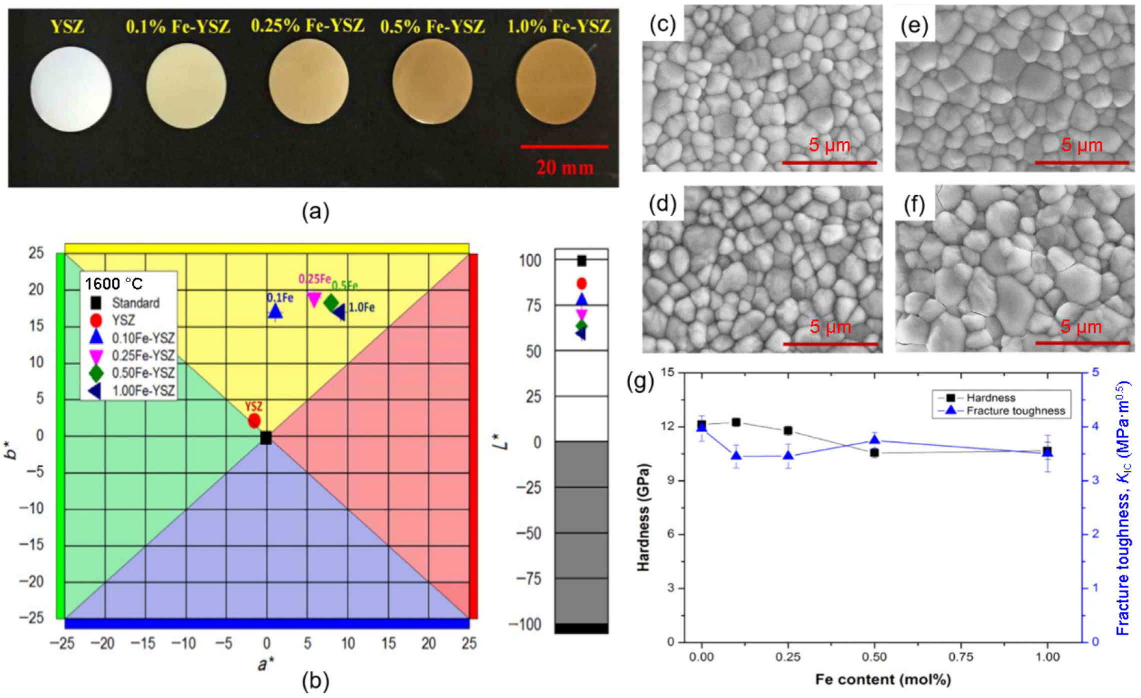

Fig. 3 (a) Color and (b) spectra of polished sections of Fe-doped YSZ samples after sintering at 1600 °C for 2 h. L^* is white–black level; a^* is red–green index; b^* is yellow–blue index. SEM micrographs of (c) YSZ, (d) 0.25% Fe-doped YSZ, (e) 1.0% Fe-doped YSZ, and (f) 3.0% Fe-doped YSZ samples after sintering at 1600 °C. (g) Hardness and toughness of sintered Fe-doped YSZ samples. Reproduced with permission from Ref. [123], © Elsevier Ltd. and Techna Group S.r.l. 2017.

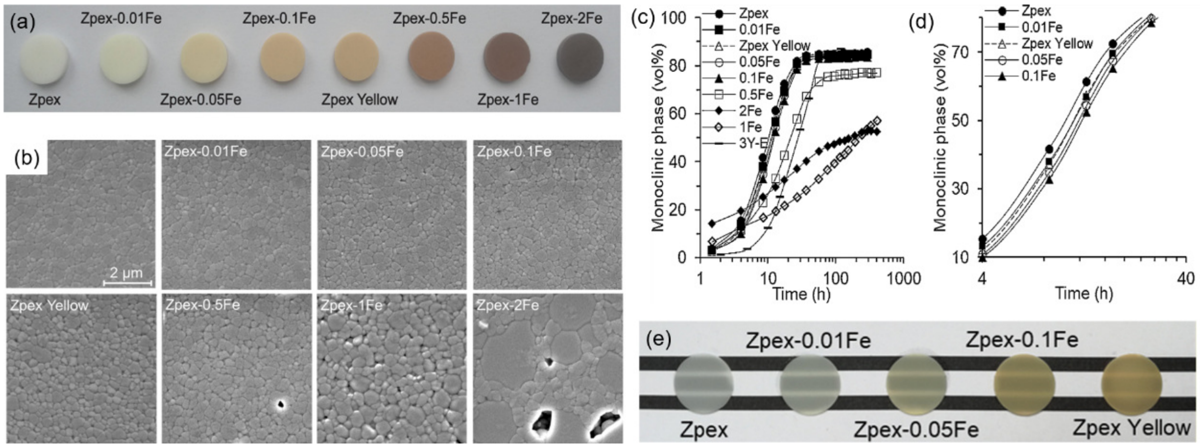

Fig. 4 (a) Photographs and (b) SEM micrographs of Fe_2O_3-doped Zpex (a nanosized co-precipitated 3YSZ powder with 0.05 wt% Al_2O_3 dopant) powder-based zirconia ceramics. Ageing curves of white (Fe_2O_3-free), and colored Zpex zirconia ceramics sintered for 2 h at 1450 °C. (c) Overview of ageing curves and comparison to TZ-3Y-E. (d) Close-up of Zpex, Zpex Yellow, Zpex-0.01Fe, Zpex-0.05Fe, and Zpex-0.1Fe. (e) Translucency of colored zirconia ceramics. Reproduced with permission from Ref. [124], © Elsevier Ltd. 2018.

brightness. Zhao *et al.* [129] introduced colored ions into 3YSZ bodies through diffusion to prepare black (Fe^{3+}, Co^{2+}, Cr^{3+}, and Al^{3+}), blue (Co^{2+} and Al^{3+}), pink (Er^{3+}), and green (Ni^{2+} and Al^{3+}) 3YSZ ceramics by the liquid-phase precursor infiltration method, as shown in Fig. 5. In addition, the mechanical property analysis revealed that the colored 3YSZ ceramics still had high strength and toughness after coloring. In 2019, Lv *et al.* [130] adopted a solid phase mixing method and used Cr_2O_3 and NiO as colorants to obtain high-toughness light green and dark green 3YSZ ceramics with stable structures and excellent performance, respectively. To achieve a specific color, in addition to doping certain colored ions alone, two or more kinds of colored ions can also be doped at the same time to regulate the color of ceramics. In 2010, Shen *et al.* [131] grew emerald-like c-ZrO_2 crystals with Pr^{3+}, Co^{3+}, and Cr^{3+} as the main doped ions by using skull crucible technology through spectral analysis of natural emeralds. Further research by the team showed that the width and intensity of the characteristic absorption peaks can be adjusted by changing the kinds and concentrations of the doped coloring ions so that the color of emerald-like c-ZrO_2 crystals can be closer to that of natural emerald [79].

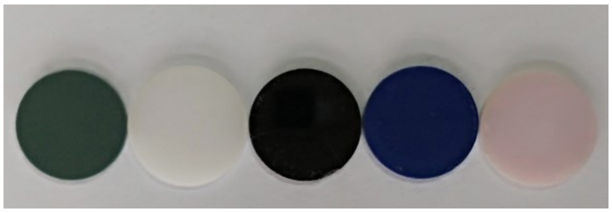

Fig. 5 Photograph of colored zirconia ceramics. Reproduced with permission from Ref. [129], © Rare Metal Materials and Engineering 2018.

Spinel compounds are traditional colorants with a long history. Sickafus *et al.* [132] summarized the crystal structures of compounds with the general formula AB_2X_4, which crystallizes with the same atomic structure as the mineral spinel $MgAl_2O_4$. The spinel-type colorant is a multi-component compound synthesized from transition metal oxides. Owing to its stable crystal structure, the ceramic or glaze layer can maintain the stability of color during high-temperature sintering [133]. For example, the colorant powders of Fe_2O_3, Cr_2O_3, and MnO were mixed in a certain proportion and then sintered at high temperature to synthesize three different types of spinel solid solutions [134]. Among them, $MnO{\cdot}Fe_2O_3$, $MnO{\cdot}Cr_2O_3$, and $FeO{\cdot}Cr_2O_3$ spinels were dark red, dark green, and dark brown, respectively. If the contents of these three metal ions can be adjusted to the right proportion, their spectral absorption curves can complement each other so that all the spectra in the visible light range will be absorbed and the spinels are bright black. In 2020, Xiong *et al.* [135] synthesized the sky-blue pigment $NiO{\cdot}nAl_2O_3$ with a spinel crystal structure using Al_2O_3 and NiO as the raw materials and prepared sky-blue aesthetic 3YSZ ceramics by solid phase method, as shown in Fig. 6. The pigment achieved the optimal chroma by calcination at 1450 °C with an Al/Ni molar ratio of 3. In addition, the prepared ceramics doped with 2.5 wt% sintered at 1450 °C achieved satisfactory comprehensive performance, with bending strengths and blueness values b^* of 964±37 MPa and −24.5, respectively.

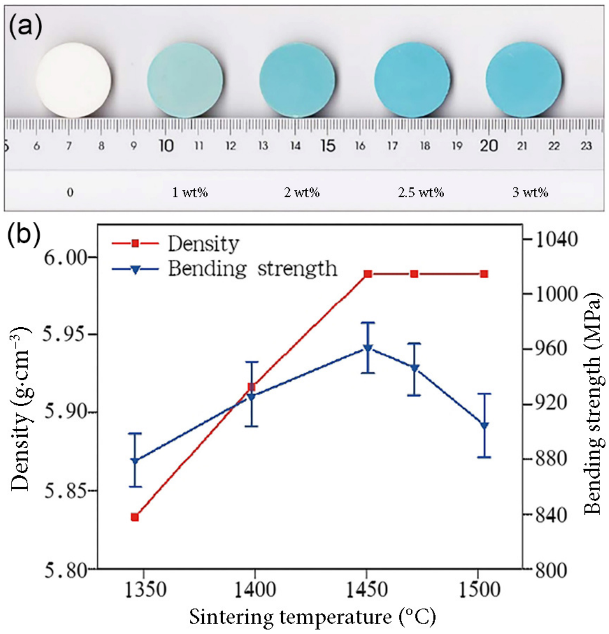

Fig. 6 (a) Photographs of sky-blue zirconia ceramics doped with different contents of pigments sintered at 1450 °C for 2 h. (b) Density and bending strength of zirconia ceramics with 2.5 wt% pigment sintered at different temperatures. Reproduced with permission from Ref. [135], © China Ceramics 2020.

2.2 Mechanism of color development

From a physical point of view, the production of color is a certain wavelength range of electromagnetic radiation. When electromagnetic radiation stimulates the visual nerves of the human eye, different colors can be produced [136]. The mechanism of color development in zirconia ceramics is mainly due to the electronic transition process and the selective absorption and reflection of colored ions under the action of visible light [137]. The color of ceramics mainly depends on the structure, ionic radius, valence, coordination number, and mutual pole of the colored ions, and the preparation method and synthesis conditions can also affect their color.

Transition metal ions have a $4s^{1-2}3d^{x}$ electronic structure, and rare earth ions have a $6s^{1-2}5d^{1-8}4f^{x}$ special electronic structure, whose outermost layer is not filled with electrons. These unfilled electrons are extremely unstable and easily transition between the suborbital orbits of each layer, and the energy required for the transition is exactly the energy required for photons in the visible light region [138]. For different elements, the energy-level discrepancy (ΔE) between the various sublayers is not equal, resulting in the corresponding wavelength of monochromatic light being selectively absorbed and reflected, thus resulting in different colors [139].

Transition metal ions have unsaturated electrons in their d or f layers and thus absorb certain wavelengths of visible light and exhibit different colors. Transition metal oxides are inexpensive and are often used in dental restorations and in the coloring of decorative materials such as gemstones [140]. Some smaller transition metal ions enter the ZrO_2 ionic lattice in the form of gap ions, forming a color center, absorbing visible light and producing color. Guo *et al.* [141] reported that the ceramics of the two components produced different colors as the doping content of Fe_2O_3 increased. When the doping content of Fe_2O_3 was 0.5 mol%, the 3YSZ ceramics were brownish-red, while the 8YSZ ceramics were light yellow, as presented in Fig. 7. In addition, Cr^{3+} can make rubies red and emeralds green, indicating that the production of color is related not only to the transition metal ion itself but also to the local crystal field [142,143].

Rare earth elements have abundant energy levels, and the electrons in the 4f layer, P layer, and O layer may transition after being excited. The transition can absorb or reflect electromagnetic radiation of various wavelengths in the ultraviolet, visible, and infrared light regions, so rare earth elements are a wide range of colored substances [144]. Rare earth ions doped with YSZ ceramics produce colors similar to those formed in materials such as crystals or glass because the 4f state that absorbs specific wavelengths of light is an electronic state in the inner shell, so the crystal field effect is negligible [145]. Chen *et al.* [146] prepared Nd:8YSZ transparent ceramics by solid-phase mixing method, and the ceramics were purple in color, as shown in Fig. 8. The results showed that the transmittance reached approximately 70% in the range from 900 to 1200 nm, which is close to the theoretical transmittance. Due to its good high-temperature resistance and high mechanical strength, this ceramic has potential application value in solid-state heat capacity lasers. In particular, the color of cerium ion is affected not only by its valence state but also by its local crystal field. Kato *et al.* [147] prepared Ce:MgO transparent ceramics via the discharge plasma sintering method, and the ceramics gradually turned yellow with increasing Ce doping concentration, while the Ce:YSZ ceramics prepared by Lv *et al.* [148] were red. This is because cerium ions exist in two charge states in the YSZ matrix: Ce^{3+} and Ce^{4+}. In a reducing atmosphere, cerium ions are present as Ce^{3+}, which causes a 4f–5d transition absorption band, giving the ceramics a red color [149,150]. When cerium ions are oxidized, since Ce^{4+} does not have 4f electrons, it does not cause absorption in the visible light band of 390–550 nm, so the color of the ceramic is yellow.

In addition to common rare earth oxides and transition metal oxide colorants, spinel structure oxides, such as the common blue $CoAl_2O_4$ spinel, are also commonly used as colorants for zirconia ceramics. The color development mechanism is that the spinel structure of the oxide and zirconia matrix forms multi-phase color zirconia ceramics, which have good high-temperature stability. Mindru *et al.* [151] believed that the deep blue color of $CoAl_2O_4$ was due to tetrahedral color development, where Co^{2+} is located in the tetrahedron and Al^{3+} is located in the octahedron. The color-producing ion Co^{2+} has a $3d^7$ unsaturated electron configuration, and the light absorption band appears in the range of visible light at 610–700 nm, resulting in changes in the excited state and ground state of the electron, which is blue in color.

2.3 Coloring methods for colored zirconia ceramics

2.3.1 *Solid-state mixing method*

The solid-state mixing method usually involves mechanical mixing ofcolorant, zirconia powder, and other additives according to a certain proportion of the mixture. After drying, forming, and high-temperature sintering treatment, colored zirconia ceramics can be produced [152]. This method has the advantages of simple process, easy operation, mature technology, low product price,

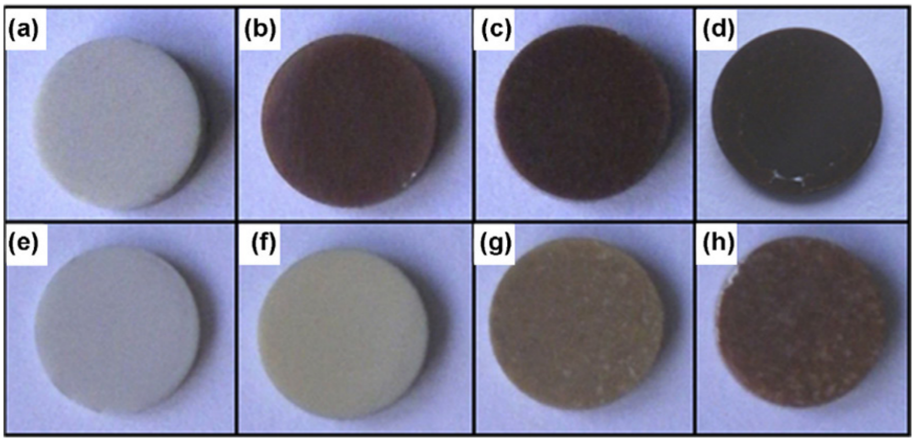

Fig. 7 Photographs of (a) 0 mol%, (b) 0.5 mol%, (c) 1.0 mol%, and (d) 1.5 mol% Fe_2O_3-doped 3YSZ samples sintered at 1400–1500 °C for 2 h and (e) 0 mol%, (f) 0.5 mol%, (g) 1.0 mol%, and (h) 1.5 mol% Fe_2O_3-doped 8YSZ samples sintered at 1400–1500 °C for 2 h. Reproduced with permission from Ref. [141], © Elsevier Ltd. 2012.

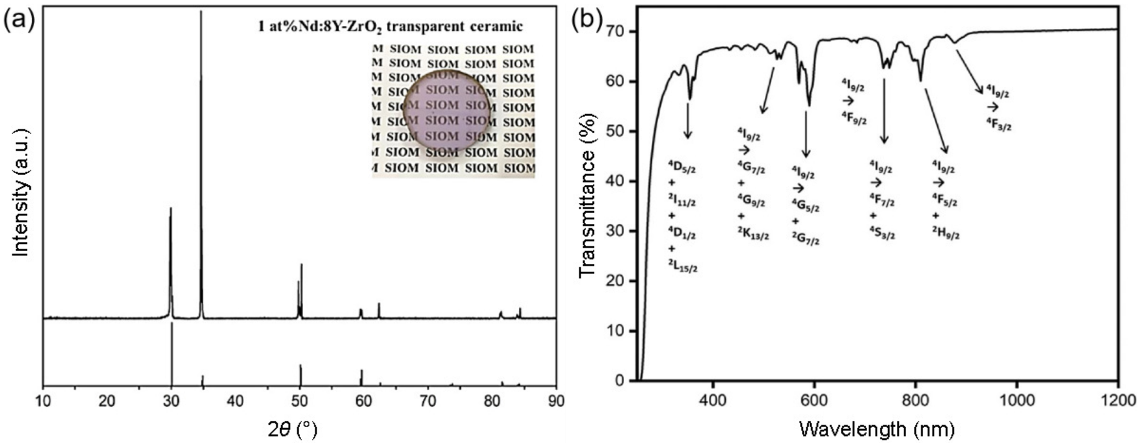

Fig. 8 (a) Sample photo, XRD pattern, and (b) transmission spectrum at room temperature of Nd:8YSZ transparent ceramics. Reproduced with permission from Ref. [146], © Elsevier B.V. 2021.

and mass production capability, so it is basically used in industry to prepare colored zirconia ceramics [153]. However, the biggest disadvantage of mechanical mixing is the agglomeration between nanoparticles during ball milling, and the diffusion mass transfer process is relatively difficult during sintering, resulting in uneven composition and coloring [154]. To overcome the agglomeration of nanoparticles, ultrasonic dispersion technology or electrolyte dispersants can be used to improve the dispersion of particles [155]. In addition, the solid phase mixing process has a long period and easily introduces impurities, which also consumes time and energy [156]. During the mechanical mixing process, the colorant structure may also be destroyed, thus affecting the integrity of the grains, which is not conducive to the color effect of the colorant in the ceramic.

Zhang *et al.* [157] used micron- or nano-scale 3YSZ powders as raw materials and Pr–Zr pigments, NH_4VO_3 or Cr_2O_3 as colorants, and added a small amount of sintering additive for mechanical mixing. Yellow, beige, or green zirconia ceramics with excellent mechanical properties could be obtained by high-temperature solid-phase sintering methods. Huang *et al.* [158] prepared yellow and red 3YSZ ceramics using Pr_6O_{11}, CeO_2, and Er_2O_3 as colorants by solid-phase mixing combined with a high-temperature sintering method. By studying the effect of colorant on the mechanical properties of ceramics, it is found that rare earth oxide colorants mainly reduce the three-point flexural strength of 3YSZ ceramics but have little effect on the Vickers hardness and fracture toughness. Gao *et al.* [159] prepared 3YSZ ceramics by doping a trace amount of rare earth oxide colorant by solid-phase mixing method and found that the prepared ceramics had excellent mechanical properties.

2.3.2 Chemical co-precipitation method

The chemical co-precipitation method involves mixing zirconium salt, a stabilizer and colored ions according to a certain proportion to prepare a salt solution, with appropriate alkaline solution as the precipitator, adjusting the solution pH = 9.0–9.5 to obtain the precursor, washing, drying, screening, calcination and a series of processes to obtain colored stable zirconia powders, and forming and sintering to prepare colored zirconia ceramics [160,161]. This method enables homogeneous mixing of the colorant and matrix elements at the atomic level, thus significantly enhancing the uniformity of coloring. Additionally, the cost-effective zirconium salt solution can serve as an initial raw material for synthesizing high-purity and ultrafine zirconia powder doped with coloring ions [162]. However, this method has the drawback of inherent complexity, primarily arising from the complexity of co-precipitated ions. During the high-temperature calcination process, unpredictable reactions between different ions may occur, leading to substantial consumption of colored ions and subsequently rendering the final impact on color and mechanical properties of colored zirconia ceramics unpredictable. Luo *et al.* [163] synthesized Ho^{3+}/Yb^{3+} co-doped 8YSZ nanoparticle by chemical co-precipitation method and studied their luminescence properties. The results revealed that the powder had a pure cubic structure and was approximately spherical after calcination at 800 °C, with an average grain size of 11 nm.

2.3.3 Liquid-phase precursor infiltration method

The liquid-phase precursor infiltration method involves infiltration of the body containing connected pores into solution containing colored ions, and the colored zirconia ceramics are prepared by drying and sintering at high temperature [164,165]. This method is relatively simple, but it is affected by many external factors, such as the type of coloring agent, soaking time, coloring solution concentration, and coloring agent ion distribution, which directly affect the coloring effect and mechanical properties of ceramics [166,167]. In particular, when the volume of the zirconia body is too large, it is difficult for the coloring ions to completely infiltrate into the body, resulting in uneven coloring. In addition, if the immersion time is not properly controlled, the surface or edge of the body will fall off, resulting in obvious cracks on the ceramic surface after sintering, thereby reducing the mechanical properties of the ceramics.

In 2011, Liu *et al.* [168] immersed zirconia body in a solution containing Co^{2+} and Al^{3+} ions and sintered it in air to prepare blue ZrO_2–$CoAl_2O_4$ ceramics. The prepared colored ceramic bars have a core-shell structure, and the thickness of the colored ceramic rod can be controlled by adjusting the soaking time and temperature, as shown in Fig. 9. In addition, zirconia ceramics of different colors were also successfully prepared by soaking in a solution containing ions of different colors, thus providing an effective way to prepare colored zirconia ceramic parts.

Ban *et al.* [169] studied the influence of six different colorants on the bending strength of zirconia ceramics by liquid phase precursor infiltration method and found that the bending strength of zirconia ceramics obviously decreased after coloring with a pink liquid containing Nd^{3+} and Er^{3+}. Kaplan *et al.* [170] immersed 3YSZ ceramics pre-sintered at 1450 °C in $NiCl_2$, $MoCl_3$, and $NiCl_2$/$MoCl_3$ solutions for 5, 30, and 60 s, respectively, and different concentrations (0.1, 0.25, and 0.5 wt%) were applied to

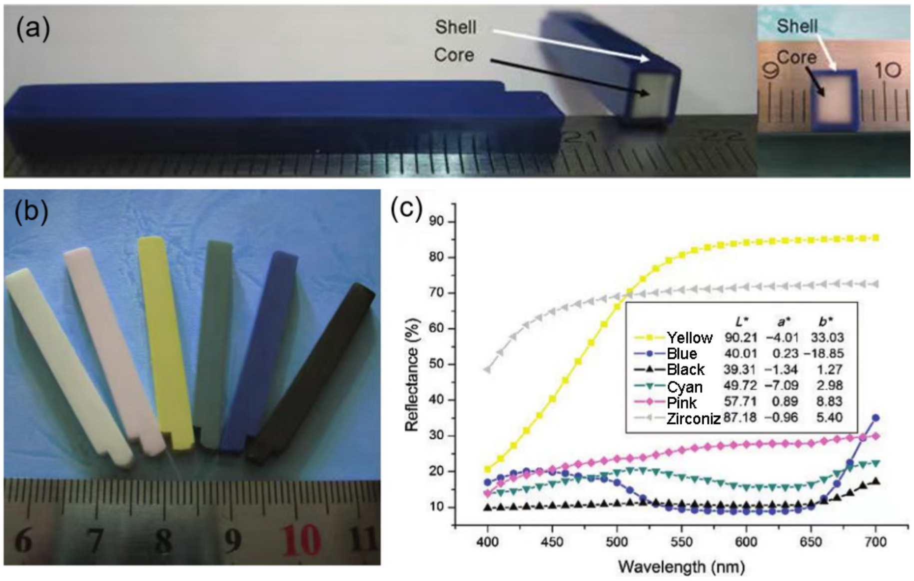

Fig. 9 (a) Cross-section of sintered blue zirconia bar immersed in blue solution for 3 h at 40 °C. (b) Photographs and (c) reflectance curves and CIE Lab coordinates of colored zirconia ceramics prepared by infiltration method. Reproduced with permission from Ref. [168], © Maney Publishing 2011.

prepare colored zirconia ceramics with a natural tooth color. It is found that the color produced depends on the type and concentration of colorant solution, whereas immersion duration has no significant effect on the color of the ceramics. In 2020, Jing *et al.* [171] dipped 3YSZ ceramics prepared via the solid-phase method in a dyeing solution, then sintered them into porcelain after immersion and dyeing, and investigated the effects of Pr and Er single elements on 3YSZ coloring and the influence of multiple factors on the overall coloring effect after mixing. The results revealed that Pr and Er were yellow and red in the process of coloring, respectively, and when the two elements were used for dyeing at the same time, the values of a^* and b^* of the mixed color presented the additivity of a^* and b^* and the value of L^* increased, as shown in Fig. 10, which is of great significance to the development of dental ceramics.

2.3.4 *Chemical coating method*

The chemical coating method involves adding dispersed zirconia powder into water to form a suspended solution and adjust the pH value so that the precipitation reaction of colored ions occurs on the surface of the zirconia matrix through a precipitating agent to form a core–shell-coated composite structure powder, which is an effective method for preparing bright-colored zirconia ceramics [172]. Compared with the solid-phase mixing method, the chemical coating method can uniformly coat and mix the colored ions on the zirconia particles and effectively overcome the agglomeration between particles to shorten the mass transfer distance for the reaction and reduce the decomposition or volatilization of colored ions at high temperatures.

Qian *et al.* [173] used Y-TZP as a raw material, $Mn(NO_3)_2{\cdot}6H_2O$, and $Al(NO_3)_3{\cdot}9H_2O$ as coloring agents and polyethylene glycol (PEG) 2000 as the dispersant to synthesize black zirconia ceramics with a uniform color and bright luster by a heterogeneous precipitation method. It is found that the prepared composite powder has a shell–core structure, and the fabricated zirconia ceramics have bright black color with a homogeneous, fine, and dense microstructure after sintering, as shown in Fig. 11. Compared with the mechanical mixing method, the mass transfer distance for the reaction of the incorporated colorant elements could be effectively reduced, which could

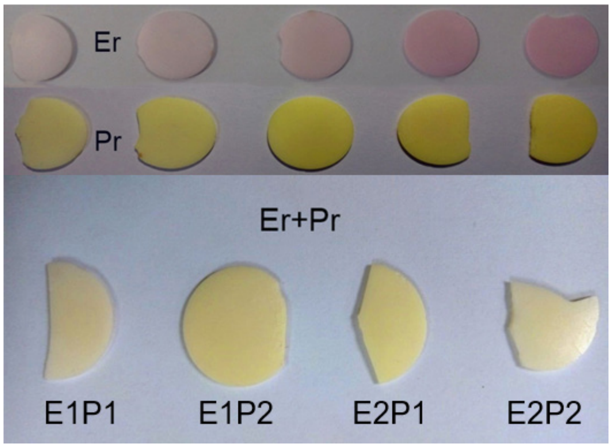

Fig. 10 Coloring effect of different concentrations of Er and Pr solutions and mixed dyeing. Reproduced with permission from Ref. [171], © The Minerals, Metals & Materials Society 2019.

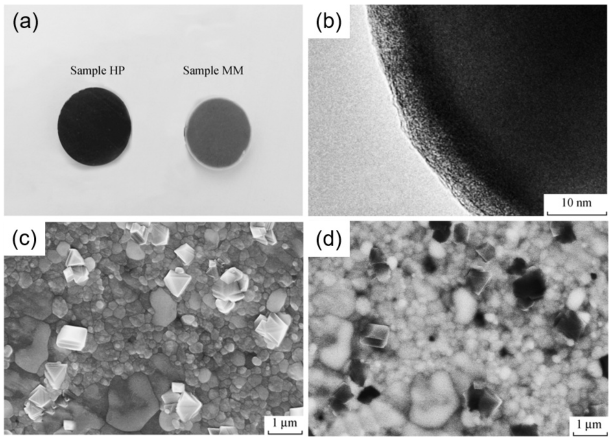

Fig. 11 (a) Photograph of samples prepared by heterogeneous precipitation (HP) method and mechanical mixing (MM) method. (b) TEM micrographs of the prepared composite powder. (c) SEM and (d) backscattered electron image (BSE)-SEM micrographs of samples prepared by heterogeneous precipitation method. Reproduced with permission from Ref. [173], © China Academic Journal Electronic Publishing House 1994–2023.

facilitate the formation of $MnAl_2O_4$ coloring phases uniformly dispersed in the zirconia matrix with less volatilization. However, this method is difficult to control due to its complicated process and many influencing factors, so it is not suitable for batch preparation.

3 Research progress on colored zirconia ceramics

In chromatics, the three primary colors, namely, red, blue, and green, serve as the foundational hues for blending various shades. Any other color can be obtained by overlaying the three primary colors. Among them, red is a culturally significant color in China, while blue is renowned for its classic coolness, embodying traits of tranquility and rationality, which enjoy widespread popularity and find extensive application in personalized decoration products and smart wearable devices. Furthermore, black, which serves as the predominant hue in high-end applications such as mobile phone backplanes, holds significant relevance in the exploration of colored zirconia ceramics. Therefore, the research progress on blue, red, and black zirconia ceramics is reviewed in this section.

3.1 Red zirconia ceramics

Red ceramics have been endowed with unique cultural connotations in Chinese history and also have a pivotal value in the field of decorative materials. As one of the three primary colors, red color is of great significance to the regulation of ceramic color, and researchers are constantly conducting research and exploration on red color. Red porcelain began in the late Tang Dynasty, with Fe_2O_3 as the main glaze applied to the surface of the porcelain, but its color was not satisfactory.

Since 1960, Pr-doped ceria has been employed as a red ceramic pigment in the ceramic industry by Aruna *et al.* [174]. In continuation of the research on ceria [175] and ceramic pigments [176,177], the $Ce_{1-x}Pr_xO_{2-\delta}$ red pigment was synthesized by the thermal decomposition of precursors and combustion of aqueous solutions at high temperature. It is found that the diffuse reflectance spectrum of the pigment shows an absorption edge at 690 nm, and the color changes from brick red to brown with increasing Pr doping. According to Masó *et al.* [178], the optimum synthesis temperature for the preparation of Pr_2O_3-doped CeO_2 red pigments by solid-state reactions is 1400–1500 °C. Bondioli *et al.* [179] used the flux method to synthesize the $Ce_{1-x}Pr_xO_2$ red pigment at low temperatures. In addition, Zhu *et al.* [180] synthesized a $Ce_{1-x}Pr_xO_2$ nano-crystalline red pigment via low-temperature combustion. In 2010, Llusar *et al.* [181] prepared homogeneous (single-phase) $Ce_{1-x}Tb_xO_2$ solid solutions at lower temperatures by optimizing the synthesis methods, which can be used as a reddish ceramic pigment.

Some sulfides can also be used as red pigment ceramics. Vasilyeva *et al.* [182] prepared red ceramics using red sulfide (Ce_2S_3) as a colorant. The Ce_2S_3 solid solutions are also red. Grazia *et al.* [183] used cadmium selenide sulfide ($CdS_{1-x}Se_x$) as a colorant to produce a series of low-temperature red ceramics with different tones. The color of the $CdS_{1-x}Se_x$ solid solutions is tuned from orange to red by increasing the amount of selenium. Its redness value is as high as 50, reaching the level of a large red color. However, sulfides cannot be stably present in a high-temperature oxidation environment. At temperatures greater than 800 °C, sulfides decompose and produce toxic sulfide fumes. In addition, cadmium sulfide is a carcinogen that is harmful to humans.

In 2020, Zhang *et al.* [184] prepared two colorants with $ErAlO_3$ and $Er_3Al_5O_{12}$ as the main crystal phases by the gel-polymerization method and then prepared pink 3YSZ ceramics via the gel-casting method. It was found that Er^{3+} might dissolve into the zirconia lattice to form a cubic solid solution at high temperatures, and the remaining Al_2O_3 was distributed in the zirconia solid solution matrix as the second phase. When up to 5 wt% $ErAlO_3$ was added, the redness value a^* of 3YSZ ceramics reached the maximum value, which was 13.6.

To obtain a red zirconia ceramic with a high redness value, Lv *et al.* [148] fabricated Ce-doped red zirconia ceramics via a high-temperature reduction method, and the absorption wavelength ranged from 480 to 500 nm, as shown in Fig. 12. As the reduction temperature and the amount of doped CeO_2 increased, the redness value of a^* of the ceramics first increased and then decreased, and the maximum a^* reached 32.5. In addition, it also had excellent mechanical properties, with the fracture toughness reaching a maximum of 9.6 $MPa \cdot m^{1/2}$. The colorant is more environmentally friendly and safer than traditional sulfide red colorants.

In addition, iron oxide has been used extensively as a red colorant for preparing red ceramics, but its color rendering was poor, and the samples were orange–red [185,186]. The value of a^* was low, at less than 20, and the yellowness value of b^* was greater than 20, which indicated that the samples had a lower red hue. Therefore, its color cannot meet the requirements of red; the addition of iron oxide greatly reduces the mechanical properties of the 3YSZ system, and the fracture toughness of Fe-doped red ceramics is distributed within the range of 5.0–6.0 $MPa \cdot m^{1/2}$, which greatly limits their application in industry [187].

Yttrium aluminum garnet (YAG) is an important optical ceramic material, especially when doped with Nd and Er, which has a specific color and is widely used in artificial gems and other decorative industries. For instance, Er:YAG is pink, and Nd:YAG is light reddish purple [188–190]. Both are light in color. Furthermore, the sintering temperature of YAG can reach 1750 °C, which increases the difficulty and energy consumption of the process.

In 2022, Wang *et al.* [191] prepared highly transparent Ce-doped yttria-stabilized zirconia ceramics with bright red color using commercial powders as raw materials via a solid-state mixing method combined with two-step sintering technology. Owing to the 4f–5d transition absorption of Ce^{3+} at 390–550 nm, the Ce-doped 8YSZ had a high in-line transmittance of 70.7% at 700 nm and a large redness of 57.1, as shown in Fig. 13. The results showed that with increasing air annealing temperature, the redness value decreased from 57.1 to 0.8 due to the oxidation of Ce^{3+} to Ce^{4+}. Furthermore, to improve the color uniformity of red ceramics, the research team prepared Ce-doped 8YSZ transparent ceramics by two-step sintering using the nano-powder synthesized via a co-precipitation method [192]. The ceramics show the best in-line transmittance of 47.6% at 700 nm and the highest redness of 52.0, as shown in Fig. 14. The successful preparation of red zirconia transparent ceramics can broaden the application of zirconia ceramics in filter materials, signal lampshades, and other fields.

3.2 Blue zirconia ceramics

In traditional blue ceramics, blue glaze is usually used as the main color, and blue color is usually used as the oxide of trivalent cobalt, which is the first element developed for blue glaze. In industry, cobalt oxide (Co_2O_3) is still mainly used to synthesize blue spinel to prepare blue ceramics. At present, blue ceramic pigments mainly include vanadium–zirconium blue materials as

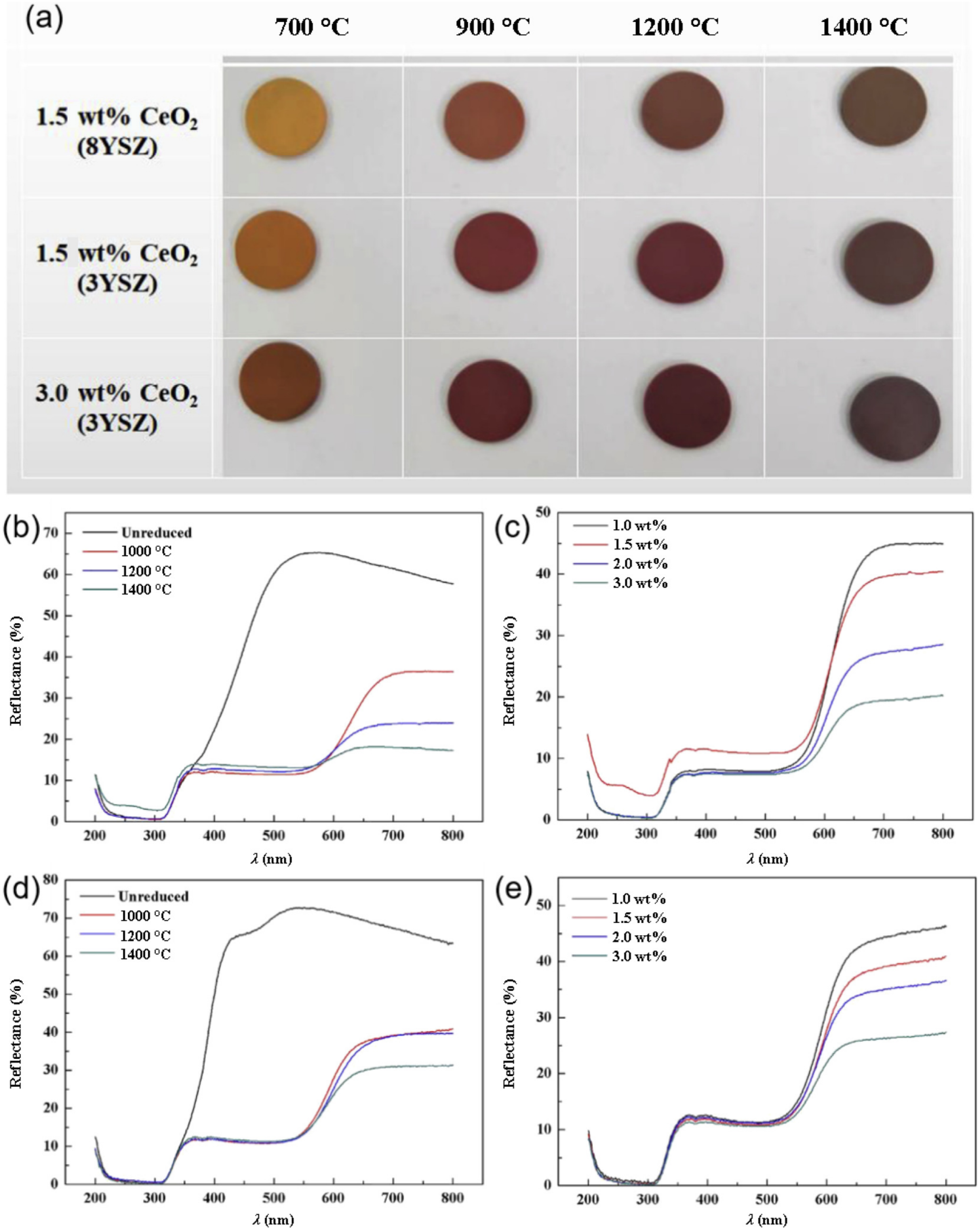

Fig. 12 (a) Photo comparing different systems of CeO_2-doped YSZ ceramics. Ultraviolet–visible (UV-Vis) diffuse reflectance spectra of (b, c) Ce-3YSZ ceramics and (d, e) Ce-8YSZ ceramics. Reproduced with permission from Ref. [148], © Elsevier B.V. 2019.

colorants, cobalt–aluminum spinels, nickel–aluminum spinels and spinel colorants, which use other ions to replace cobalt ions, and hexaluminate and lanthanide colorants as representatives. On the basis of ensuring the color rendering and mechanical properties, the exploration of environmentally friendly and economic blue colorants is still the focus of current research.

Chen *et al.* [193] synthesized blue cobalt-aluminum spinels via a hydrothermal method using $CoCl_2·6H_2O$ and $AlCl_3·6H_2O$ as raw materials. However, owing to the long cycle of this method, it is not conducive to industrial production.

By using the solid phase method, Wang *et al.* [194] used nanoscale 3YSZ powder as a raw material, V–Zr pigments or Co_3O_4 as colorants, and added a small amount of sintering additives to prepare bright blue 3YSZ ceramics. The prepared blue 3YSZ ceramics exhibited impressive hardness values of 13.96 and 13.21 GPa, respectively, accompanied by bending strengths of 690 and 613 MPa, respectively.

Wang *et al.* [195] prepared uniformly blue-colored $CoAl_2O_4$–ZrO_2 ceramics via a heterogeneous nucleation method, as shown in Fig. 15, with a blueness value b^* of 31.41. PEG 2000 was used as the dispersant for the ZrO_2 powders, followed by introduction of Al and Co hydrates to the suspension for preparation of coated powders, and then, NH_3 was added into the suspension to tailor the pH values to favor the heterogeneous nucleation of $CoAl_2O_4$ spinels. It is believed that heterogeneous nucleation could be a facile and cost-saving route to facilitate solid-state reactions and guarantee the uniform distribution of new phases within the matrix for the fabrication of colored ceramics.

Zhou *et al.* [196] made some improvements on the basis of the traditional $CoAl_2O_4$ spinel colorant and used the sol–gel method

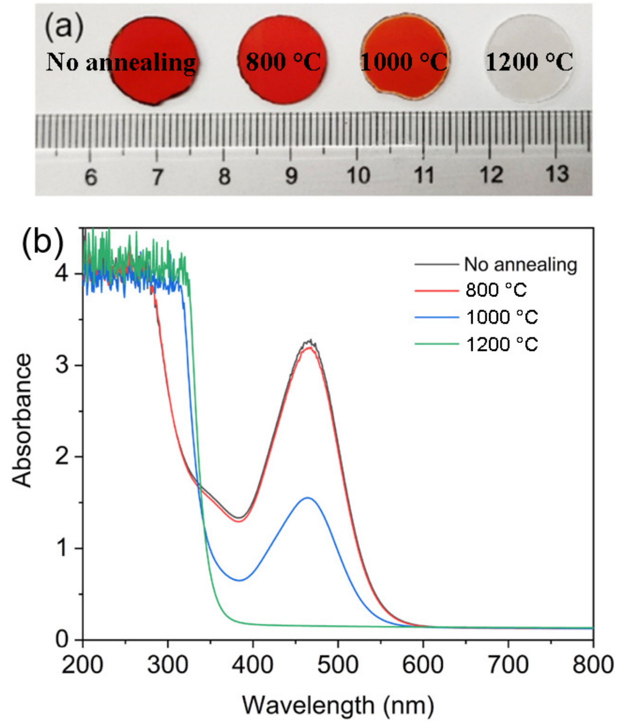

Fig. 13 (a) Photograph and (b) absorption spectra of Ce-doped 8YSZ ceramics un-annealed and annealed at different temperatures for 3 h. Reproduced with permission from Ref. [191], © Elsevier B.V. 2022.

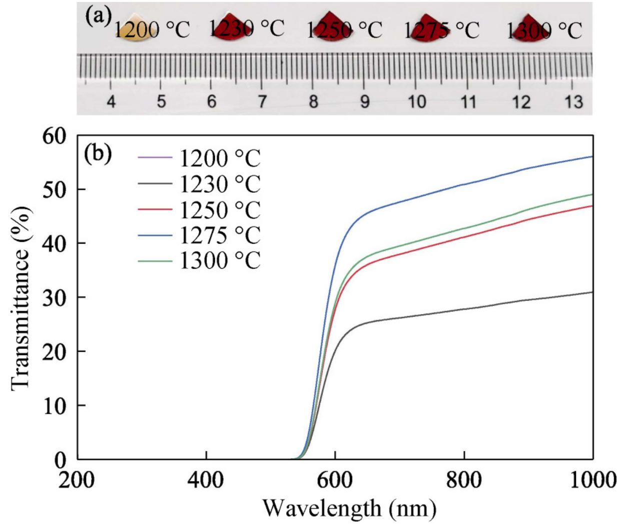

Fig. 14 (a) Photograph and (b) in-line transmittance of Ce-doped 8YSZ ceramics (1 mm thick) pre-sintered at 1200–1300 °C in air for 2 h and subjected to hot isostatic pressed (HIP) post-treatment at 1700 °C for 3 h under 176 MPa in Ar atmosphere. Reproduced with permission from Ref. [192], © China Academic Journal Electronic Publishing House 1994–2022.

as an experimental method to replace cobalt ions with calcium ions to form $Co_{1-x}Ca_xAl_2O_4$ solid solution. With increasing calcium content, the brightness of the blue pigment increased, and the blueness value changed from 12 to approximately 30. In the range of visible wavelengths from 530 to 670 nm, the light absorption intensity tended to decrease.

Hu *et al.* [197] prepared cobalt blue ceramic ink via three different chemical coagulation methods: gelling first and then dissolving, mixing and gelling, and co-dissolving and gelling; they found that the ceramic hair color ink produced by co-dissolving and gelling had relatively excellent performance and high stability.

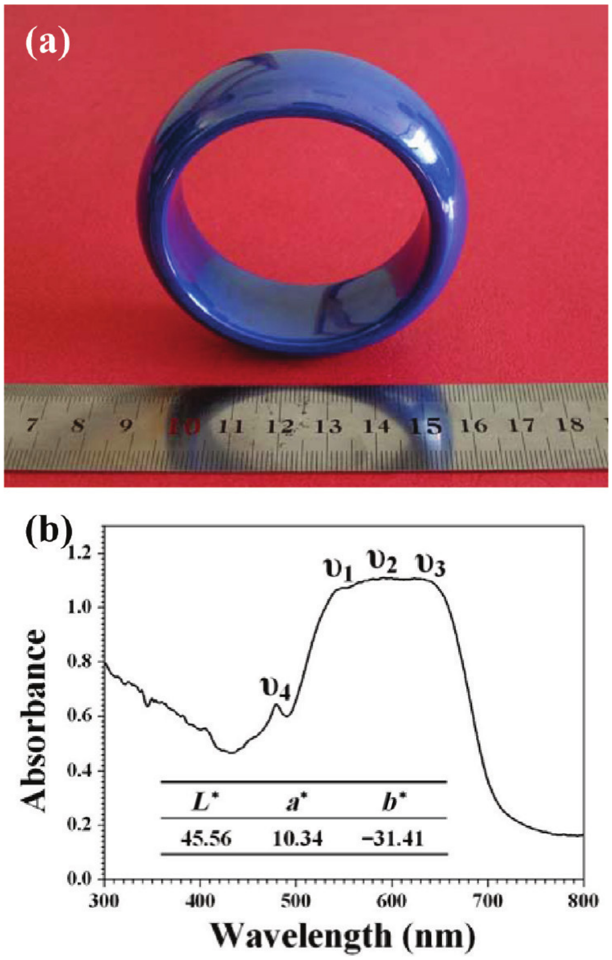

Fig. 15 (a) As-fabricated ring-shaped and blue-colored ZrO_2 parts formed via heterogeneous nucleation. (b) UV–Vis spectrum of blue-colored ZrO_2 ceramics. Reproduced with permission from Ref. [195], © American Chemical Society 2009.

Xu *et al.* [198] prepared blue zirconia ceramics with high toughness and strength using Co_2O_3 and Al_2O_3 as composite colorants together through the combination of cold isostatic pressing and high-temperature sintering. The prepared zirconia ceramics have a uniform color distribution and bright color. When the molding pressure was 200 MPa and the sintering temperature is 1500 °C, the mechanical properties of blue zirconia are better; the density is up to 99%, and the bending strength is 740 MPa.

Wang *et al.* [199] synthesized solid solutions based on $BaAl_{12-x}M_xO_{19}$ (M = Co^{2+}, Ni^{2+}) with intense blue shades by conventional solid-state reaction. Novel zirconia ceramics with intense blue hues were then prepared upon the addition of 5 wt% synthesized chromophore to a ZrO_2 white matrix containing 5 wt% Y_2O_3 and sintered at 1400 °C for 6 h. This is a promising blue colorant for zirconia ceramics that exploit only minimal amounts of toxic metal elements.

Cheng *et al.* [200] successfully synthesized novel solid solutions based on $Ba_{0.956}Mg_{0.912}Al_{10.088-x}Ni_xO_{17}$ and $Ba_{0.956}Mg_{0.912}Al_{10.088-x}Co_xO_{17}$ via the solid-state method. The as-prepared blue oxides were sintered with ZrO_2 powders at 1400 °C to prepare blue zirconia ceramic materials. The results revealed that the pigment phase was stable after high-temperature sintering with ZrO_2, and the grain boundaries were clear. Additionally, the prepared blue ceramics were dense, and the mechanical properties of the fabricated blue ceramics were maintained compared to pure ZrO_2 ceramics.

In summary, the current blue ceramic pigments mainly include V–Zr blue pigments, $CoAl_2O_4$, $NiAl_2O_4$, and spinel-type pigments, where cobalt ions are substituted by other ions, hexaluminate, and lanthanide colorants represented by related colorants. On the basis of ensuring color rendering and mechanical properties, the exploration of environmentally friendly and economic blue colorants is still the focus of current research directions.

3.3 Black zirconia ceramics

With the development of electronic ceramic decorations, black ceramics are deeply loved by people for their solemn and unique colors. The preparation of black ceramics is inseparable from the research and development of black pigments [201–203]. Owing to the scarcity and high price of cobalt oxide sources, researchers have been actively investigating alternative options to replace cobalt in zirconia ceramic colorants. Using MnO_2, Fe_2O_3, and Cr_2O_3 as raw materials, three different colors of spinel can be obtained through high-temperature calcination, including dark brown chromium iron spinel, dark red iron manganese spinel, and dark green chromium manganese spinel. By adjusting the ratio of components to control the proportion of each spinel, different colors interact with each other to obtain a stable black color. This strategy greatly reduces production costs and improves economic viability.

In the preparation of traditional black ceramics, cobalt oxide is usually introduced as a raw material. Etoh *et al.* [204] synthesized black zirconia ceramics co-doped with Co_3O_4, Cr_2O_3, and TiO_2 using a high-temperature solid-state reaction method. However, the phase structure of black spinel is unstable when the colorant is doped into zirconia. The phase structure of black ceramics becomes disorganized, resulting in drastic changes in volume and even cracking, which further reduces the color stability and mechanical properties of the ceramics. In addition, the color stability of the prepared black zirconia ceramics is poor, and the colorant volatilizes seriously at high temperature; there are certain environmental problems. Briod [205] prepared black zirconia by doping $CoFe_2O_4$ into the zirconia matrix. Although Cr ions were not introduced via this method, the coloring ion was not high-temperature-resistant or volatile, which not only polluted the sintering equipment but also affected the chroma and mechanical properties of the ceramic products.

Gargori *et al.* [206] used Cr/Ni/Cu electroplating waste as a raw material and prepared black ceramic pigments by sintering at 1100 °C for 1 h. However, the L^* value of this ceramic color is low, less than 40, and the values of a^* and b^* are discreetly distributed, with maximum values of −8.4 and 53.5, respectively. This indicates that the sample has a low blackness value, and the pigment is not pure black; the brightness is low, and the effect of bright black cannot be achieved.

In recent years, black TiO_{2-x} materials have been obtained via H_2 reduction treatment, chemical reduction, chemical oxidation, electrochemical reduction, anode oxidation annealing, and other methods to produce oxygen vacancies and defects on the surface and body phase of TiO_2 [207]. To improve the color unity and phase stability of black ceramics, Lv *et al.* [208] used TiO_2 instead of traditional composite black spinel and prepared black titanium-doped zirconia ceramics via a high-temperature reduction method, replacing the traditional dyeing method. After high-temperature reduction, the concentrations of Ti^{3+} and oxygen vacancies increased, and strong absorption of visible light occurred, as shown in Fig. 16. The lightness value L^* of the prepared ceramic is approximately 44, and both the a^* and b^* values are close to 0, which indicates that the sample is bright black.

However, during the preparation process of black zirconia ceramics, the multi-phase spinel structure is prone to react with the zirconia matrix. This interaction will lead to destruction of the spinel structure and a decrease in phase stability, which is not conducive to the mechanical properties of ceramics. In addition, the color of the reduced zirconia powder preparation is gray-black, which also brings greater challenges to the preparation of bright black zirconia.

Table 1 summarizes CIE coordinates L^*, a^*, and b^* ($L^* = 0$ indicates black; $L^* = 100$ indicates white; $a^* > 0$ indicates red; $a^* < 0$ indicates green; $b^* > 0$ indicates yellow; $b^* < 0$ indicates blue) and main mechanical properties of colored zirconia ceramics.

4 Conclusions and outlook

In summary, this paper reviews the research progress on colored zirconia ceramics. Owing to their diverse color characteristics, excellent mechanical properties, and good biocompatibility, colored zirconia ceramics have been widely used in communication, advanced decorations, biomaterial fields and so on. In future research, the evolution of color zirconia ceramics will

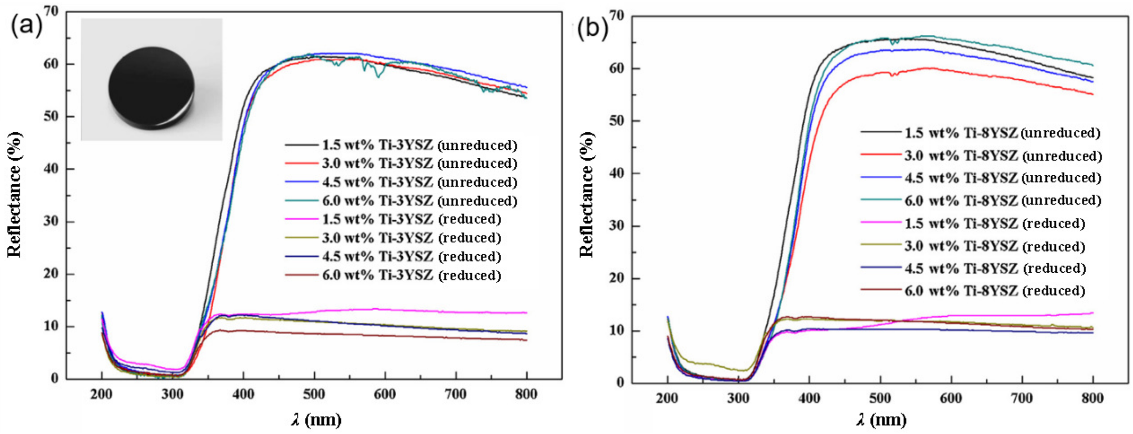

Fig. 16 UV–Vis diffuse reflectance spectra of Ti-doped YSZ ceramics: (a) Ti-doped 3YSZ ceramics and (b) Ti-doped 8YSZ ceramics. Reproduced with permission from Ref. [208], © The Authors 2020.

Table 1 Chromaticity values and mechanical properties of colored zirconia ceramics

Color	Colorant	Chromaticity value			Mechanical property			Ref.
		L^*	a^*	b^*	Bending strength (MPa)	Vickers hardness (GPa)	Fracture toughness ($MPa\cdot m^{1/2}$)	
Yellow	Fe_2O_3	~65.00	~5.00	~12.00	~1100.00	—	—	[120]
		—	—	—	—	~13.00	~3.60	[124]
	VO_3^-	90.21	−4.01	33.03	—	—	—	[168]
	CeO_2	~80.00	−4.15	8.30	842.00	—	—	[120]
	Pr_6O_{11}	79.54	−2.07	23.50	—	—	—	[171]
	Pr_6O_{11}, Er_2O_3	82.82	1.40	16.22	—	—	—	[171]
Red	Fe_2O_3	~55.00	~10.00	~17.00	—	~11.00	~3.50	[123]
	$Ce_{(1-x)}Pr_xO_2$	57.32	25.04	23.42	—	—	—	[180]
	CeO_2	47.20	32.50	22.40	—	13.70	9.60	[148]
	CeO_2	42.40	52.00	72.90	—	—	—	[192]
Pink	Er^{3+}	78.11	6.16	−5.52	1470.65	13.25	12.18	[129]
	Er_2O_3	~70.00	~1.00	~−5.00	~1100.00	—	—	[120]
	$ErAlO_3$, $Er_3Al_5O_{12}$	87.35	13.63	0.75	—	—	—	[184]
Blue	Vr–Zr	—	—	—	690.00	13.96	—	[194]
	Co_3O_4	—	—	—	613.00	13.21	—	[194]
	$CoAl_2O_4$	45.56	10.34	−31.41	—	—	—	[195]
	$CoAl_2O_4$	40.01	0.23	−18.85	—	—	—	[168]
	Co^{2+}, Al^{3+}	48.56	1.22	−20.38	1185.42	13.08	13.32	[129]
	Co_2O_3, Al_2O_3	37.34	4.19	−28.01	740.00	—	—	[198]
	$NiO\cdot nAl_2O_3$	64.70	−22.20	−24.50	964.00	—	—	[135]
	$CoAl_2O_4$, $Nd_2Zr_2O_7$	43.79	0.34	−21.26	—	14.10	9.00	[209]
Green	Ni^{2+}, Al^{3+}	54.71	−7.59	−1.99	2088.48	12.72	14.52	[129]
	Cr_2O_3, Al_2O_3	47.27	−5.27	2.34	631.98	13.10	5.31	[130]
	NiO, Al_2O_3	54.10	−11.66	10.55	581.98	9.38	5.14	[130]
Purple	Nd_2O_3	71.70	5.57	−7.58	—	12.37	5.77	[210]
	$Nd_2Zr_2O_7$	68.23	8.94	15.22	—	12.93	8.15	[211]
Black	$MnAl_2O_4$	43.58	0.37	0.90	—	—	—	[173]
	MnO_2	45.10	~4.00	~−4.00	—	—	—	[128]
	Fe^{3+}, Co^{2+}, Cr^{3+}, Al^{3+}	42.01	0.22	−0.73	1064.45	12.00	13.96	[129]
	TiO_2	~44.00	~0	~0	—	—	—	[208]

be underscored by the exploration of novel colorant combinations alongside advancements in mechanical properties. Concurrently, the pursuit of innovative coloring methodologies represents a pivotal avenue for future investigations. Specifically, with respect to red zirconia ceramics, the transformation from the initial sulfide and Fe_2O_3 coloring to the reduction of CeO_2 is aimed at improving the red chrominance of the ceramics, but it is a significant challenge to mitigate the adverse impact of oxygen vacancy-induced color darkening when Ce^{4+} is reduced to Ce^{3+}. It is a future development direction to adjust the valence state balance in ceramics by designing other sintering additives. For blue zirconia ceramics, exploring other ions to replace cobalt ion coloring in $CoAl_2O_4$ provides a new idea for the development of blue ceramic colorants and balances the relationship between ceramic performance and economy. The elucidation and manipulation of the intricate relationships among the composition, microstructure, color, and properties of colored zirconia ceramics are poised to become central pursuits in future research endeavors. This holistic approach is indispensable for informing the design and fabrication of colored zirconia ceramics with tailored properties to meet the demands of various industries, including but not limited to biomedicine, electronics, and structural engineering.

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Declaration of competing interest

The authors have no competing interests to declare that are relevant to the content of this article. The author Jiang Li is the Editorial Committee member of this journal.

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