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Research Article

Synergistic regulation of electron and phonon transport in SrTiO₃-based thermoelectric perovskites via entropy engineering of A-site cations

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Abstract: SrTiO₃-based thermoelectric oxides are promising candidates for high-temperature energy conversion because of their low cost, environmental compatibility, and excellent thermal stability. However, their relatively low thermoelectric performance remains a key limitation to practical applications. In this work, a series of multicomponent A-site medium- and high-entropy perovskite oxides, (Sr_{1/2}Ba_{1/4}Ca_{1/4})_{1-x}(La_{1/3}Nd_{1/3}Sm_{1/3})_xTiO₃ (SBC-RE_x; $x = 0.08\text{--}0.18$), were developed to simultaneously regulate electron and phonon transport. All samples maintain a cubic SrTiO₃ perovskite phase with $Pm\text{--}3m$ symmetry and homogeneous elemental distribution. The doping of La³⁺, Nd³⁺ and Sm³⁺ electron donors modulates the electron carrier concentration, thereby increasing electrical conductivity while preserving moderate carrier mobility and effective mass, leading to enhanced power factor at $x = 0.14$. Meanwhile, entropy engineering via co-incorporation of multielement rare earth cations in the A-site induces chemical disorder, local lattice distortion and strain fields, which suppress grain growth and intensify phonon scattering, leading to reduced lattice thermal conductivity. Consequently, the optimized SBC-RE_{0.14} sample achieves a power factor of 1.24 mW m⁻¹ K⁻² and thermal conductivity of 3.08 W m⁻¹ K⁻¹, reaching a maximum ZT of 0.31 at 773 K. This work demonstrates that A-site entropy engineering offers an effective strategy to balance electron and phonon transport in SrTiO₃-based oxide thermoelectrics.

Keywords: thermoelectric materials, strontium titanate, entropy engineering, thermal conductivity

1 Introduction

Thermoelectric materials can directly convert a temperature gradient into electrical energy (vice versa) and therefore have great potential in waste heat recovery, solid-state refrigeration, and thermal

management of microelectronic devices [1–4]. Their performance is generally evaluated by the dimensionless figure of merit, $ZT = S^2\sigma T/\kappa$, where S is the Seebeck coefficient, σ is the electrical conductivity, T is the absolute temperature, and κ is the total thermal conductivity. Because the transport parameters are strongly interdependent, an increase in electrical conductivity is often accompanied by a reduction in the Seebeck coefficient and an increase in electronic thermal conductivity [5]. Therefore, the synergistic regulation of carrier and phonon transport is crucial for improving thermoelectric performance [6].

In recent years, remarkable progress has been achieved in chalcogenide [7,8] and intermetallic [9,10] based thermoelectric materials. However, these materials generally suffer from resource scarcity, elemental toxicity, and insufficient stability arising from oxidation, volatilization, and degradation at high temperatures. In contrast, oxide thermoelectric materials have attracted sustained attention owing to their excellent chemical stability, superior oxidation resistance at elevated temperatures, abundant raw materials, and environmental friendliness [11]. Representative systems include n -type oxides, such as ZnO [12], TiO₂ [13], and perovskite oxides (SrTiO₃ [14] and CaTiO₃ [15]), as well as p -type oxides, such as layered cobaltites [16] and BiCuSeO [17].

Among various oxide thermoelectric systems, SrTiO₃-based materials have become an important research focus because of their highly tunable perovskite structure and excellent chemical and thermal stability [18]. Nevertheless, pristine SrTiO₃ still suffers from intrinsically high lattice thermal conductivity and limited electrical transport performance, which restrict further enhancement of its overall thermoelectric performance [18]. To address these issues, strategies such as elemental doping [19–21], vacancy engineering [22], and nanostructure design [23] have been widely adopted to optimize its transport behavior. However, although these approaches can effectively suppress lattice thermal conductivity, they often introduce stronger carrier scattering, thereby deteriorating carrier

mobility and limiting the power factor. Therefore, how to enhance phonon scattering while retaining favorable electrical transport remains a central issue in the study of SrTiO₃-based thermoelectric oxides.

In recent years, entropy engineering has emerged as a promising strategy for regulating the performance of SrTiO₃-based thermoelectric materials [24–26]. By introducing multicomponent configurations at the A/B sites of perovskite oxide to form medium-/high-entropy solid solutions [27,28], the atomic mass/size mismatch, point-defect scattering, and local lattice disorder can be significantly enhanced, thereby effectively suppressing phonon transport and reducing lattice thermal conductivity. For example, Zhu et al. reported a multicomponent high-entropy perovskite oxide prepared under a reducing atmosphere, the equimolar A/B site multielement doping significantly increased chemical disorder while maintaining structural stability, introducing abundant point defects and local lattice distortions [29]. As a result, the material exhibited glass-like ultralow lattice thermal conductivity over 300–873 K, reaching a minimum of about 1.6–1.9 W m⁻¹ K⁻¹. However, the power factor of the material at 873 K was only 39.5 μW·m⁻¹ K⁻². This relatively poor electrical performance may be attributed to excessive lattice disorder and complex B-site substitution, which can enhance carrier scattering, reduce carrier mobility, and disturb the Ti–O–Ti electron transport pathway [30]. In this context, appropriate selection of substitution elements, their relative compositions, and valence states may enable the synergistic optimization of electron and phonon transport, thereby reducing thermal conductivity without excessively compromising electrical performance. Notably, Zheng et al. demonstrated carrier–phonon decoupling in entropy-engineered SrTiO₃-based thin films [14], entropy-engineering can simultaneously suppress phonon transport via A-site compositional disorder, while improving carrier mobility by tuning Ti displacement and local symmetry, achieving a room-temperature lattice thermal conductivity of 1.25 W m⁻¹ K⁻¹ and a weighted mobility of 65 cm² V⁻¹ s⁻¹. In bulk ceramics, Bi et al. demonstrated that medium-entropy La-doped (Sr_{1/3}Ba_{1/3}Ca_{1/3})_{1-x}La_xTiO₃ could maintain favorable electrical transport through La³⁺-induced carrier regulation while reducing

thermal conductivity through A-site entropy engineering and porous structures [31]. However, though the A-site doping of a single rare earth La^{3+} donor can modulate the carrier concentration and thus electrical properties, it cannot enhance chemical disorder of the lattice significantly and thus cannot further reduce thermal conductivity. Compared with single- La^{3+} substitution, co-doping of La^{3+} , Nd^{3+} , and Sm^{3+} retains the electron donor effect while introducing greater A-site ion size and mass disorder, thereby enhancing local lattice distortion and phonon scattering.

In this work, co-doping of La, Nd, and Sm rare-earth elements were used to construct a medium and high entropy perovskite system with multicomponent A-site composition, $(\text{Sr}_{1/2}\text{Ba}_{1/4}\text{Ca}_{1/4})_{1-x}(\text{La}_{1/3}\text{Nd}_{1/3}\text{Sm}_{1/3})_x\text{TiO}_3$ (SBC-RE_x; $x = 0.08\text{--}0.18$), and the effects of entropy engineering on structural evolution, defect chemistry, electrical and thermal transport, and thermoelectric performance were systematically investigated. The results show that all samples are dominated by a cubic perovskite SrTiO_3 phase (space group $Pm\text{--}3m$) with uniform elemental distribution. The introduction of multielement rare-earth cations effectively suppresses grain growth during sintering and promotes the formation of oxygen vacancies and Ti^{3+} , thereby increasing carrier concentration while maintaining moderate mobility and effective mass, ultimately leading to an enhanced power factor. In terms of thermal transport, multicomponent entropy engineering introduces multiscale defect structures, including point defects and local strain fields, which effectively strengthen phonon scattering and reduce lattice thermal conductivity in the high temperature region. Benefiting from the synergistic optimization of electrical and thermal transport, the SBC-RE_{0.14} sample achieves the highest ZT value of 0.31 at 773 K, demonstrating competitive thermoelectric performance among SrTiO_3 -based oxide thermoelectrics. These results indicate that entropy engineering of A-site cations via La, Nd, and Sm co-doping provides an effective strategy for simultaneously regulating defect chemistry, carrier transport, and phonon scattering in SrTiO_3 -based thermoelectric oxides.

2 Experimental section

2.1 Sample preparation

The raw materials used in this study included TiO_2 (Aladdin, 99.8%), SrCO_3 (Aladdin, 99.95%), BaCO_3 (Aladdin, 99.95%), CaCO_3 (Aladdin, 99.99%), La_2O_3 (Aladdin, 99.99%), Sm_2O_3 (Aladdin, 99.99%), and Nd_2O_3 (Aladdin, 99.9%). To avoid moisture absorption during weighing and mixing, all powders were pre-dried in an oven at 100 °C before use. Graphene nanoplatelets (GNPs) with an average lateral size of approximately 15 μm and a specific surface area of 40–80 $\text{m}^2 \text{g}^{-1}$ were purchased from Aladdin. The GNPs were used to prepare an oxygen-scavenging sacrificial powder bed consisting of 95 wt% SBC- RE_x powder and 5 wt% GNPs. Medium- and high- entropy perovskite ceramics with different doping levels of rare earth cations, $(\text{Sr}_{1/2}\text{Ba}_{1/4}\text{Ca}_{1/4})_{1-x}(\text{La}_{1/3}\text{Nd}_{1/3}\text{Sm}_{1/3})_x\text{TiO}_3$ ($x = 0.08\text{--}0.18$) were prepared by a solid-state reaction method. Stoichiometric amounts of the starting powders were weighed and ball-milled in anhydrous ethanol for 24 h using zirconia balls as the milling media to ensure homogeneous mixing. The resulting slurry was dried at 100 °C for 24 h. The dried mixed powders were then placed in alumina crucibles and calcined at 1473 K for 8 h to obtain SBC-RE precursor powders. After calcination, the powders were ball milled at 350 rpm for 4 h, reducing the average particle size to approximately 500 nm. The powders were then uniaxially pressed into disk-shaped pellets with a diameter of about 15 mm and a thickness of about 2.5 mm. The green pellets were sintered at 1700 K for 24 h under a reducing atmosphere of 5% H_2 + 95% Ar. To further regulate the oxygen partial pressure during sintering, the sacrificial powder bed consisting of 95 wt% SBC- RE_x powder and 5 wt% GNPs was placed around the samples without direct contact with the pellets during sintering [32]. The obtained samples were denoted as SBC- $\text{RE}_{0.08}$, SBC- $\text{RE}_{0.10}$, SBC- $\text{RE}_{0.12}$, SBC- $\text{RE}_{0.14}$, SBC- $\text{RE}_{0.16}$, and SBC- $\text{RE}_{0.18}$, corresponding to rare-earth cation contents x of 0.08, 0.10, 0.12, 0.14, 0.16, and 0.18, respectively. For comparison, the $\text{Sr}_{1/2}\text{Ba}_{1/4}\text{Ca}_{1/4}\text{TiO}_3$ (SBC) ceramic sample without rare earth cation substitution was also prepared using the identical method.

2.2 Characterization

The phase composition and crystal structure of the prepared ceramic samples were analyzed by X-ray diffraction (XRD, Rigaku SmartLab SE) using Cu K_{α} radiation over a 2θ range of 10° – 90° . Rietveld refinement was carried out using the *Jana* software to determine the lattice parameters [33]. The diffraction peak profiles were fitted with a pseudo-Voigt function, and the refinement quality was evaluated by the reliability factors R_p and R_{wp} , and the goodness of fit parameter χ^2 . Raman spectra were collected using a Raman microscope (InVia Reflex, Renishaw) equipped with a 532 nm laser excitation source over a wavenumber range of 100 – 1000 cm^{-1} . The microstructure of the bulk ceramics was characterized using a field-emission scanning electron microscope (SEM, Gemini SEM 500, Zeiss), combined with an energy-dispersive X-ray spectrometer (EDS, XFlash 6-100, Bruker) and an electron backscatter diffraction detector (EBSD, QUANTAX, Bruker) to analyze the elemental distribution, grain orientation, and crystallographic features of the samples. To obtain high quality surfaces suitable for SEM and EBSD measurements, the samples were finely polished using oxide polishing suspensions. Grain orientation and grain size were analyzed using the *Atex* software. High-angle annular dark-field scanning transmission electron microscopy (HAADF–STEM) imaging and scanning transmission electron microscopy energy dispersive X-ray spectroscopy (STEM–EDS) elemental mapping were performed using a 300 kV double spherical aberration-corrected transmission electron microscope (Spectra 300, Thermo Fisher Scientific). Geometric phase analysis (GPA) was carried out using the *Strain++* software [34]. X-ray photoelectron spectroscopy (XPS, Thermo Fisher Scientific Escalab 250Xi) was used to investigate the chemical states of the elements in the SBC–RE ceramic series.

2.3 Thermoelectric measurements

Electrical resistivity and Seebeck coefficient were measured using a thermoelectric measurement system (ZEM-3, ULVAC-RIKO) under a low-pressure helium atmosphere over a temperature range

of 300–900 K. The samples used for these measurements were bar shaped specimens with dimensions of approximately 2 mm × 2 mm × 12 mm. The Hall coefficient was measured at room temperature using a Hall effect measurement system (CH-100, CH-Magtech) based on the Van der Pauw method [35], with sample dimensions of approximately 5 mm × 5 mm × 0.5 mm. The carrier concentration n and Hall mobility μ_H were calculated according to $n = \frac{1}{eR_H}$ and $\mu_H = \frac{\sigma}{ne}$, respectively, where e is the elementary charge, R_H is the Hall coefficient, and σ is the electrical conductivity. The thermal conductivity κ was calculated using $\kappa = \rho \times D \times C_p$, where the sample density ρ was determined by the Archimedes method, while the specific heat capacity C_p and thermal diffusivity D were measured using a differential scanning calorimeter (DSC-300, Netzsch) and a laser flash apparatus (LFA-467, Netzsch), respectively.

2.4 Theoretical calculations

Detailed methods and parameters for the first-principles calculations are provided in the Supplementary Calculation Details section of the Electronic Supplementary Material.

3 Results and discussion

3.1 Physical and chemical characterization

As shown in Fig. 1(a), the SBC-RE_x ($x = 0.08$ – 0.18) with different doping levels of rare-earth cations all exhibit typical X-ray diffraction features of a perovskite structure. All the diffraction peaks can be indexed to cubic SrTiO₃ (space group $Pm\bar{3}m$), without obvious diffraction peaks from impurity phases, indicating that the multielement A-site cations are well accommodated within the perovskite lattice. To further evaluate the structural compatibility of the designed A-site multicomponent perovskite system, the Goldschmidt tolerance factor t was calculated using the following Eq. (1):

$$t = \frac{r_A + r_O}{\sqrt{2}(r_B + r_O)} \quad (1)$$

where r_A is the average ionic radius of A-site cations, r_B is the average ionic radius of B-site cations, and r_O is the ionic radius of oxygen. The calculated t values remain close to unity from 1.003 to 0.997, further confirming the structural stability of the cubic perovskite phase. Meanwhile, A-site configurational entropy (ΔS_{conf}) increases from $1.323R$ (medium entropy: $1 R \leq \Delta S_{\text{conf}} < 1.5 R$) to $1.522R$ (high entropy: $\Delta S_{\text{conf}} \geq 1.5R$) and ionic size disorder (δ_{rA}) increases from 7.24% to 7.78% with increasing x , indicating that A-site configurational disorder and ionic-size mismatch are progressively enhanced within a structurally tolerable range (Table 1, Fig. S1, Eqs. (S2) and (S3)). Further Rietveld refinement results (Fig. 1(b) and Figs. S2(a)-S2(f)) show good agreement between the calculated and experimental patterns, with relatively smooth difference curves and acceptable values of R_{wp} , R_p , and χ^2 , indicating the high reliability of the structural refinement. The lattice parameters (a) obtained from the refinement do not show a monotonic trend with increasing rare-earth cation contents (Table 1), which is likely related to differences in defects and local disorder induced by high entropy.

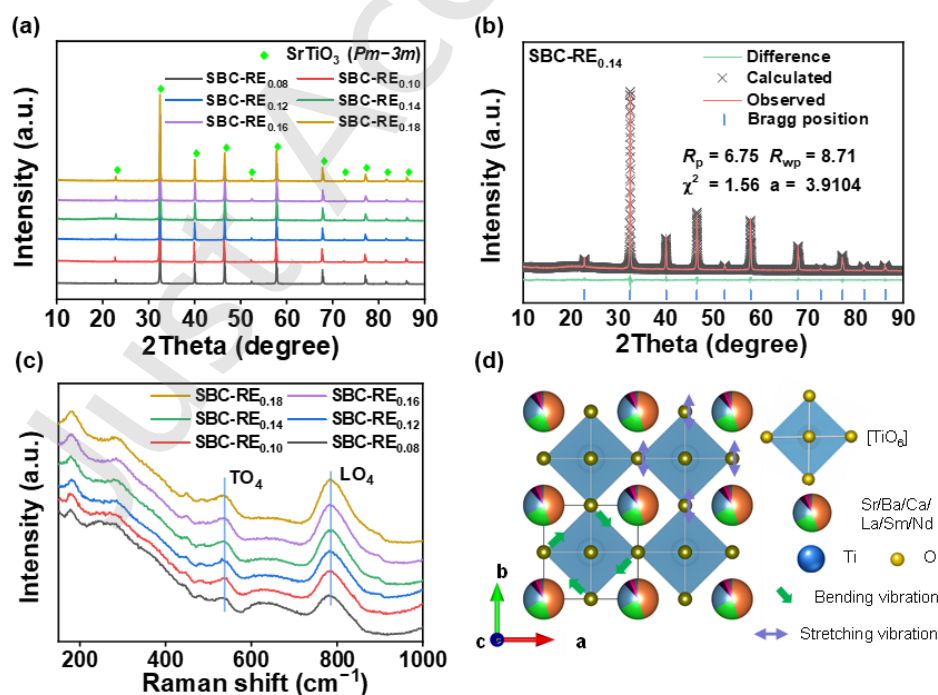


Fig. 1 Structural characterization of SBC-RE_x ceramics: (a) XRD patterns of the six annealed SBC-RE_x samples. (b) Rietveld refinement result of SBC-RE_{0.14}. (c) Room-temperature Raman spectra of the calcined SBC-RE_x powder samples. (d) Schematic illustration of the Raman vibration modes.

Table 1 Structural information of the SBC-RE_x ceramics.

Sample	$a = b = c$ (Å)	$\alpha = \beta = \gamma$ (°)	t	δ_{rA} (%)	$\Delta S_{\text{conf}}(R)$	Density (%)
SBC-RE _{0.08}	3.9113	90	1.003	7.24	1.32	97.5
SBC-RE _{0.10}	3.9129	90	1.002	7.37	1.37	97.7
SBC-RE _{0.12}	3.9112	90	1.000	7.48	1.41	98.0
SBC-RE _{0.14}	3.9104	90	0.999	7.59	1.45	98.3
SBC-RE _{0.16}	3.9102	90	0.998	7.69	1.49	99.6
SBC-RE _{0.18}	3.9114	90	0.997	7.78	1.52	99.5

As shown in Fig. 1(c), the Raman spectra of SBC-RE_x samples show two characteristic modes at approximately 542 and 760 cm⁻¹, corresponding to the TO₄ and LO₄ modes, respectively, which are associated with the bending and stretching vibrations of Ti–O–Ti bonds (Fig. 1(d)) [36,37]. The appearance of these Raman active modes indicates that multicomponent A-site doping induces local symmetry breaking in the nominally cubic lattice [37]. Compared with the calcined powders, the sintered bulk samples show smoother but broader spectral features, likely due to oxygen vacancies generated under the reducing atmosphere (Fig. S3) [36,38]. These oxygen vacancies have a spatially inhomogeneous distribution and can modify the local Ti–O bonding configuration, giving rise to diversified vibrational environments of TiO₆ octahedra and thus broadened Raman bands. Meanwhile, oxygen-vacancy-induced local strain distorts the TiO₆ octahedra and modifies the Ti–O bond lengths, locally increasing the Ti–O force constants and thereby shifting the TO₄/LO₄ modes to higher wavenumbers [39,40]. With increasing doping level of rare-earth cation, both TO₄ and LO₄ modes shift to higher wavenumbers and broaden progressively, suggesting enhanced lattice disorder. This behavior can be ascribed to the combined effects of entropy-induced chemical disorder, local lattice distortion, and complexed bond environment (mainly A–O bond-length variation and neighboring

TiO₆ octahedral distortion), which are beneficial for enhancing phonon scattering and reducing lattice thermal conductivity [41].

As shown in Figs. 2(a) and 2(b), the EBSD inverse pole figure (IPF) map and the corresponding grain boundary map of SBC-RE_{0.14} reveal the absence of preferred orientation, indicating that the grains are randomly distributed and that the SBC-RE_x composition does not induce significant texture formation (Fig. S4). Meanwhile, Fig. 2(c) shows that the average grain size decreases monotonically with the increase of doping level x , from 12.90 μm of SBC-RE_{0.08} to 5.04 μm of SBC-RE_{0.18}. This result demonstrates that multicomponent rare earth cations effectively suppress grain growth during sintering. Such grain refinement can be mainly attributed to the enhanced local lattice distortion and chemical disorder arising from the coexistence of multielement rare earth cations with different ionic radii, which hinder grain-boundary migration and thereby inhibit grain coarsening. From the BSE-SEM images in Figs. 2(d)–2(i), all samples exhibit relatively dense ceramic microstructures with clearly distinguishable grains, while several dark regions marked by white circles are identified as pores. Although a small amount of porosity remains, the overall pore fraction is limited, consistent with the relatively high bulk densities of the samples (above 95%, Table 1). In addition, the EDS elemental mapping and point analysis of SBC-RE_{0.14} show that all constituent elements are uniformly distributed throughout the matrix without obvious elemental segregation, enrichment, or depletion (Fig. S5). The measured local composition is also close to the nominal stoichiometry, indicating that the processing route enables homogeneous incorporation of the multicomponent rare earth cations and the formation of a stable solid solution.

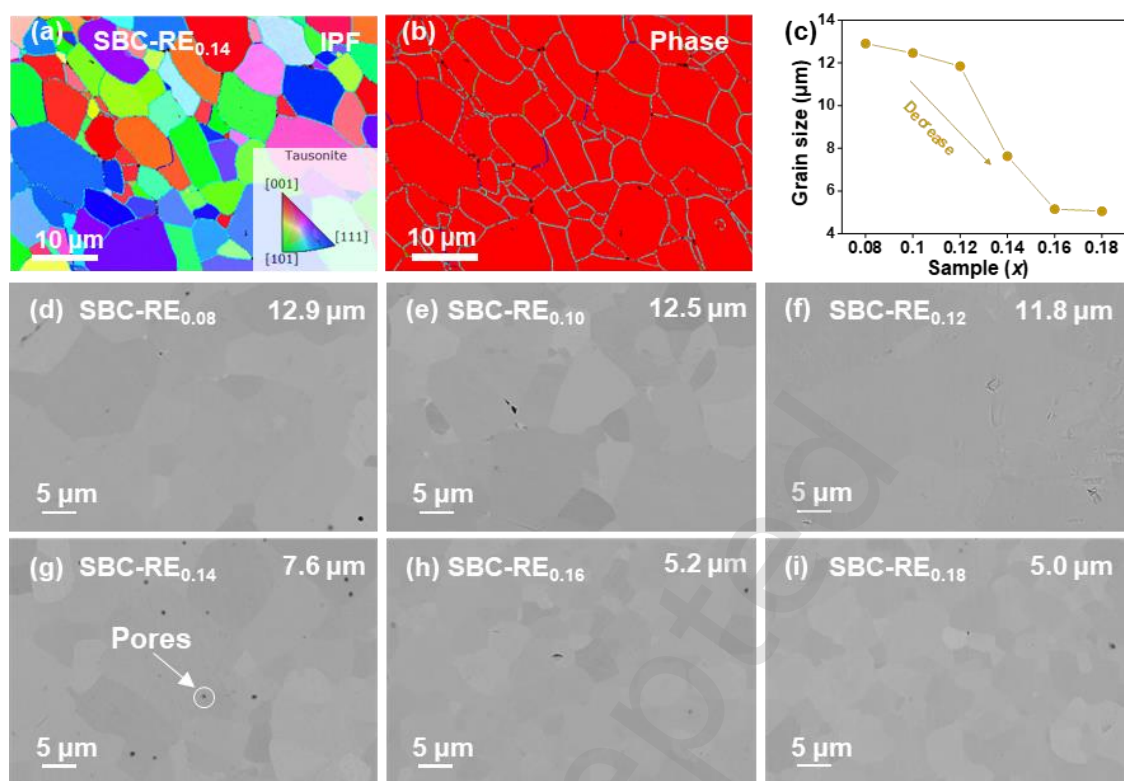


Fig. 2 Microstructure characterization of SBC-RE_x ceramics: (a) EBSD inverse pole figure (IPF) orientation map of SBC-RE_{0.14}. (b) Corresponding grain boundary map of SBC-RE_{0.14}. (c) Average grain size as a function of x . (d–i) Backscattered electron (BSE) SEM images of SBC-RE_x ceramics annealed at 1700 K for 24 h at different doping levels of (d) $x = 0.08$, (e) $x = 0.10$, (f) $x = 0.12$, (g) $x = 0.14$, (h) $x = 0.16$, and (i) $x = 0.18$, the dark regions marked by white circles indicate pores.

As shown in Fig. 3(a), STEM characterization was employed to further characterize the microstructure and atomic-scale features of the SBC-RE_{0.14} sample. The low-magnification STEM image reveals a representative ceramic grain with a dense morphology, while the enlarged image shows defect/dislocation-rich regions, indicating the presence of lattice defects within the grain (Fig. 3(b)). The atomic-resolution HAADF-STEM image exhibits a clearly ordered lattice arrangement (Fig. 3(c)), and the corresponding FFT pattern (the inset in Fig. 3(c)) confirms a well-defined perovskite crystal structure viewed along the [100] zone axis. Fig. S6 presents a HAADF-STEM image viewed along the [111] zone axis. The measured interplanar spacing of approximately 0.40 nm in Fig. 3(c) can be

assigned to the (001) planes of the SrTiO₃-based perovskite lattice. In addition, the iDPC-STEM image clearly resolves the A-site, B-site, and O atomic columns, further demonstrating that SBC-RE_{0.14} retains an ordered perovskite framework (Fig. 3(d)). As shown in Fig. 3(e), the intensity profiles extracted from the marked A-site and O-site atomic columns exhibit distinct periodic peaks, while the local intensity fluctuations suggest the possible coexistence of multielement A-site occupation and oxygen-vacancy-induced local structural disorder.

To further evaluate the local lattice distortion, GPA analysis was performed on the atomic-resolution STEM image. The resulting ϵ_{xx} , ϵ_{xy} , ϵ_{yx} , and ϵ_{yy} strain maps show pronounced spatially heterogeneous strain fluctuations, indicating that multicomponent A-site solid solution induces local lattice distortion and nonuniform strain fields (Fig. 3(f)). These local distortions can be mainly attributed to ionic-radius mismatch among the Sr, Ba, Ca, La, Nd, and Sm A-site cations. Such atomic-scale lattice distortion and strain fluctuation can act as effective phonon-scattering centers, thereby contributing to the reduction in lattice thermal conductivity [15,42]. In addition, the HAADF-STEM image and corresponding STEM-EDS elemental maps reveal the uniform distribution of Ca, Sr, Ba, La, Nd, Sm, Ti, and O throughout the observed region, confirming the successful formation of a compositionally complex multicomponent A-site-substituted SrTiO₃ lattice, as shown in Figs. 3(g) and 3(h).

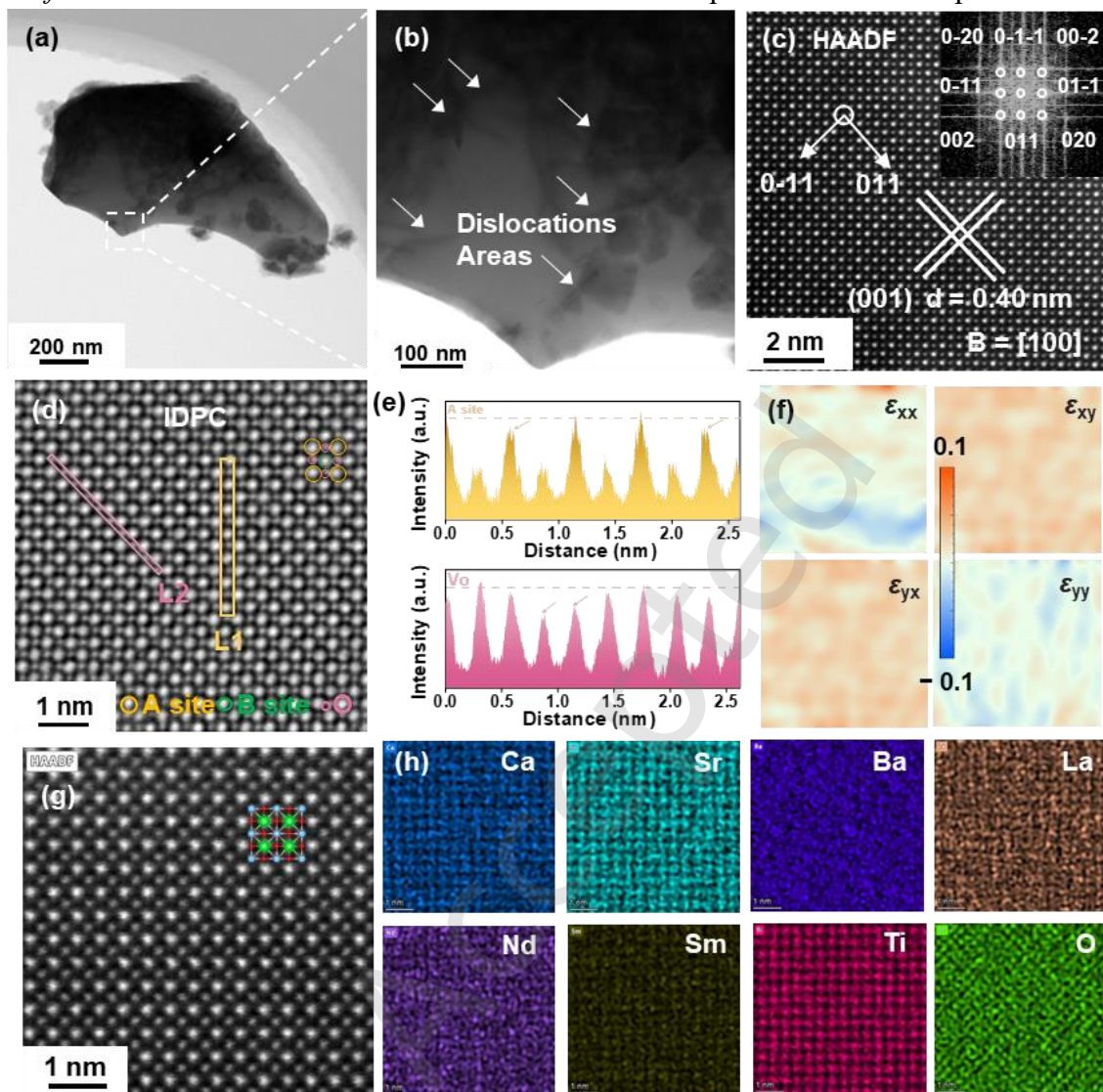


Fig. 3 STEM analysis of the SBC-RE_{0.14} sample: (a) Low-magnification TEM image of a representative ceramic grain. (b) Magnified TEM image showing local dislocation-rich regions. (c) Atomic-resolution HAADF-STEM image and corresponding FFT pattern, revealing a well-defined perovskite lattice viewed along the [100] zone axis. (d) IDPC-STEM image displaying the A-site, B-site, and O atomic columns; L1 and L2 indicate the selected regions for intensity-profile analysis. (e) Intensity profiles extracted from the A-site and O-site atomic columns marked in d). (f) GPA-derived strain maps of ϵ_{xx} , ϵ_{xy} , ϵ_{yx} , and ϵ_{yy} , indicating local lattice distortion and strain fluctuation. (g) Atomic-resolution HAADF-STEM image of the SBC-RE_{0.14}, with the inset showing the local perovskite atomic configuration. (h) STEM-EDS elemental maps of Ca, Sr, Ba, La, Nd, Sm, Ti, and O, showing their uniform distribution in the observed region.

3.2 Electrical transport properties

Fig. 4(a) displays the temperature-dependent Seebeck coefficients of all samples, which remain negative throughout the measured temperature range, indicating a typical electron-dominated *n*-type conduction behavior. The absolute value of Seebeck coefficient gradually increases with increasing temperature, consistent with the transport characteristics of *n*-type degenerate semiconductors. At 300 K, the Seebeck coefficient generally decreases with increasing doping level of rare-earth cations, from $-114.32 \mu\text{V K}^{-1}$ for SBC-RE_{0.08} to $-57.20 \mu\text{V K}^{-1}$ for SBC-RE_{0.18}. As displayed in Fig. 4(b), the electrical conductivity shows an overall increasing trend with increasing content of rare earth cations at 300 K, rising from 112.63 S cm^{-1} for SBC-RE_{0.08} to 284.97 S cm^{-1} for SBC-RE_{0.18}. The overall improvement in electrical conductivity with increasing rare-earth cations can be mainly attributed to the donor effect of trivalent rare-earth ions, which introduce additional electron carriers upon substitution of the Sr^{2+} cations at the A site, thereby enhancing the electronic transport capability of the material.

With increasing temperature, the electrical conductivity first increases, reaches a maximum at approximately 500–600 K, and then gradually decreases. This behavior suggests that the transport mechanism evolves from thermally activated conduction at low temperatures to degenerate-semiconductor-like transport at higher temperatures [43]. Similar temperature-dependent transport tendencies have been reported in SrTiO_3 -based thermoelectric materials, where high carrier concentrations can lead to metallic or degenerate-semiconductor-like transport behavior [32,44]. In the low-temperature region, the increase in electrical conductivity with temperature indicates that carrier transport is mainly governed by thermal activation. By contrast, the gradual decrease in conductivity at elevated temperatures implies enhanced carrier scattering, with acoustic phonon scattering

becoming dominant. Among all samples, SBC-RE_{0.18} exhibits the highest electrical conductivity of 803.16 S cm⁻¹ at 550 K.

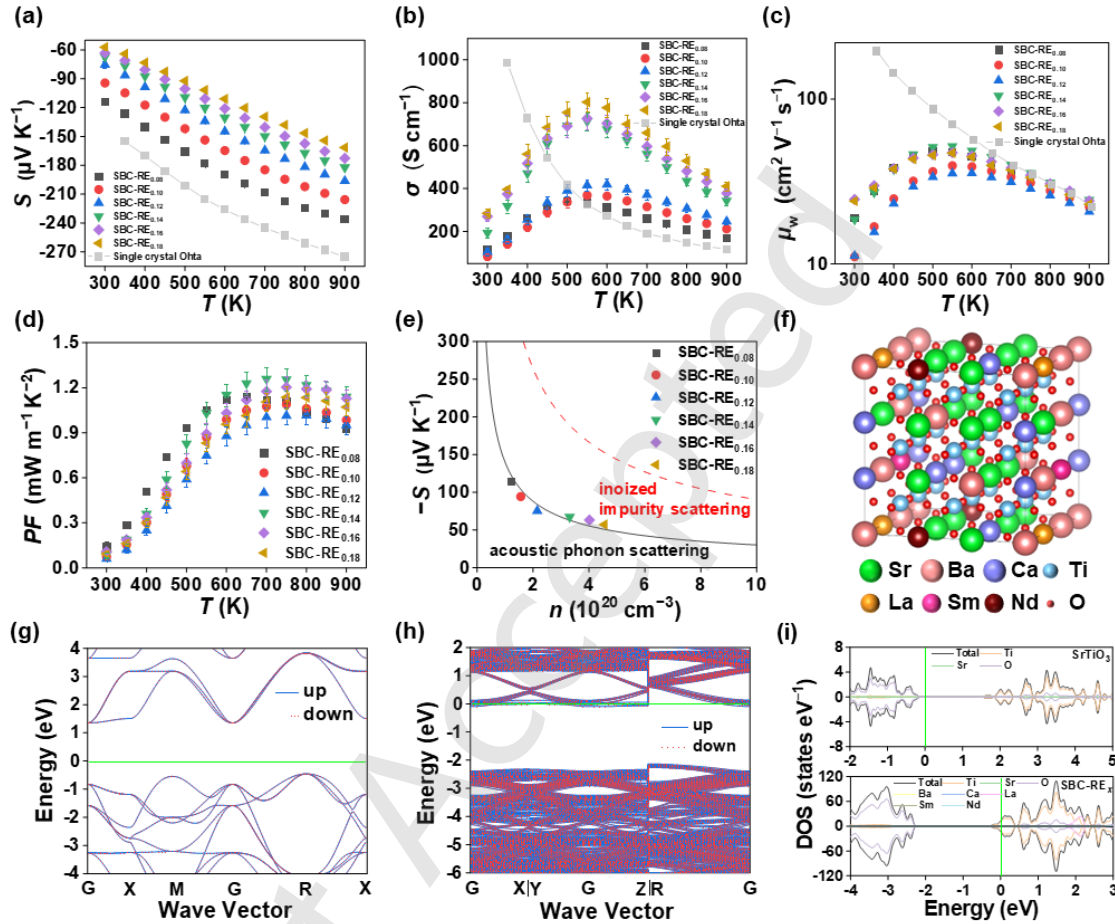


Fig. 4 Electrical transport properties and electronic-structure calculations of the SBC-RE_x samples: Temperature-dependent (a) Seebeck coefficient, (b) electrical conductivity and (c) weighted mobility μ_w , the data of single-crystal SrTiO₃ were taken from literature [43]. (d) Temperature dependent power factor. (e) Hall carrier concentration–Seebeck coefficient relationship, showing the influence of different carrier scattering mechanisms on S . (f) Crystal structure of the SBC-RE model (Sr₁₂Ba₆Ca₆La₁Nd₁Sm₁Ti₂₇O₈₁) used for DFT calculation. (g) and (h) Calculated electronic band structures of pristine SrTiO₃ and the representative SBC-RE model, respectively. (i) Projected density of states (PDOS) of pristine SrTiO₃ and the SBC-RE model.

To further understand the low-temperature carrier transport behavior, Arrhenius fitting of $\ln(\sigma T)$ versus $1000/T$ was performed over the temperature range of 300–500 K, yielding activation energies of 0.096–0.126 eV for the different samples (Eqs. (S4), (S5) and Fig. S8(a)), which are consistent with the ~ 0.12 eV activation-energy scale reported in literature for reduced La-doped SrTiO_3 [45]. This indicates reduced carrier transport barriers and helps explain the enhanced electrical conductivity of the SBC-RE_x ceramics. Considering the relatively weak temperature dependency of carrier concentration in SrTiO_3 -based materials [23], the temperature-dependent mobility was estimated using $\mu_H = \sigma/(en)$ with the room-temperature Hall carrier concentration, where μ_H is the Hall carrier mobility, σ is the electrical conductivity, e is the elementary charge, and n is the Hall carrier concentration. The estimated μ_H values exhibit a nonmonotonic temperature dependency, increase with temperature roughly at 300–500 K and then decrease at higher temperatures (Fig. S8(b)). In addition to μ_H , the carrier transport property was further evaluated using the weighted mobility μ_w , which is estimated from the measured S and σ using the Drude–Sommerfeld-based relation (Eq. (S6) and Fig. 4(c)) [44,46]. Consistent with the temperature dependency of μ_H , the μ_w values for each SBC-RE_x sample first increases and then decreases with increasing temperature. This nonmonotonic behavior arises from thermally activated carrier transport across grain-boundary barriers and localized defect states at low temperatures, followed by the dominance of phonon scattering at higher temperatures, which suppresses carrier mobility [32,43]. The μ_w value of $\text{SBC-RE}_{0.14}$ reaches a maximum of $51.83 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ at 550 K; when the temperature is above 600 K, μ_w gradually approaches the mobility of single-crystal SrTiO_3 , suggesting that the carrier transport behavior in the high-temperature region becomes increasingly governed by intrinsic lattice scattering. Based on the measured Seebeck coefficient and electrical conductivity, the temperature-dependent power factor was calculated to evaluate the overall electrical transport performance. As shown in Fig. 4(d), the power factor first increases and then decreases with increasing doping level of rare-earth cations. Among all samples, $\text{SBC-RE}_{0.14}$ exhibits the highest power factor, reaching $1.26 \text{ mW m}^{-1} \text{ K}^{-2}$ at 700 K. Moreover, compared with the SBC

ceramic sample without rare earth cation doping ($x = 0$), the enhanced PF and electrical conductivity of SBC-RE_{0.14} confirm the beneficial electron carrier modulation effect of La/Nd/Sm substitution (Figs. S7(a–c)).

Oxygen vacancies are intrinsic defects in SrTiO₃-based ceramics and can be further generated during sintering under a reducing atmosphere. They play an important role in thermoelectric transport by donating electrons for n -type conduction and serving as phonon scattering centers. The formation of oxygen vacancies can be expressed by Eq. (2) [47]:



indicating that each oxygen vacancy contributes to the generation of electron carriers. In SBC-RE ceramics, the oxygen vacancy concentration is closely related to the Ti³⁺ content through the charge compensation relationship (Eq. (3)):

$$[\text{Ti}'_{\text{Ti}}] \approx [\text{RE}^\bullet_{\text{Sr}}] + [\text{V}_\text{o}^{\bullet\bullet}] \quad (3)$$

where RE represents the incorporated rare-earth cations (La, Nd, and Sm). Accordingly, XPS analysis of the Ti 2p spectra was performed to clarify the defect evolution (Fig. S9(b) and Table S1). The coexistence of Ti⁴⁺ and Ti³⁺ is observed in SBC-RE_{0.08} and SBC-RE_{0.14}, with the Ti⁴⁺ peaks located at 458.10 eV (2p_{3/2}) and 463.87 eV (2p_{1/2}), and the Ti³⁺ peaks located at 456.30 eV (2p_{3/2}) and 462.30 eV (2p_{1/2}) [25], confirming the partial reduction of Ti⁴⁺ during sintering. The Ti³⁺/Ti ratio increases from 3.23% for SBC-RE_{0.08} to 4.34% for SBC-RE_{0.14} with increasing rare-earth cation content, which can be rationalized by donor-type charge compensation induced by trivalent RE³⁺ substitution at the divalent A site, together with oxygen loss under the reducing sintering atmosphere. Both processes can supply electrons that occupy the Ti 3d orbital, thereby promoting the formation of Ti³⁺ species [48,49]. Consistently, Hall measurements show that the carrier concentration increases from $1.22 \times 10^{20} \text{ cm}^{-3}$ for SBC-RE_{0.08} to $4.55 \times 10^{20} \text{ cm}^{-3}$ for SBC-RE_{0.18}. Meanwhile, μ_H decreases overall from $5.76 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ for SBC-RE_{0.08} to $3.91 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ for SBC-RE_{0.18}, with a partial recovery at $x = 0.14$ – 0.18 .

This recovery may reflect the improved grain-boundary transport at higher doping levels (carrier concentrations), which partly offset grain boundary barrier [32]. Local structural changes may also contribute to the recovery of mobility, as reduced Ti off-centering via entropy-engineering has been shown to improve TiO_6 symmetry and carrier transport in SrTiO_3 [14]. Nevertheless, all samples remain dominated by acoustic phonon scattering over the entire measured temperature range, with a scattering factor of $r = 0$ (Fig. 4(e), Table 2, and Eq. (S7)) [50]. Furthermore, the carrier effective mass (m^*) was estimated using the single parabolic band (SPB) model based on the measured Seebeck coefficient and the carrier concentration. Specifically, the reduced chemical potential ξ was first obtained from the Seebeck relation using Eqs. (4) and (5) [51,52] :

$$S = \frac{k_B}{e} \left(\frac{(r+2)F_{r+1}(\xi)}{(r+1)F_r(\xi)} - \xi \right) \quad (4)$$

where the Fermi integral is given by

$$F_r(\xi) = \int_0^\infty \frac{x^r}{1 + e^{x-\xi}} dx \quad (5)$$

the extracted ξ was then substituted into Eq. (6) to determine m^* .

$$n = 4\pi \left(\frac{2m^*k_B T}{h^2} \right)^{\frac{3}{2}} F_{\frac{1}{2}}(\xi) \quad (6)$$

The calculated effective mass remains within a relatively narrow range of approximately 1.3–1.7 m_e (Table 2), indicating that the band structure does not undergo a drastic change with increasing rare-earth content. Among all samples, SBC-RE_{0.14}, which exhibits the highest power factor, shows a well-balanced combination of transport parameters, including a moderate carrier concentration ($3.31 \times 10^{20} \text{ cm}^{-3}$), mobility ($3.64 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$), and effective mass ($1.58 m_e$). These results indicate that SBC-RE_{0.14} achieves a more favorable balance among carrier concentration, mobility, and band transport characteristics, thereby leading to an enhanced power factor.

Table 2 Transport properties of SBC-RE_x ceramics at room temperature

Sample	S ($\mu\text{V K}^{-1}$)	σ (S cm^{-1})	n (10^{20} cm^{-3})	μ_{H} ($\text{cm}^2 \text{ V}^{-1} \text{ s}^{-1}$)	m^*/m_e
SBC-RE _{0.08}	− 114.32	112.63	1.22	5.76	1.47
SBC-RE _{0.10}	− 94.37	82.92	1.56	3.32	1.38
SBC-RE _{0.12}	− 75.42	105.24	2.15	3.06	1.33
SBC-RE _{0.14}	− 67.29	193.15	3.31	3.64	1.58
SBC-RE _{0.16}	− 63.22	269.94	4.02	4.19	1.69
SBC-RE _{0.18}	− 57.20	284.97	4.55	3.91	1.65

To further understand the effect of multicomponent A-site substitution on electronic transport, first-principles calculations were performed for pristine SrTiO₃ and a representative SBC-RE model. The pristine SrTiO₃ model was constructed using a $1 \times 1 \times 1$ unit cell (Fig. S10), while the representative SBC-RE model was constructed based on a $3 \times 3 \times 3$ supercell with a nominal composition of Sr₁₂Ba₆Ca₆La₁Nd₁Sm₁Ti₂₇O₈₁, corresponding to $x \approx 0.11$ in the designed SBC-RE system (Fig. 4(f)). The calculated band structures of pristine SrTiO₃ and the representative SBC-RE model are shown in Figs. 4(g) and 4(h), respectively. For pristine SrTiO₃, the band structure exhibits a clear band gap, with the Fermi level located below the conduction band, indicating its limited intrinsic carrier concentration. In comparison, the band structure of the representative SBC-RE model shows that the Fermi level shifts into the conduction band, suggesting an increased electron carrier concentration. This result helps explain the experimentally observed increase in electrical conductivity and the decrease in the absolute Seebeck coefficient after multicomponent rare-earth substitution. The projected density of states (PDOS) results are shown in Fig. 4(i). For pristine SrTiO₃, the conduction band is mainly derived from Ti-3d orbitals, while the valence band is dominated by O-2p orbitals, consistent with the typical electronic structure of SrTiO₃-based perovskite oxides [53,54]. For the representative SBC-RE model, the electronic states near the conduction band edge are still mainly associated with Ti-3d orbitals, while La-5d-, Nd-5d-, and Sm-5d-related states also contribute to the conduction-band region. The valence band remains primarily dominated by O-2p orbitals.

3.3 Thermal transport properties

Fig. 5(a) presents the total thermal conductivity (κ) of the SBC-RE_x ceramics. Consistent with the thermal diffusivity (D) trend (Fig. S11(a)), κ gradually decreases with increasing temperature for all samples. Compared with the literature thermal conductivity values of pristine SrTiO₃ (8.61 W m⁻¹ K⁻¹ at ~300 K) and La_{0.08}Sr_{0.9}TiO_{3- δ} (7.53 W m⁻¹ K⁻¹ at ~320 K) [32,55], the SBC-RE_x ceramics exhibit substantially reduced κ values (4.26–4.93 W m⁻¹ K⁻¹) at room temperature, indicating that multicomponent A-site substitution plays a decisive role in establishing intrinsically low thermal transport in the SBC-RE_x system. Nevertheless, the κ shows a slight increasing trend with increasing rare-earth-cation content at 300 K. For example, κ increases from 4.26 W m⁻¹ K⁻¹ for SBC-RE_{0.08} to 4.93 W m⁻¹ K⁻¹ for SBC-RE_{0.18} at 300 K, corresponding to an increase of approximately 16% (Fig. 5(a)). This enhancement is closely related to the increased electronic thermal conductivity (κ_{ele}) [56]. The κ_{ele} was estimated using the Wiedemann–Franz law, $\kappa_{\text{ele}} = L \sigma T$, where the Lorenz number L (Fig. S11(b)) was calculated according to the following Eq. (7) [57]:

$$L = 1.5 + \exp\left(-\frac{|S|}{116}\right) \quad (7)$$

As shown in Fig. 5(b), the variation of κ_{ele} is consistent with that of the electrical conductivity, with an increasing trend at higher doping level of rare earth cations. This indicates that increasing the rare-earth doping level x can enhance electrical conductivity but also unfavorably increases the electronic contribution to thermal conduction.

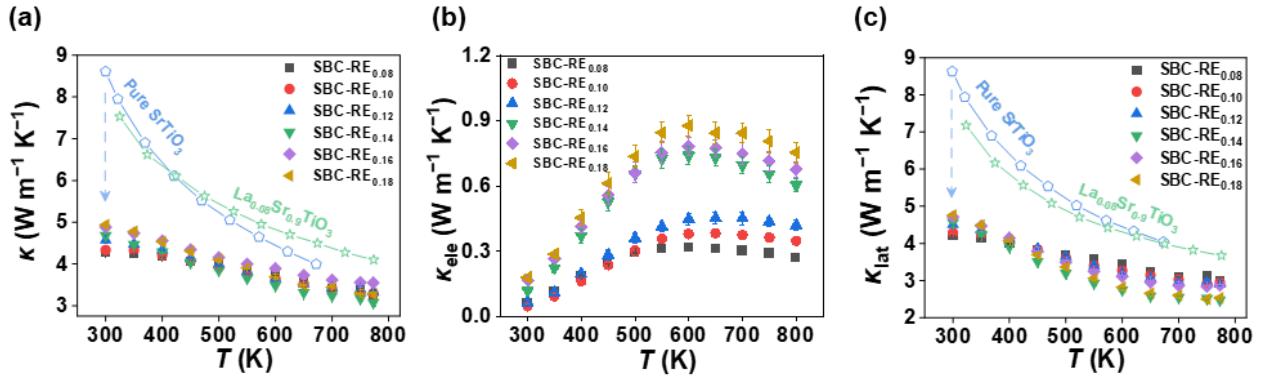


Fig. 5 Thermal transport properties results of the SBC-RE_x ceramics: (a) Total thermal conductivity, (b) electronic thermal conductivity, and (c) lattice thermal conductivity of the SBC-RE_x samples, thermal conductivities of two SrTiO₃-based thermoelectric oxides (SrTiO₃ and La_{0.08}Sr_{0.9}TiO₃) reported in literature [32,55] are plotted in (a) and (c) as references.

Nevertheless, lattice thermal conductivity (κ_{lat}) remains the dominant contribution to the total thermal conductivity and can be suppressed effectively by entropy engineering of the A-site cations (Fig. 5(c)). Compared with pure SrTiO₃ (8.60 W m⁻¹ K⁻¹ at ~300 K) and La_{0.08}Sr_{0.9}TiO_{3- δ} (7.18 W m⁻¹ K⁻¹ at ~320 K), the SBC-RE_x samples show substantially reduced κ_{lat} values (~4.5 W m⁻¹ K⁻¹ at ~300 K), indicating that multicomponent A-site substitution effectively suppresses lattice heat transport [32,55]. Direct comparison of the κ_{lat} of SrTiO₃, SBC and SBC-RE_{0.14} was conducted to decouple the effects of Sr/Ba/Ca and La/Nd/Sm substitution in reducing thermal conductivity (Fig. S7(e)), the results indicate that Sr/Ba/Ca A-site substitution can effectively reduce lattice conductivity from 8.6 to ~4.5 W m⁻¹ K⁻¹ at 300 K, while La/Nd/Sm substitution can further reduce thermal conductivity at high temperature range. In particular, at 773 K, compared with the $x = 0$ SBC reference, the κ_{lat} of SBC-RE_{0.14} is reduced by 26.5%, dropping from 3.44 to 2.53 W m⁻¹ K⁻¹ (Figs. S7(d,e)). In addition, as shown in Fig. 5(c), at 773 K, κ_{lat} of SBC-RE_x can be further suppressed by increasing doping level, for instance, SBC-RE_{0.14} (2.53 W m⁻¹ K⁻¹) is smaller than that of SBC-RE_{0.08} (2.95 W m⁻¹ K⁻¹), corresponding to an approximately 14% reduction. With the increase of RE doping content from 0.08 to 0.18, the slopes extracted from plots of $\ln \kappa_{lat}$ versus $\ln T$ (Fig. S11(c)) generally becomes more

negative, changing from -0.36 to -0.76 , suggesting the recovery of Umklapp scattering dominated temperature dependency of κ_{lat} ($\kappa_{\text{lat}} \propto T^{-1}$) [58]. Note that pure SrTiO_3 has a relationship of $\kappa_{\text{lat}} \propto T^{-0.93}$ (Fig. S11(c)), which is very close to the ideal Umklapp scattering mechanism. This change of temperature dependency of κ_{lat} with entropy involves complex lattice dynamics and requires further study to be understood.

According to the Debye Callaway model, κ_{lat} can be expressed by Eq. (8) [59]:

$$\kappa_{\text{lat}} = \frac{k_{\text{B}}}{2\pi^2 v} \left(\frac{k_{\text{B}} T}{\hbar} \right)^3 \int_0^{\theta_{\text{D}}/T} \frac{\tau_{\text{c}} x^4 e^x}{(e^x - 1)^2} dx \quad (8)$$

where v , θ_{D} , τ_{c} , and $\hbar$ denote the average sound velocity, Debye temperature, total phonon relaxation time, and reduced Planck constant, respectively. As a key parameter describing phonon transport, τ_{c} comprehensively reflects the suppressing effect of multiple scattering centers on phonon propagation. According to Matthiessen's rule, the total scattering rate τ_{c}^{-1} can be expressed as the sum of the scattering rates associated with different mechanisms, as shown in Eq. (9) [19].

$$\tau_{\text{c}}^{-1} = \tau_{\text{U}}^{-1} + \tau_{\text{N}}^{-1} + \tau_{\text{PD}}^{-1} + \tau_{\text{GB}}^{-1} + \tau_{\text{EP}}^{-1} \quad (9)$$

here, the frequency dependences of the individual scattering processes are given as follows: Umklapp (U) scattering follows $\tau_{\text{U}}^{-1} \propto \omega^2$, normal (N) scattering follows $\tau_{\text{N}}^{-1} \propto \omega^2$, point defect (PD) scattering follows $\tau_{\text{PD}}^{-1} \propto \omega^4$, grain boundary (GB) scattering is approximately frequency independent, i.e., $\tau_{\text{GB}}^{-1} \propto \omega^0$, and electron phonon (EP) scattering typically follows $\tau_{\text{EP}}^{-1} \propto \omega^2$, where τ^{-1} represents the phonon scattering rate and ω represents the phonon angular frequency [60]. Semi-quantitative Debye–Callaway analysis of $\text{SBC-RE}_{0.14}$ further suggests that point-defect scattering dominates the additional suppression of κ_{lat} , while grain-boundary and electron–phonon scattering make relatively smaller contributions (Fig. S12). For the medium and high entropy $(\text{Sr}_{1/2}\text{Ba}_{1/4}\text{Ca}_{1/4})_{1-x}(\text{La}_{1/3}\text{Nd}_{1/3}\text{Sm}_{1/3})_x\text{TiO}_3$ ($x = 0.08\text{--}0.18$) system investigated in this work, increasing the doping level of multielement rare-earth cations intensifies the atomic mass fluctuation, ion size mismatch, and chemical disorder, thereby

creating more abundant scattering centers in the lattice, including point defects, local strain fields, and grain boundary related defects. These multiscale defective structures can effectively enhance phonon scattering and shorten the phonon relaxation time τ_c , thereby suppressing lattice heat transport and reducing κ_{lat} . In addition, the coexistence of A-site cations with different ionic radii leads to a broader distribution of local A–O bond lengths and induces distortion of the neighboring TiO_6 octahedra. Such local bond-length and force-constant fluctuations further strengthen the scattering of mid- to high-frequency phonons, which is beneficial for achieving lower lattice thermal conductivity in the high temperature region [41,61].

Fig. 6(a) shows the temperature-dependent ZT values of the SBC-RE_x samples. The ZT values of all samples increase continuously with temperature, indicating a favorable thermoelectric response in the medium to high temperature range. Among them, SBC-RE_{0.14} exhibits the best overall thermoelectric performance, achieving a peak ZT of approximately 0.31 at 773 K. This improvement mainly originates from the synergistic optimization of electrical transport and suppression of thermal transport. Appropriate multicomponent doping with La^{3+} , Nd^{3+} , and Sm^{3+} effectively regulates the carrier concentration, mobility, and effective mass, thereby maintaining a high power factor. At the same time, the enhanced lattice disorder effectively suppresses phonon-mediated heat transport, enabling a favorable balance between electrical and thermal transport in SBC-RE_{0.14}. The thermoelectric conversion efficiency (η) is an important indicator for evaluating the application potential of thermoelectric materials. The theoretical maximum conversion efficiency (η_{max}) of a thermoelectric generator can be estimated using the following Eq. (10) [62]:

$$\eta_{\text{max}} = \frac{T_{\text{H}} - T_{\text{C}}}{T_{\text{H}}} \times \frac{\sqrt{1 + ZT_{\text{ave}}} - 1}{\sqrt{1 + ZT_{\text{ave}}} + T_{\text{C}}/T_{\text{H}}} \quad (10)$$

where T_{H} and T_{C} are the hot side and cold side temperatures, respectively, and ZT_{ave} is the average thermoelectric figure of merit over the temperature range from T_{C} to T_{H} . When T_{C} is fixed at 300 K,

the $\eta_{\max}$ of SBC-RE_{0.14} increases with increasing T_H and ZT_{ave} , reaching approximately 3.0% at 773 K (Fig. 6(b)), suggesting its potential for medium to high temperature thermoelectric conversion.

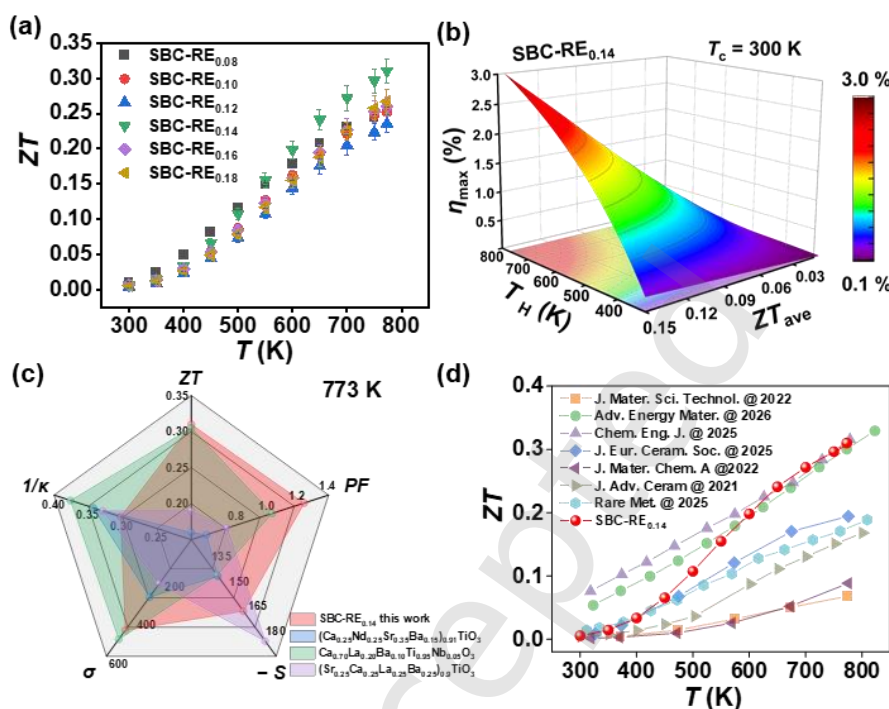


Fig. 6 Overall thermoelectric performance of SBC-RE_x ceramics: (a) Temperature-dependent ZT values of SBC-RE_x samples. (b) Calculated maximum thermoelectric conversion efficiency $\eta_{\max}$ of SBC-RE_{0.14} as a function of the hot-side temperature T_H and the engineering figure of merit ZT_{ave} , with the cold side temperature fixed at $T_c = 300$ K. (c) Radar-chart comparison of ZT , PF , $-S$, σ , and $1/\kappa$ at 773 K between SBC-RE_{0.14} and representative previously reported n -type medium- and high-entropy SrTiO₃ based thermoelectric ceramics [15,39,63]. (d) Comparison of temperature-dependent ZT values between SBC-RE_{0.14} and previously reported n -type medium- and high-entropy SrTiO₃-based thermoelectric ceramics [15,19,27,39,64–66].

Fig. 6(c) compares the key thermoelectric parameters of SBC-RE_{0.14} with those of reported n -type medium and high entropy titanate thermoelectric ceramics at 773 K [15,39,63]. The SBC-RE_{0.14} maintains favorable electrical transport while exhibiting reduced thermal conductivity at 773 K, it has a Seebeck coefficient of $-163 \mu\text{V K}^{-1}$, an electrical conductivity of 467 S cm^{-1} , and a power factor of

1.24 mW m⁻¹ K⁻², together with a relatively low thermal conductivity of 3.08 W m⁻¹ K⁻¹. These results demonstrate a synergistic optimization between electrical and thermal transport. Fig. 6(d) further compares the ZT values of this work with those of reported n -type medium and high entropy SrTiO₃-based thermoelectric ceramics [15,19,27,39,64–66]. The SBC-RE_{0.14} shows competitive ZT values over the measured temperature range and reaches approximately 0.31 at 773 K. These results indicate that multicomponent rare-earth doping at the A-site can enhance phonon scattering and reduce thermal conductivity while maintaining favorable electrical transport, providing an effective strategy for improving the thermoelectric performance of SrTiO₃-based oxide ceramics.

4 Conclusions

In summary, we systematically investigated the synergistic regulation of electron and phonon transport in SrTiO₃-based thermoelectric perovskites through A-site entropy engineering. A series of multicomponent medium and high entropy ceramics, (Sr_{1/2}Ba_{1/4}Ca_{1/4})_{1-x}(La_{1/3}Nd_{1/3}Sm_{1/3})_xTiO₃ (SBC-RE_x; $x = 0.08$ – 0.18), were successfully synthesized by a solid-state reaction method. All samples maintained a cubic SrTiO₃ perovskite structure with homogeneous elemental distribution, indicating that the multicomponent A-site cations were effectively incorporated into the lattice. The co-doping of La³⁺, Nd³⁺, and Sm³⁺ introduced donor type substitution and promoted the formation of Ti³⁺ species, thereby increasing carrier concentration while maintaining a favorable balance among carrier mobility and effective mass. As a result, the optimized SBC-RE_{0.14} sample achieved a maximum power factor of 1.26 mW m⁻¹ K⁻². Meanwhile, the multicomponent A-site occupation induced atomic mass fluctuation, ionic-size mismatch, local lattice distortion, strain fields, and grain refinement, which effectively strengthened phonon scattering and suppressed lattice thermal conductivity. Benefiting from this coordinated optimization of electrical and thermal transport, SBC-RE_{0.14} reached a peak ZT of 0.31 at 773 K. This work demonstrates that rational A-site entropy engineering is an effective strategy for balancing carrier transport and phonon scattering in SrTiO₃-based oxide thermoelectrics.

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Author contributions

Hui Zhang: Conceptualization, Data curation, Investigation, Methodology, Software, Validation, Visualization, Writing – original draft, and Writing – review & editing. **Jianyun Cao:** Conceptualization, Data curation, Formal analysis, Funding acquisition, Investigation, Methodology, Project administration, Resources, Supervision, Validation, Visualization, and Writing – review & editing. **Yifan Sun:** Data curation and Software. **Yu Tan:** Data curation and Software. **Liping Hao:** Formal analysis, Funding acquisition, Investigation, Methodology, and Validation. **Jiyang Xie:** Formal analysis and Resources. **Guoliang Chen:** Formal analysis, Investigation, Methodology, Software, and Validation. **Yaming Wang:** Methodology, Resources, and Validation. **Wanbiao Hu:** Conceptualization, Formal analysis, Funding acquisition, Investigation, Methodology, Project administration, Resources, Supervision, Validation, Visualization, and Writing – review & editing. All authors reviewed and approved the final manuscript.

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.

Electronic Supplementary Materials

Supplementary materials (supplementary calculation details, theoretical calculation details, Figures. S1–S12, and Table S1) are available in the online version of this article.

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