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REVIEW ARTICLE

A review on structure–property relationships in dielectric ceramics using high-entropy compositional strategies

Deqin Chen[1] | **Xiaowei Zhu**[1] | **Xinrong Yang**[1] | **Na Yan**[1] | **Yongbao Cui**[1] | **Xiuyun Lei**[1] | **Laijun Liu**[1] | **Jibran Khaliq**[2] | **Chunchun Li**[1,3,4]

[1]Guangxi University Key Laboratory of Non-ferrous Metal Oxide Electronic Functional Materials and Devices, College of Material Science and Engineering, Guilin University of Technology, Guilin, China

[2]Department of Mechanical and Construction Engineering, Faculty of Engineering and Environment, Northumbria University, Newcastle, Newcastle upon Tyne, UK

[3]Guangxi Key Laboratory of Embedded Technology and Intelligent System, Guilin University of Technology, Guilin, China

[4]Electronic Materials Research Laboratory, Key Laboratory of the Ministry of Education and International Center for Dielectric Research, Xi'an Jiaotong University, Xi'an, Shaanxi, China

Correspondence
Chunchun Li, Guangxi University Key Laboratory of Non-ferrous Metal Oxide Electronic Functional Materials and Devices, College of Material Science and Engineering, Guilin University of Technology, Guilin 541004, China.
Email: lichunchun2003@126.com

Jibran Khaliq, Department of Mechanical and Construction Engineering, Faculty of Engineering and Environment, Northumbria University, Newcastle, Newcastle upon Tyne, NE1 8ST, UK.
Email: jibran.khaliq@northumbria.ac.uk

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Abstract

High-entropy strategy is a design that uses multiple elements to increase configurational entropy. As an emerging research field, high-entropy was originally applied to metals, but it has shown great potential in inorganic nonmetallic materials. Since the first discovery of colossal dielectric permittivity in high-entropy ceramics, it indicates that the high-entropy strategy is feasible in dielectric ceramics. The multi-ion solid solution will bring a broader space for the material structure and property tailoring. With the research of domestic and foreign scholars in high-entropy dielectrics, we believe that entropic engineering can effectively regulate dielectric properties. The review paper explores the potential of the high-entropy strategy in dielectric ceramics. It provides a detailed overview of the structural properties and the potential benefits of utilizing high-entropy materials in dielectric ceramics. The review also covers the current research advancements in the field and provides insights into future directions for further development of high-performance dielectric ceramics.

KEYWORDS
dielectric materials, high-entropy ceramics, permittivity, structure–property relationship

1 | INTRODUCTION

To keep up with the demands of scientific and technological advancements, novel advanced ceramics with superior properties and size reduction are crucial. The majority of the ceramics are based on one single composition. Different kinds of dopants are added to the base material to enhance its properties. Recently, high-entropy mixing emerged as a design strategy that is making waves across the materials science community.[1–3] High-entropy ceramics are a new and emerging class of materials that have gained significant attention in recent years. These materials are defined as solid solutions formed by the presence of five or more elements occupying one or more Wyckoff sites. However, with advancements in research, the definition of high-entropy ceramics has expanded to include materials with multicomponent designs occupying multiple lattice sites. This has allowed for the exploration and development of a wide range of high-entropy ceramics, including oxides,[4,5] nitrides,[6] carbides,[7] and borides.[8,9] High-entropy ceramics differ from traditional ceramics in their composition, as they contain a high degree of disorder and randomness.

One of the key characteristics of high-entropy ceramics is their unique properties, which are a result of the combination of different elements with varying physical and chemical properties. These properties include improved thermal stability, enhanced mechanical properties, and increased wear resistance.[10–14] Furthermore, high-entropy ceramics exhibit improved chemical stability,[15] as the presence of multiple elements helps to prevent the formation of undesirable phases. The formation of high-entropy ceramics is facilitated by the use of a thermodynamic approach, where the Boltzmann formula[16] is used to connect the number of microscopic states (ω) to entropy as shown in Equation (1). In thermodynamics, entropy represents the disorder in materials, and the configurational entropy (S_c) can be calculated using the formula in Equation (2).[17] The configurational entropy, in turn, influences the properties of high-entropy ceramics and determines their unique characteristics:

$$S = k \ln \omega \tag{1}$$

$$S_c = -R \sum_{i=1}^{n} x_i \ln (x_i) \tag{2}$$

High-entropy ceramics have potential applications in various fields, such as energy conversion and storage,[18,19] catalysis,[20,21] and biomedicine.[22] In the field of electronics, high-entropy dielectric ceramics have been investigated for their potential in dielectric materials for capacitors, substrates, and other electronic components.[23–25] Their unique compositions and structures offer opportunities for developing new materials with enhanced electrical and thermal properties.

In energy conversion and storage, high-entropy ceramics can be used as electrodes in lithium-ion batteries, due to their improved stability and higher voltage windows. In catalysis, high-entropy ceramics exhibit improved activity and selectivity compared to traditional catalysts due to their high surface area and tunable electronic structure, making them a promising material for environmental applications such as hydrogen evolution and carbon dioxide reduction.[26] In biomedicine, high-entropy ceramics can be used as scaffolds for tissue engineering, due to their biocompatibility and improved mechanical properties.

Pyroelectric materials can generate electric charge upon temperature changes, making them valuable in energy harvesting and sensing applications. High-entropy dielectric ceramics exhibit excellent thermal stability and can be tailored to possess desirable pyroelectric properties. Research by Zhang et al.[27] investigated the pyroelectric properties of high-entropy dielectric ceramics and their potential for energy conversion. The results indicated 10 times increase in pyroelectric effect compared to lead-based materials indicating the potential of lead-free pyroelectric materials synthesized using high-entropy strategy.

Furthermore, high-entropy dielectric ceramics have shown promise in other fields, such as thermoelectric materials, energy storage devices, and electronic applications. Their unique combination of properties, including high dielectric constant, low dielectric loss, and excellent thermal stability, makes them suitable for these applications. Research by Zheng et al.[28] demonstrated the potential of high-entropy dielectric ceramics in thermoelectric devices, with improved thermoelectric performance compared to traditional materials. Similarly, studies have explored their application in energy storage devices, such as capacitors, where their high dielectric constant and low dielectric loss contribute to improved energy storage capabilities.[29]

According to the S_c value, the system can be divided into low-entropy ($S_c \leq 1R$), medium-entropy ($1R < S_c \leq 1.5R$), and high-entropy ($S_c > 1.5R$),[23] as shown in Figure 1. However, the rate of increase in configurational entropy slows down with increasing the number of components. When the entropy (ΔS) of the system increases, ΔG decreases, which is beneficial to stabilize the phase structure, as shown in the following equation[30]:

$$\Delta G = \Delta H - T\Delta S \tag{3}$$

Like high-entropy alloys, high-entropy ceramics have four special effects. High-entropy effect: The high-entropy

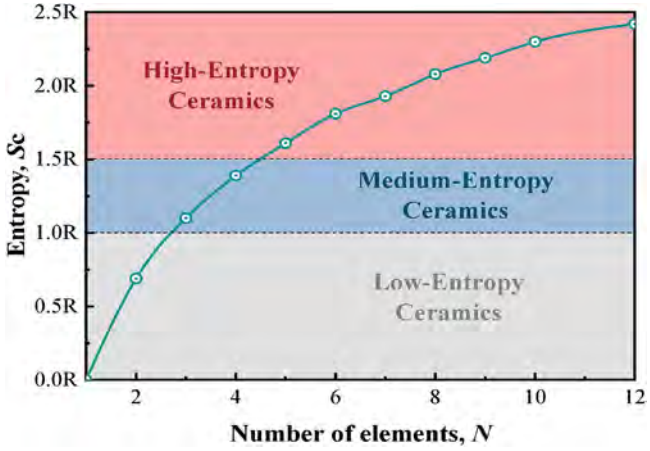

FIGURE 1 Relationship between configuration entropy and the number of components (in equal). The maximum configuration entropy S_c is obtained by an equal ratio of various components. *Source*: Ref. [31]. Reproduced with permission of Elsevier.

effect arises from the high configurational entropy in multicomponent mixtures with nearly equal molar ratios. This effect favors the formation of a single-phase solid solution, where multiple elements are uniformly distributed throughout the lattice. The high configurational entropy stabilizes the solid solution, resulting in a homogeneous microstructure and improved mechanical and functional properties.[32] Sluggish diffusion effect: The presence of multiple elements in high-entropy ceramics hinders the synergistic diffusion of components. The sluggish diffusion effect leads to slower diffusion rates, which can influence various properties, including thermal conductivity, electrical conductivity, and magnetic behavior. This effect can be advantageous in certain applications, such as thermoelectric materials, where low thermal conductivity is desired to enhance efficiency.[33,34] Lattice distortion effect: The random arrangement of multiple components in the lattice sites of high-entropy ceramics induces lattice distortion. This distortion can lead to increased lattice strain, dislocations, and slip systems, which can enhance mechanical properties such as hardness, strength, and fracture toughness. The lattice distortion effect provides a mechanism for improving mechanical performance beyond what is achievable in traditional single-component ceramics.[35,36]

Lastly, synergy in components (cocktail effect): The combination of different elements in high-entropy ceramics can result in synergistic interactions, also known as the "cocktail" effect. These interactions can lead to unexpected performance enhancements or mutations in material properties. The synergy between elements in high-entropy ceramics can give rise to new phase formations, improved electrical conductivity, altered optical properties, or enhanced catalytic activity. This effect opens up exciting opportunities for tailoring material properties by carefully selecting and combining elements.[20,37] These unique effects enable high-entropy ceramics to overcome the limitations of traditional materials, and the complex compositions of high-entropy ceramics provide more opportunities for improving material properties.[33] By understanding and harnessing these effects, high-performance ceramics with enhanced mechanical, thermal, electrical, and functional properties can be designed and developed which hold great promise for further advancements in material science and technology.

Currently, the use of high-entropy ceramics is concentrated on the thermal barrier, environmental barrier, friction resistance, corrosion resistance, and other thermodynamic materials. The high-entropy effect ensures a homogeneous distribution of multiple elements, reducing the propensity for phase separation and improving the stability of thermal barrier coatings coating. Additionally, the lattice distortion effect contributes to the ability of high-entropy ceramics to withstand thermal cycling and mechanical stresses, thus enhancing the durability ability to withstand mechanical stresses and reducing the likelihood of abrasive wear.[3,9,38,39] Research on its dielectric properties is still in its early stages, although the number of publications on high-entropy dielectric is increasing (Figure 2). As the high-entropy strategy was applied to ceramic in 2015, high-entropy ceramic shows an excellent potential in energy storage, piezoelectric, ferroelectric, and microwave dielectric materials. Surprisingly, the local ion disorder from the interaction among multiple ions exhibits a multiple polarized configuration, efficiently. The weakened long-range polarization order enhances the polarization flexibility under the external field, which result in improved piezoelectricity. In addition, the construction of inhomogeneous nanophase region and local polymorphic distortion by multi-ion solid solution has been proven to be an effective approach to improve the relaxation of ferroelectric materials. The above explains the unique charm of the high-entropy strategy in dielectric material tailoring. Figure 3 presents statistics on the number of publications from 2015 to 2022 using the keywords "high entropy, dielectric" and "high entropy, ferroelectric." In 2015, the publications containing the keywords "high entropy, dielectric" and "high entropy, ferroelectric" are 0. The numbers in 2022 increased to 36 and 7, respectively. The number increasing in published papers indicates that high-entropy strategies in electroceramics have drawn great attention in research.

One of the key advantages of high-entropy ceramics in dielectric materials is the potential for enhanced dielectric permittivity. The high configurational entropy resulting

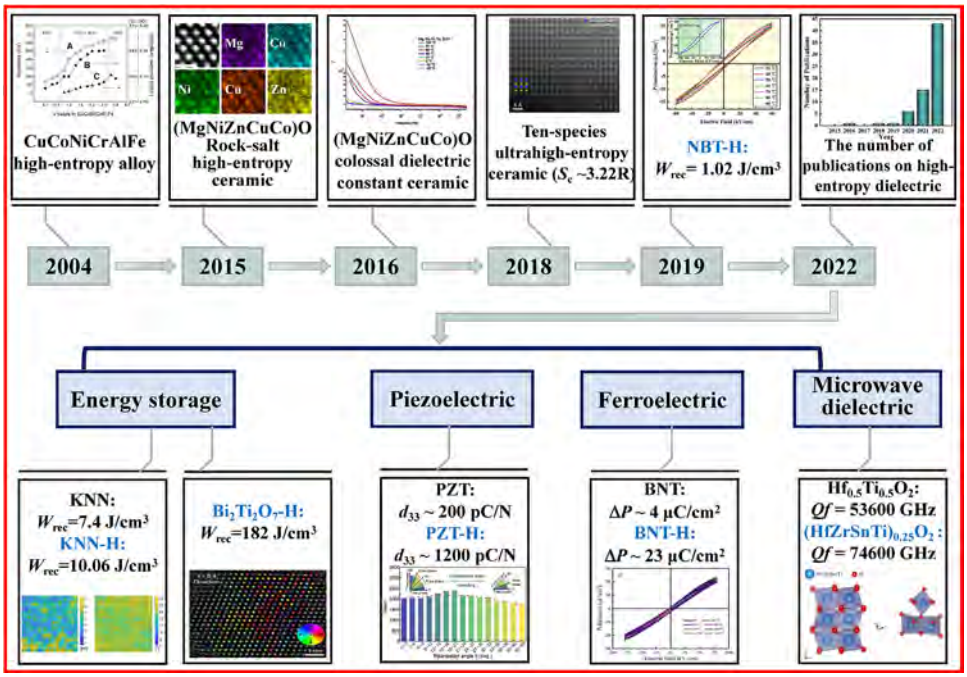

FIGURE 2 Historic development of high-entropy ceramic (KNN, PZT and BNT-H represents the high-entropy material). *Source*: Refs. [1], [2], [24], [25], [29], [40–44]. Reproduced with permission of Springer, Elsevier, and Wiley.

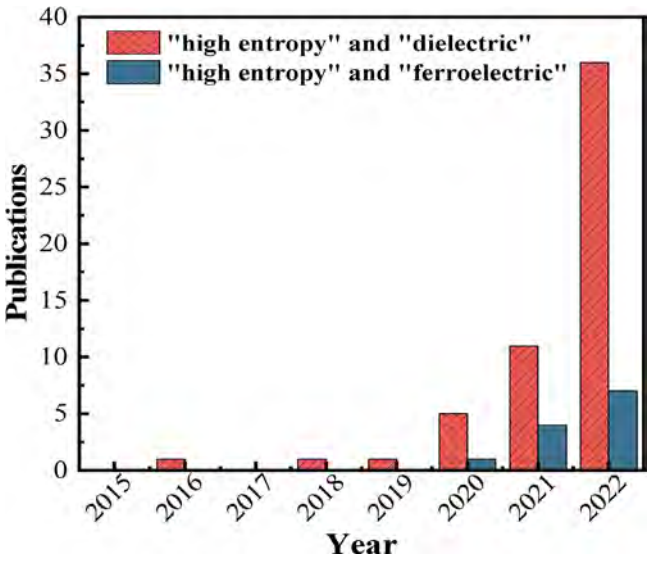

FIGURE 3 Annual number of publications on high-entropy electroceramics in the refereed journals from 2015 to 2022 (collected from Web of Science Core Collection with the keywords "high entropy, ceramic," "high entropy, dielectric," and "high entropy, ferroelectric").

from the presence of multiple elements in equimolar or near-equal molar ratios leads to a highly disordered atomic arrangement.[45] This disorder contributes to a more uniform distribution of charges throughout the material, facilitating greater polarization. As a result, high-entropy ceramics can exhibit significantly higher dielectric permittivity compared to traditional ceramics. The ability to tune the dielectric permittivity is of great importance in various applications, such as capacitors, energy storage devices, and dielectric resonators.[46–49] Additionally, the disordered atomic arrangement and lattice distortion effects in high-entropy ceramics can contribute to the reduction of dielectric loss. The random distribution of multiple elements and the resulting lattice distortion can impede the propagation of phonons and charge carriers, leading to decreased energy dissipation and improved dielectric loss characteristics. This reduction in dielectric loss is particularly advantageous in high-frequency applications where minimizing energy dissipation is essential, such as in telecommunications, microwave devices, and integrated circuits.[29,42] Additionally, the presence of multiple elements in the material can lead to the creation of bandgap states, which can be utilized for a variety of electronic and photonic applications proving that the high-entropy strategy is feasible.[1] In dielectric material, according to the examples mentioned in Figure 2, they are the competitive candidate materials used in dielectric capacitors, energy harvesters, dielectric pulses, resonators, and so on. This review aims to fill this gap by introducing and presenting the research progress of high-entropy dielectric materials in energy storage, ferroelectric, and microwave dielectric ceramics. The review explores the relationship between structure and properties in high-entropy oxide materials and discusses their potential for future development. Figure 4 provides an outline of the review's content.

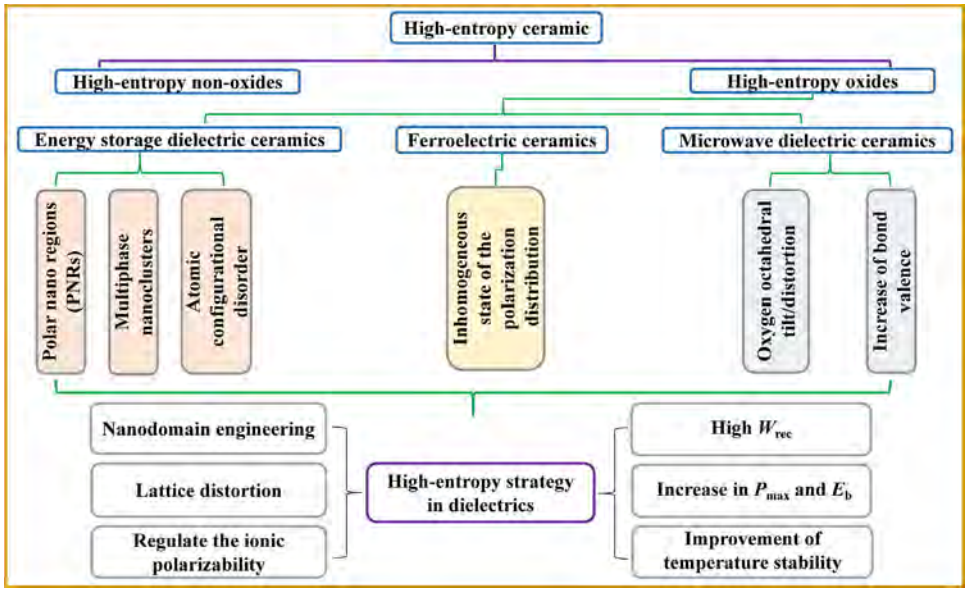

FIGURE 4 Outline of this review.

2 | ENERGY STORAGE DIELECTRIC CERAMICS

Dielectric capacitors are being utilized as pulse power devices in microelectronic, medical, aerospace, and other fields due to their high energy density, fast charging and discharging speed, and excellent chemical stability.[50–52] The energy storage density and efficiency of the dielectric material can be calculated using the following equations:

$$W = \int_0^{P_{max}} E\mathrm{d}P \tag{4}$$

$$W_{rec} = \int_{P_r}^{P_{max}} E\mathrm{d}P \tag{5}$$

$$\eta = \frac{W_{rec}}{W} \tag{6}$$

where W is the total energy storage density of the material that includes recyclable energy (W_{rec}) (density and lost energy density), η is the energy storage efficiency, E is the external electric field strength, P is the polarization strength, P_{max} is the maximum polarization strength, and P_r is the residual polarization strength. It is necessary to have small residual polarization P_r, large polarization P_{max}, and high breakdown field strength E to obtain high energy storage density and efficiency. By reducing P_r to a minimum, the dielectric material can effectively store and release energy without significant losses. This is particularly important for applications requiring efficient energy storage and rapid charging and discharging capabilities such as batteries.

Dielectric capacitors can be categorized into two types based on the relationship between dielectric permittivity and electric field strength: linear dielectrics and nonlinear dielectrics. For linear dielectric, the polarization strength changes linearly with the increase of electric field strength without hysteresis. However, the energy storage density is constrained by the low saturated polarization strength. Nonlinear dielectrics, on the other hand, exhibit a more complex relationship between polarization and electric field strength. Typical nonlinear dielectrics include paraelectrics, ferroelectrics, relaxor ferroelectrics, and antiferroelectrics (double relaxor ferroelectrics), with their respective hysteresis loops as shown in Figure 5. The choice between linear and nonlinear dielectrics depends on the potential response speed, and stability requirements. The hysteresis loops indicate their unique properties, which helps to create tailored design and optimization of dielectric capacitors for respective applications.

Among them, the relaxor ferroelectrics have low residual polarization and dielectric loss while retaining high saturation polarization and breakdown field strength, making them potential energy storage dielectric materials.[54] The unique feature of high-entropy materials, highly disordered cations, or anions, provides advantages for the design of the relaxor characteristics.

2.1 | Research status of high-entropy energy storage dielectric ceramics

High-entropy ceramics has shown enormous potential in dielectric energy storage and a significant reduction in loss tangent. The loss tangent remained remarkably low

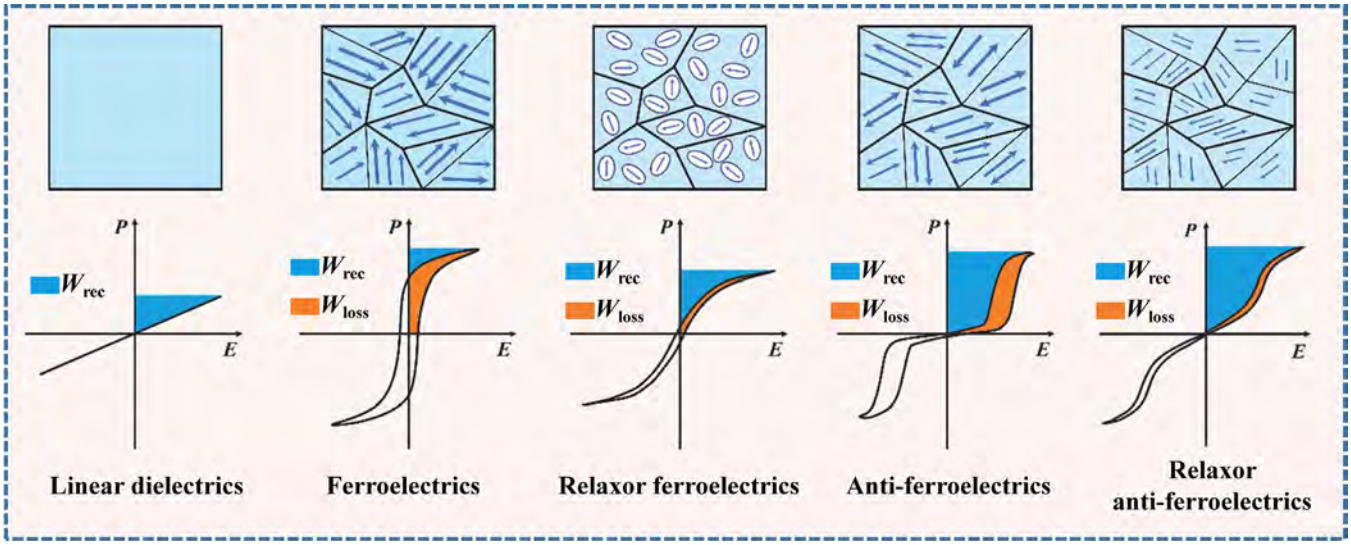

FIGURE 5 Domain structures and hysteresis loops of different dielectrics. *Source*: Ref. [53]. Reproduced with permission of RSC.

(<0.015) from room temperature to 125°C, attributed to the presence of polar nano-regions with a short relaxation time resulting from the high atomic disorder induced by the high-entropy effect in the A-sites.[55] Pu et al.[41,56,57] successfully synthesized $(Na_{0.2}Bi_{0.2}Ba_{0.2}Sr_{0.2}Ca_{0.2})TiO_3$ high-entropy ceramics and investigated their energy storage properties. Results indicate that the discharge energy density achieves 1.02 J/cm^3 under the electric field strength of 145 kV/cm, which is improved to 1.56 J/cm^3 by incorporating 0.5 wt% Mn^{2+}. The addition of ions with various sizes and valences to form solid solutions increases the disorder of the ions, which has an impact on the long-range ordered state of the ferroelectric domain to a certain extent. As a result, the nonhomogeneous micro-region appears, and the dispersion coefficient (γ) of the ceramics is 1.72, which enhances the relaxor properties of the ferroelectrics.

Bismuth titanate-based ceramics have attracted interest due to their high dielectric constant, large remnant polarization, and excellent energy storage properties.[58,59] However, one of the challenges associated with bismuth titanate ceramics is their high loss tangent, which limits their efficiency and performance in energy storage applications.[60–62] For example, $(Na_{0.5}Bi_{0.5})TiO_3$ (BNT) ferroelectric ceramics have large coercivity field (E_c), low resistivity (ρ), and low depolarization temperature (T_d).[63,64] To this end, high-entropy design is introduced through multi-cation incorporation in the A-site of BNT to instruct polar nano-regions (PNRs). Ferroelectric materials with the PNRs show greater polarization strength (P_{max}) and smaller residual polarization (P_r), making them competitive candidates for energy storage properties and random-access memory.[65,66] Similarly, the multi-ion solid solution provides highly disordered structural characterization, it is the key to breaking macroscopic ferroelectric domains and improving relaxor phenomenon.

Additionally, high-entropy strategy was also applied in $BaTiO_3$ and $Bi_{1.5}ZnNb_{1.5}O_7$ to enhance the energy storage characteristics. By introducing doped cations with random occupation, these high-entropy compositions produced various nonhomogeneous structures that had promising properties for energy storage applications. The energy storage density, breakdown field strength, and energy storage efficiency of high-entropy ceramics are summarized in Table 1.

In a recent study, Chen et al.[29] employed a high-entropy strategy to design a unique composition $[(K_{0.2}Na_{0.8})_{0.8}Li_{0.08}Ba_{0.02}Bi_{0.1}](Nb_{0.68}Sc_{0.02}Hf_{0.08}Zr_{0.1}Ta_{0.08}Sb_{0.04})O_3$ which exhibited multiphase nanoclusters and randomly aligned oxygen octahedral tiling in by (as shown in Figure 6). This high-entropy strategy disrupted the long-range ferroelectric ordering through R–O–T–C multiphase nanoclusters. This resulted in PNRs with small sizes, giving rise to a high energy storage density of up to 10.06 J/cm^3 with high efficiency of 90.8%.[75] Similarly, the addition of $Bi(Li_{0.2}Y_{0.2}Mg_{0.2}Ti_{0.2}Ta_{0.2})O_3$ ceramic into $(0.9Ba(Ti_{0.97}Ca_{0.03})O_3–0.1Bi_{0.55}Na_{0.45}TiO_3)$ ceramic to synthesize (0.9BTC–0.1BNT–BLYMTT) resulted in high energy storage density (4.89 J/cm^3 at 240 kV/cm) and energy storage efficiency (91.2%).[76] Another design concept aimed to disrupt the long-range ordered ferroelectric state by introducing an enhanced local random field was utilized in $(Bi_{0.5}Na_{0.5})(Ti_{1-2x}Fe_xNb_x)O_3$ ($x = 1/3$) ceramic using $NaNbO_3$, $(Bi_{0.5}Na_{0.5})TiO_3$, and $BiFeO_3$ (the design concept is shown in Figure 7). This disruption resulted in the formation of smaller PNRs and significantly increased the maximum polarization strength P_{max}.[77] The weakly coupled PNRs, presence of polymorphic oxygen octahedra,

TABLE 1 Energy storage density, breakdown field strength, and energy storage efficiency of the high-entropy dielectric.

System	W_{rec} (J/cm^3)	E_b (kV/cm)	η (%)	Ref.
$Ba(Zr_{0.2}Ti_{0.2}Sn_{0.2}Hf_{0.2}Nb_{0.2})O_3$	0.55	290	–	67
$Ba(Zr_{0.2}Ti_{0.2}Sn_{0.2}Hf_{0.2}Ta_{0.2})O_3$	0.68	370	–	67
$(Bi_{0.2}Na_{0.2}K_{0.2}Ba_{0.2}Ca_{0.2})TiO_3$	0.68	129	87.5	68
$(Bi_{0.2}Na_{0.2}K_{0.2}La_{0.2}Sr_{0.2})TiO_3$	0.96	180	90.0	69
$(Bi_{0.2}Na_{0.2}K_{0.2}La_{0.2}Sr_{0.2})(Ti_{0.8}Sc_{0.2})O_3$	1.09	220	80.0	49
$(Bi_{0.2}Na_{0.2}Ba_{0.2}Ca_{0.2}Sr_{0.2})TiO_3$	1.37	127	72.7	70
$(Na_{0.2}Bi_{0.2}Ca_{0.2}Sr_{0.2}Ba_{0.2})TiO_3$	1.32	110	91.0	24
$(Bi_{0.2}Na_{0.2}Ba_{0.2}Ca_{0.2}Sr_{0.2})TiO_3$–0.5 wt% $MnCO_3$	1.56	160	70.2	57
$Bi_{1.5}Zn_{0.75}Mg_{0.25}Nb_{0.75}Ta_{0.75}O_7$	2.72	650	91.0	71
$0.88(Bi_{0.4}Na_{0.2}K_{0.2}Ba_{0.2})TiO_3$–$0.12Sr(Mg_{1/3}Nb_{2/3})O_3$	3.37	310	90.8	72
0.8 $Na_{0.5}Bi_{0.5}TiO_3$–$0.2Sr(Zr_{0.2}Sn_{0.2}Hf_{0.2}Ti_{0.2}Nb_{0.2})O_3$	3.52	283	73.4	73
$0.9(0.75BaTiO_3$-$0.25Na_{0.5}Bi_{0.5}TiO_3)$–$0.1Bi(Zn_{0.2}Mg_{0.2}Al_{0.2}Sn_{0.2}Zr_{0.2})O_3$	3.74	273	82.2	74

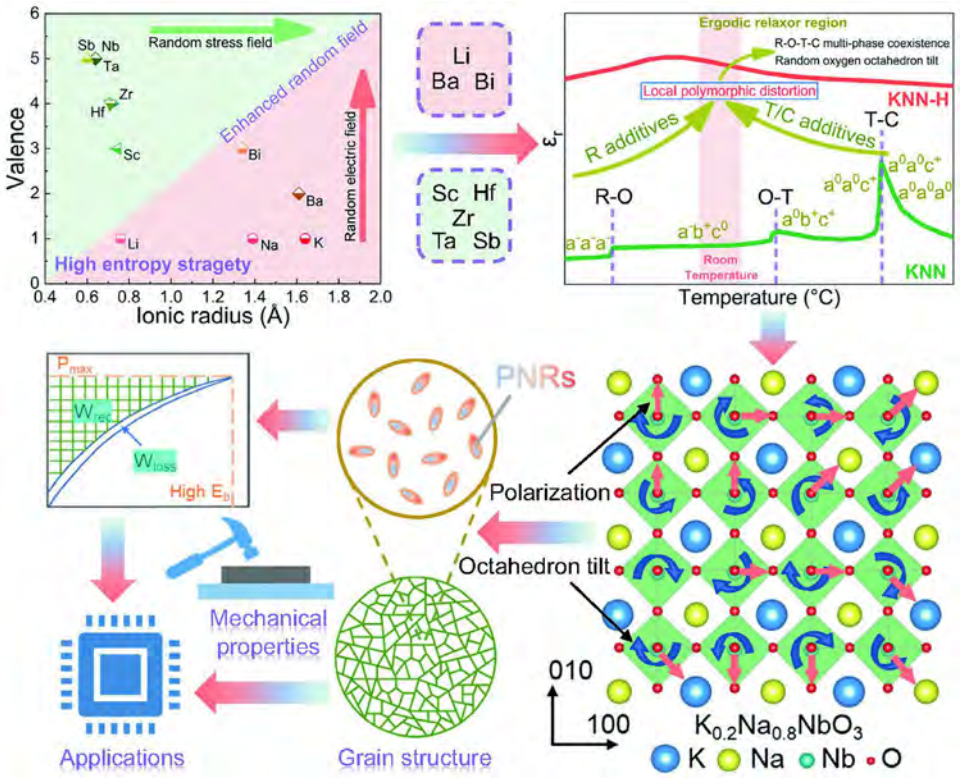

FIGURE 6 Schematic diagram of high-entropy design for $K_{0.2}Na_{0.8}NbO_3$ strategy for local polymorphic distortion and giant energy storage performance. *Source*: Ref. [29]. Reproduced with permission of Springer.

and higher grain boundary density collectively contributed to a higher breakdown field strength E_b. Furthermore, the long-range ordered ferroelectric state transformed to a macroscopic nonpolar state in an unloaded electric field, resulting in a near-zero P_r. The combination of these characteristics led to a giant energy storage density of 13.8 J/cm^3 and large efficiency of 82.4%.

High-entropy strategy was also employed to thin films through which $(Bi_{3.25}La_{0.75})(Ti_{3-3x}Zr_xHf_xSn_x)O_{12}$ (x = 0.4) film forms a stabilized pyrochlore $Bi_2Ti_2O_7$-based dielectric films. By carefully controlling the atomic configurational entropy, the film exhibited favorable and stable microstructural characteristics. This was evident from the presence of lattice distorted nanocrystalline grains and a disordered amorphous-like phase as shown in Figure 8.[42] As a result, the breakdown strength was enhanced resulting in overall stability and durability of the film under high electric field conditions. Furthermore,

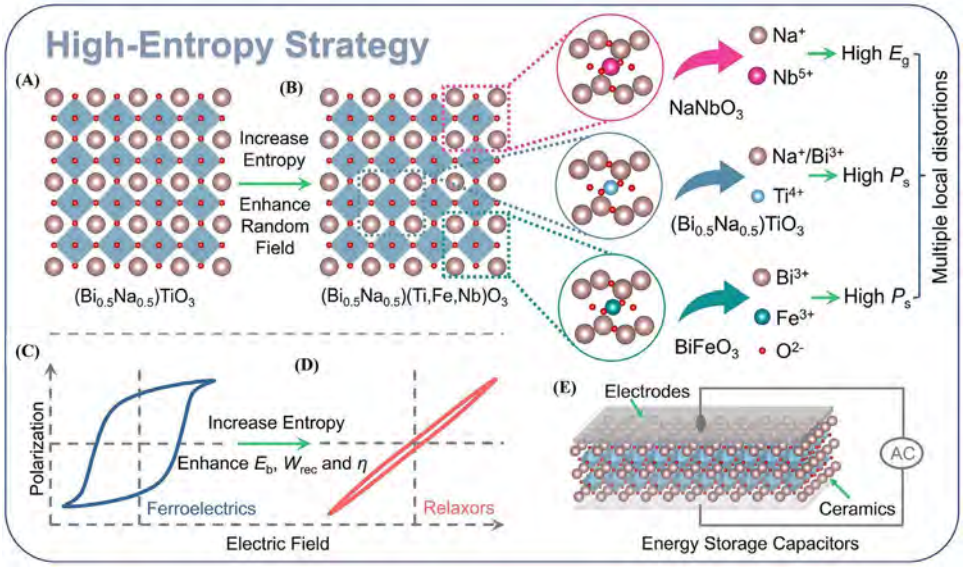

FIGURE 7 The design concept of $(Bi_{0.5}Na_{0.5})(Ti_{1-2x}Fe_xNb_x)O_3$ high-entropy ceramic. The structure of BNT (A) and BNTFN (B). *P–E* loops of BNT (C) and BNTFN (D). (E) Schematic diagram of energy storage capacitors. *Source*: Ref. [77]. Reproduced with permission of Springer.

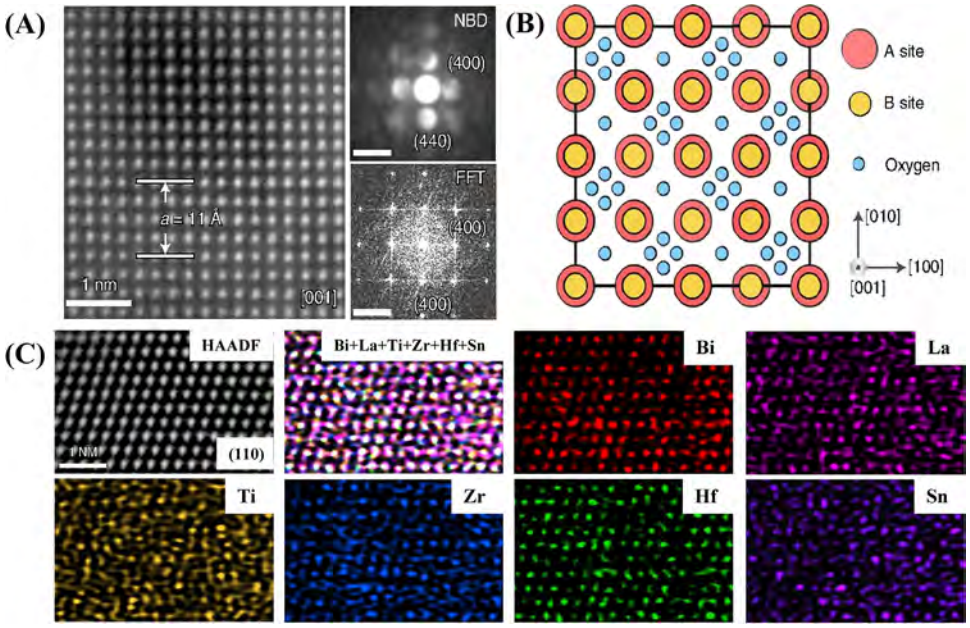

FIGURE 8 Structure evolution (A, B) and TEM–EDS mapping (C) with the increase of entropy. *Source*: Ref. [42]. Reproduced with permission of Springer.

the presence of the disordered amorphous-like phase and lattice-distorted nanocrystalline grains reduced the polarization switching hysteresis allowing the film to exhibit a more rapid and reversible polarization response. The combination of these effects enhanced the energy storage performance with a high energy density value of 182 J/cm^3 and achieved a storage efficiency of 78%.

The introduction of cations with varying sizes and valence states into parent ceramics exhibits a high degree of structural disorder resulting in high-entropy ceramics. This disorder creates a favorable environment for the formation of relaxor behavior, characterized by a broadened and frequency-dependent dielectric response. The resulting high-entropy effect, from the random distribution of multiple cations, improves the breakdown field

strength because of the long-range order of the crystal lattice, leading to the formation of local fields. Additionally, polarization strength and temperature stability were also improved due to the presence of multiple cations and the associated lattice distortion. This resulted in a more stable dielectric response over a wide temperature range which is essential for the reliable performance of dielectric energy storage materials under varying operating conditions.

3 | FERROELECTRIC CERAMICS

High-entropy ferroelectric ceramics have recently emerged as a promising approach to enhance ferroelectric properties. The doping of multiple ions and their occupation in different positions in the lattice can induce local structural and compositional changes, resulting in improved ferroelectric properties such as Curie temperature and electromechanical response.

In addition to high-entropy strategies, researchers have also explored other approaches to improve the performance of ferroelectric ceramics. For example, the use of multiphase nanoclusters and randomly aligned oxygen octahedral tiling has been shown to enhance the relaxor characteristics of ferroelectric ceramics, leading to high energy storage density and efficiency. Similarly, the manipulation of the atomic configurational entropy has been used to stabilize pyrochlore-based dielectric films and reduce polarization switching hysteresis, resulting in improved energy storage performance.[78]

The high-entropy strategy is a method used to control the size of ferroelectric domains in a material by incorporating multiple ions with different sizes and valences. This combination of ions leads to local structural disorder in the material, which in turn allows for dipoles in the ferroelectric domain to be rapidly reversed under smaller external electric fields. This disorder disrupts the long-range ordered ferroelectric state and facilitates the rapid reversal of dipoles under smaller external electric fields. By manipulating the composition and structure of the material, the high-entropy strategy enables the creation of a material that can be more easily polarized, leading to enhanced ferroelectric properties. This approach is a promising direction for the development of new materials with improved ferroelectric performance. High-entropy strategy can control preparation to reduce E_c and increase P_r, regulating the relaxor characteristics. Relaxor behavior refers to the presence of a broad and frequency-dependent dielectric response, which is desirable for energy storage applications. By inducing local structural disorder through the high-entropy strategy, the relaxor characteristics of the material can be enhanced, leading to improved dielectric energy storage capabilities.

High-entropy perovskite ceramics can achieve stable ferroelectric polarization through various factors such as A-site ion radius and valence difference, lattice distortion, and oxygen octahedron deflection. This stability results in disorder in the crystal structure, ultimately enhancing the material's polarization reaching 20 $\mu C/cm^2$ under the electric field of 50 kV/cm[79] as shown in Figure 9. Anisotropic high-entropy relaxor ferroelectric ceramics have multiple ions doped in the A-site, leading to an increased degree of disorder and heterogeneity of microregion composition.[24] The introduction of disorder and heterogeneity through the high-entropy strategy provides a means to tailor the polarization characteristics of perovskite ceramics. By precisely controlling the composition and structural variations, it becomes possible to achieve ceramics with unique and desired polar properties. As there can be various cations occupying the A-site in the material, their robust polar ferroelectric properties and regions displaying weaker polar relaxor-like characteristics might coexist. The variation in polarization can be attributed to fluctuations or variances in the composition of the material due to the high-entropy design strategy.[55]

The concept of a morphotropic phase boundary strategy was proposed as an approach to achieve ferroelectricity in high-entropy perovskite oxides.[76] This strategy takes an advantage of the unique properties and interactions among different lead-based components to create new design ideas exhibiting enhanced ferroelectric and piezoelectric properties.[80] The understanding and manipulation of high-entropy strategy in relaxor ferroelectrics create new compositional designs with improved performance. Researchers continue to explore novel ways to enhance the tunability and functionality of relaxor ferroelectrics through the control of configuration entropy.[56,78] The $(NaBiBa)_x(SrCa)_{(1-3x)/2}TiO_3$ ($x = 0.18$–0.22) high-entropy ceramics have been investigated for their ferroelectric properties, and the results have revealed significant achievements in terms of polarization. A high maximum polarization of $P_m \sim 16.59$ μC/cm^2 and the residual polarization of $P_r \sim 1.44$ μC/cm^2 was obtained under the external electric field of 100 kV/cm with $x = 0.21$. These values indicate the strong ferroelectric behavior and the ability of the material to retain a certain polarization even in the absence of an external field. Similarly, single-phase four-layered Aurivillius structure ferroelectric ceramic exhibits relaxor ferroelectric behavior due to the thermal stability of dipoles, cocktail effect, and local stress field. The presence of both long-range ordered domains and nanodomains introduces additional local disorder

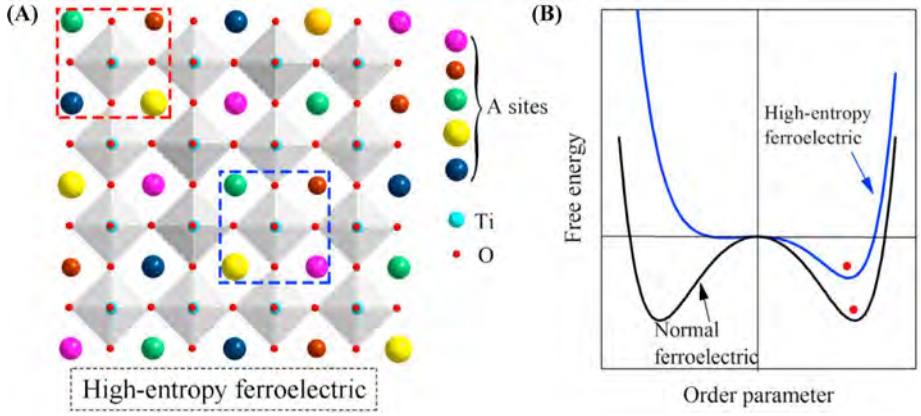

FIGURE 9 (A) Structural schematic and (B) Landau free energy for ferroelectrics. *Source*: Ref. [79]. Reproduced with permission of Elsevier.

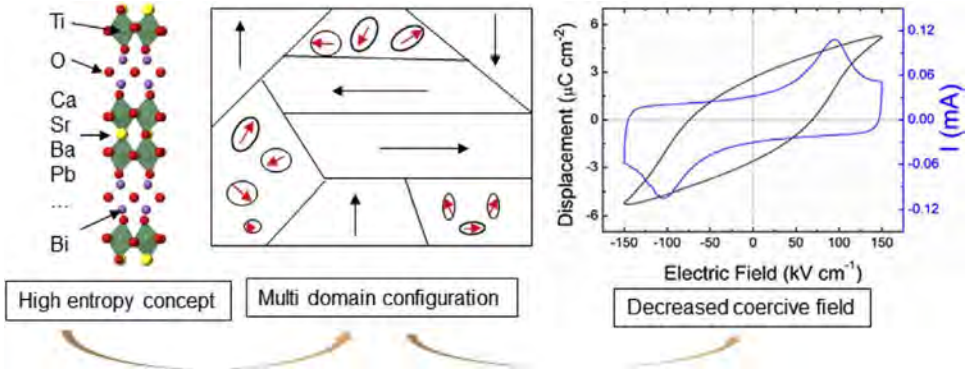

FIGURE 10 The high-entropy concept to regulate the coercive field. *Source*: Ref. [82]. Reproduced with permission of Elsevier.

and provides sites for enhanced polarization switching (as shown in Figure 10).[81,82]

This distinctive domain configuration plays a pivotal role in reducing the coercive fields of ceramics and an improved piezoelectric constant (d_{33}). The ferroelectric performance of these ceramics can also be extended to thin films by epitaxial film growth method.[83] The doping of B-site cations in $Ba(Ti_{0.2}Sn_{0.2}Zr_{0.2}Hf_{0.2}Nb_{0.2})O_3$ [Ba(5B)O] led to local disorder, which affected its phase transition, enhanced the local chemical disorder, and aggravated polarization disorder. They demonstrated that the maximum dielectric permittivity at the lowest frequency (100 Hz) appeared at the temperature T_m of 570 K. These materials, characterized by their diverse compositions and structural features, have shown promising performance in terms of their dielectric properties, phase transitions, and polarization behaviors make them highly attractive for applications requiring precise and efficient control of electrical polarization, such as in sensors, actuators, and transducers. It was found that the local configuration disorder provides a way for designing new relaxor ferroelectrics. The summary of high-entropy ferroelectric materials is shown in Table 2.

4 | MICROWAVE DIELECTRIC CERAMICS

In recent years, there have been significant research and development efforts focused on advancing novel microwave dielectric ceramics with improved dielectric properties. These properties include higher permittivity, improved quality factor, and a desired temperature coefficient of the resonant frequency. By combining multiple ions, the local structural disorder is induced, which leads to reduced oxygen octahedral tilt and improved temperature coefficient. High-entropy design strategy also leads to an ordered–disordered phase transition and improved quality factor. Recent studies have shown promising results in synthesizing high-entropy ceramics with enhanced microwave dielectric properties. These ceramics have the potential to outperform traditional

TABLE 2 The permittivity (ε_r), Curie temperature (T_c), maximum polarization (P_{max}), residual polarization (P_r), and coercive field (E_c) of high-entropy ferroelectric materials.

Composition	ε_r	T_c (°C)	P_{max} ($\mu C/cm^2$)	P_r ($\mu C/cm^2$)	E_c (kV/cm)	Ref.
$(PbBaSrCa)_{0.25}TiO_3$	15 000	65	15	5	~10	55
$0.675Pb(MgZnNbTaW)_{0.2}O_3$–$0.325PbTiO_3$	9029	~70	26	12	~8	80
$(BiNaBaSrPb)_{0.2}TiO_3$	3994	232	24.2	20.5	~30	79
$(NaBiBa)_{0.21}(SrCa)_{0.185}TiO_3$	2902	~35	16.59	1.44	~12	56
$(NaBiCaSrBa)_{0.2}TiO_3$	~2650	~40	23.59	~0	~10	24
$(Ca_{0.2}Sr_{0.2}Ba_{0.2}Pb_{0.2}Nd_{0.1}Na_{0.1})Bi_4Ti_4O_{15}$	~2000	560	~4.5	~2	~125	81
$(Ca_{0.2}Sr_{0.2}Ba_{0.2}Pb_{0.2}Nd_{0.1}Na_{0.1})Bi_2Nb_2O_9$	~350	405	~4.0	~1.7	~65	82

materials and meet the demanding requirements of microwave applications. Microwave dielectric ceramics find widespread use in various applications, including dielectric resonators, filters, substrates, and antennas. Dielectric permittivity (ε_r), quality factor ($Q \times f$), and temperature coefficient of resonant frequency (τ_f) are the critical parameters determining the specific applications of microwave dielectric ceramics. The calculation formulas of the three parameters are as follows[84]:

$$\varepsilon(\omega) = \varepsilon(\infty) + \frac{(ze)^2}{\beta V \varepsilon_0 - \frac{(ze)^2}{3}} \tag{7}$$

$$Q \times f = \frac{\beta - \frac{(ze)^2}{3V\varepsilon_0}}{2\pi m\gamma} \tag{8}$$

$$\tau_f = \frac{f(T_1) - f(T_0)}{f(T_0)(T_1 - T_0)} \tag{9}$$

where $\varepsilon(\infty)$ is the optical frequency dielectric permittivity, ε_0 is the low-frequency dielectric permittivity, e is the unit charge, V is the unit cell volume, β is the force constant between adjacent ions, m is the ion conversion mass, and $f(T_1)$ and $f(T_0)$ refer to resonant frequencies at test temperature range.

When it comes to microwave dielectric ceramics, achieving high dielectric permittivity (ε_r) and high quality factor ($Q \times f$) simultaneously poses a challenge. This is due to the inherent trade-off among the three critical parameters: ε_r, $Q \times f$), and the temperature coefficient of the resonant frequency (τ_f). Based on Equations (7) and (8), there exists an inverse relationship between ε_r and $Q \times f$. Because of the trade-off among the three parameters, therefore, it is difficult to synthesize and prepare materials with high ε_r, low dielectric loss, and low τ_f. In recent years, researchers have turned their attention to the high-entropy strategy as a potential solution to simultaneously regulate microwave dielectric properties.

The successful synthesis and optimization of $Li(Gd_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2}Lu_{0.2})GeO_4$ ceramics highlight the potential of high-entropy strategies in the development of microwave dielectric materials. The dielectric properties of $\varepsilon_r = 7.2$, $Q \times f = 29\,000$ GHz, $\tau_f = -2.9$–5.3 ppm/°C.[85] Comparing with A_2BO_4 (A = Ca, Mg; B = Si, Ge) and $LiREGeO_4$ olivines,[86–90] high-entropy $LiREGeO_4$ simultaneously exhibited improved $Q \times f$ and τ_f values, whereas the dielectric properties of $LiYGeO_4$ is $\varepsilon_r = 9.41$, $Q \times f = 18\,860$ GHz, $\tau_f = -27.7$ ppm/°C.[89] The improvement in τ_f was explained by the reduction of $[ReO_6]$ oxygen octahedral tilt in $Li(Gd_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2}Lu_{0.2})GeO_4$ via high-entropy strategy. Under the dominance of the high-entropy effect and lattice distortion effect, the oxygen polyhedron tilt or distortion is effectively regulated. This may be the most important reason for the improvement of microwave dielectric properties. The summary of high-entropy microwave dielectric ceramics along with their properties is shown in Table 3.

The utilization of high-entropy strategies in microwave dielectric ceramics holds promise for achieving materials with superior performance and expanded applications. The resulting ordered–disordered phase transition and

TABLE 3 Dielectric permittivity (ε_r), quality factor ($Q \times f$), and temperature coefficient of resonant frequency (τ_f) of high-entropy microwave dielectric ceramics.

System	ε_r	$Q \times f$ (GHz)	τ_f (ppm/°C)	Ref.
$Li(Gd_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2}Lu_{0.2})GeO_4$	7.2	29 000	−2.9–5.3	85
$(Mg_{0.2}Ni_{0.2}Zn_{0.2}Co_{0.2}Mn_{0.2})_2SiO_4$	8.02	28 400	−38.2	91
$(Hf_{0.25}Zr_{0.25}Sn_{0.25}Ti_{0.25})O_2$	25.6	74 600	−47	44
$Sr(La_{0.2}Nd_{0.2}Sm_{0.2}Eu_{0.2}Gd_{0.2})AlO_4$	18.6	14 509	−6	92
$(La_{0.2}Nd_{0.2}Sm_{0.2}Ho_{0.2}Y_{0.2})NbO_4$	19.48	47 770	−13.5	93

lattice distortion also provide a way to regulate the quality factor and the temperature coefficient of the resonant frequency.

5 | SUMMARY AND OUTLOOK

In this work, we reviewed the theory and development of this cutting-edge technology and summarize the latest research findings on its use in energy storage. High entropy holds enormous potential for improving the relaxor characteristics of ferroelectric materials. Particularly, it can help to create more PNRs in dielectric energy storage, leading to enhanced performance. Moreover, high-entropy microwave dielectric ceramics offer a wealth of structural possibilities, with numerous lattice distortions that can be adjusted to optimize quality factors and resonant frequency temperature coefficient. Although the potential benefits of high-entropy dielectrics are clear, there is still much work to be done in terms of broadening and deepening our understanding of this technology. In particular, the lack of reference samples can make it challenging to form a complete system and theory. Nevertheless, this also presents an exciting opportunity for future research to overcome these challenges and unlock the full potential of high-entropy dielectrics. Following are a few conclusions and potential future directions arising from the above review:

1. In-depth research of the relationship between the structure and properties of high-entropy dielectrics expands the number of research samples to eliminate the contingency in the case.
2. It is well known that compounds of the same structure are most likely to form solid solutions. However, it is more meaningful to dissolve a variety of different structural compounds to form a single phase. It is still unknown that the solid solution of various crystal structures brings changes in the final structure and properties.
3. High-entropy structures have more complex conditions than single structures, and small differences in the preparation environment can also lead to large differences in structures and even single-phase structures cannot be obtained. It is necessary to use advanced structure detection techniques to explore and analyze high-entropy structures, such as neutron diffraction, extended X-ray absorption fine structure, and atom probe tomography.
4. In the complex case of multi-ion solid solution, especially in the case of heterovalent ions, the synergy of multicomponent is difficult to be explained by density functional theory and kinetic model, and it is necessary to find high-throughput calculation methods to predict the synthesis of high-entropy compounds.
5. By controlling the configuration entropy, the ordered–disordered phase transition of ions in the structure can be precisely regulated, providing a broad space for the performance regulation of dielectrics due to the abundant electronic and phonon modes in ceramics.

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CONFLICT OF INTEREST STATEMENT

The authors declare that they have no known conflicts of interest.

ORCID

Laijun Liu https://orcid.org/0000-0002-6889-2506
Jibran Khaliq https://orcid.org/0000-0001-5835-9102
Chunchun Li https://orcid.org/0000-0002-3657-5856

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