Structural, Dielectric and Magnetic Properties of $\text{Ba}_3\text{SrLn}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$ (Ln=La, Nd, Sm) Filled Tungsten Bronze Ceramics

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Abstract

Structural, dielectric and magnetic properties have been investigated for $\text{Ba}_3\text{SrLn}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$ (Ln=La, Nd, Sm) ceramics. The single phase tetragonal filled tungsten bronze structure in space group $P4/mbm$ is obtained for $\text{Ba}_3\text{SrLa}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$, while such tungsten bronze major phase is determined together with minor amount of secondary phases in $\text{Ba}_3\text{SrNd}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$ and $\text{Ba}_3\text{SrSm}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$. The saturated magnetic hysteresis loops with enhanced M_r are obtained in the present ceramics at room temperature comparing to the $\text{Ba}_4\text{Ln}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$ (Ln=La, Nd, Sm) ceramics. Meanwhile, the typical relaxor behaviors are determined: a broad dielectric peak with strong frequency dispersion and the peak temperature following well with the Vogel-Fulcher relationship.

Keywords: A. ceramics, B. solid state reactions, C. dielectric response, magnetism, D. magnetic measurements, X-ray diffraction.

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1. Introduction

Ceramics with tungsten bronze structure compose one of the largest classes of dielectrics just next to the perovskites, and they consist of ten distorted BO_6 octahedra sharing corners. The octahedra are linked by their corners in such a way that three different types of tunnels run right through the structure parallel to the c-axis in a general formula $[(\text{A}_1)_2(\text{A}_2)_4\text{C}_4][(\text{B}_1)_2(\text{B}_2)_8]\text{O}_{30}$ [1]. As the smallest interstice C is generally empty, the filled tungsten bronze structure could have a general formula $\text{A}_6\text{B}_{10}\text{O}_{30}$. These features make them capable of performing a wide range of substitutions, either in the channels or within the octahedral network itself. Thus the properties of tungsten bronzes could be modified further to make it interesting and useful for devices because of the great flexibility of structure.

Recently, Zhu *et al.* published a comprehensive review on the ferroelectric transition and low-temperature dielectric relaxations in filled tungsten bronze ceramics together with the possible multiferroicity [2]. The ferroelectric nature of $\text{M}_4\text{R}_2\text{Ti}_4\text{Nb}_6\text{O}_{30}$ and $\text{M}_5\text{RTi}_3\text{Nb}_7\text{O}_{30}$ tungsten bronzes have been intensively investigated [3-10]. It is believed that the phase transition nature and transition temperature are primarily determined by structural modulation through the oxygen octahedral tilting [2]. A large radius difference between A1- and A2-site ions (Δr) will lead to the commensurate modulation in tetragonal tungsten bronze structure and the normal ferroelectric transition with a high T_C . The ferroelectric nature changes from normal ferroelectric toward diffuse and relaxor ferroelectric in the tetragonal tungsten bronze structures with incommensurate modulation as Δr decreases. The order of ferroelectric transition in filled tungsten bronzes will also be disturbed by the order/disorder in A1 and A2 sites, their random cross occupancy, and the order/disorder in B-sites.

The filled tungsten bronze ceramics have also been investigated as the possible multiferroic materials by introducing magnetic elements [11-17]. The electrical polarization and ferroelectricity are associated with the off center displacement of the B site cations and octahedra distortion. Meanwhile, the magnetism are associated with magnetic ions with partially filled d shells. Josse *et al.* reported the coexistence of

room-temperature ferromagnetism and ferroelectricity in $\text{Ba}_4\text{Ln}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$ (Ln=Nd, Sm, Eu, Gd, and Pr) [12]. In $\text{Ba}_4\text{Ln}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$ compounds, the ordered tungsten bronze structure is generally indicated, in which Ba and Ln ions properly occupy the A2 and A1 site, respectively. The problem is that the magnetism is weak for these materials in practical application. The $\text{Ba}_4\text{La}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$ compound is paramagnetic with M_{2T} of 0.09 emu/g at room temperature. In $\text{Ba}_4\text{Nd}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$ compound, $M_r=0.10$ emu/g and $H_C=1860$ Oe is obtained with M_{2T} of 0.34 emu/g. $M_r=0.15$ emu/g and $H_C=2150$ Oe is obtained in $\text{Ba}_4\text{Sm}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$ compound with M_{2T} of 0.42 emu/g [12]. Whether the disturbance of order/disorder in A and B sites have effects on the ferroelectric or magnetic nature are not confirmed in Fe-based filled tungsten bronze ceramics. There is an important issue to further understand how do the cations occupation affect these properties. A convenient way to determine this issue is to substitute one Ba^{2+} cation by smaller Sr^{2+} and then to investigate the variation of properties in $\text{Ba}_3\text{SrLn}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$ to form the filled tungsten bronze structures with disordered distribution in A site.

In the present work, dielectric and magnetic characteristics of $\text{Ba}_3\text{SrLn}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$ (Ln=La, Nd, Sm) ceramics are investigated together with the structure. The Vogel-Fulcher law and Curie-Weiss law fittings confirm the relaxor nature, and all compositions display the improved magnetic hysteresis loops at room temperature.

2. Material and methods

$\text{Ba}_3\text{SrLn}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$ (Ln=La, Nd, Sm) ceramics were prepared by a standard solid-state reaction process. High-purity BaCO_3 (99.93%), SrCO_3 (99.95%), La_2O_3 (99.99%), Nd_2O_3 (99.99%), Sm_2O_3 (99.99%), Fe_2O_3 (99.9%), and Nb_2O_5 (99.99%) powders were used as the raw materials. Before weighing, La_2O_3 , Nd_2O_3 and Sm_2O_3 raw powders were pretreated at 1273 K in air for 3 h to remove the hydroxides. These raw powders were mixed by ball milling in a polyethylene jar with ZrO_2 media in ethanol for 24 h. Then the mixtures were calcined in a high-purity alumina crucible at 1473-1623 K (depending on composition) in air for 3 h. The calcined powders were

ground again to reach the homogeneous granulometric distribution. Adding with 6 wt% polyvinyl alcohol (PVA) as the binder, the reground powders were pressed into disks with 12 mm in diameter and 2 mm in thickness under a pressure of about 98 MPa. These discs were sintered at 1523-1623 K in air for 3 h to yield the dense ceramics.

The crystal structure was identified by powder X-ray diffraction (XRD) analysis with Cu K α radiation (PANalytical, Almelo, the Netherlands) at room temperature. For structure refinement, the XRD data were collected over the range of $2\theta=10-130^\circ$ with a step width of 0.01° . The Rietveld structural refinement was performed using the FULLPROF program. The microstructures were evaluated by a scanning electron microscopy (S-4800, Hitachi, Tokyo, Japan).

Dielectric characteristics of $\text{Ba}_3\text{SrLn}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$ (Ln= Nd and Sm) ceramics were measured with a broadband dielectric spectrometer (Turnkey Concept 50, Novocontrol Technologies, Hundsangen, Germany) in a broad temperature (133-573 K) and frequency (1 Hz-5 MHz) range. The low-temperature dielectric characteristics of $\text{Ba}_3\text{SrLa}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$ ceramics were measured with an impedance analyzer (Agilent4294A, Agilent Technologies Inc., Santa Clara, CA) equipped with a cooling system (HC-4E 1, Janis, Woburn, Massachusetts) in a temperature (30-200 K) and frequency (100 kHz-1 MHz) range. The magnetization measurements were conducted with a magnetic property measurement system (MPMS-XL-5, Quantum Design, San Diego, CA) in a temperature (10-300 K) range.

3. Results and discussion

3.1 Phase and structure analysis

Fig.1 shows the XRD patterns of the polycrystalline $\text{Ba}_3\text{SrLn}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$ (Ln=La, Nd, Sm) ceramics. The tetragonal tungsten bronze single phase structure is determined for Ln=La, while a few additional peaks of LnNbO_4 are observed for the compositions of Ln=Nd and Sm beside the tungsten bronze major phase. The phase fraction of LnNbO_4 is approximately 2% for Ln=Nd and 7% for Ln=Sm according to a rough estimate. As the ionic radius of rare earth decreases from 0.136 nm (Ln=La) to 0.124 nm (Ln=Sm), the tolerance factors of filled tungsten bronze are diminished

from 0.967 to 0.954, while the tolerance factors for A1 site cation are diminished from 0.954 to 0.913, and this results in the reduced structure stability.

The Rietveld refinement of XRD data for $\text{Ba}_3\text{SrLa}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$ confirms the tetragonal tungsten bronze structure in space group $P4/mbm$ (see **Fig.2**). As shown in **Table 1**, the experimental parameters ($R_{wp}=3.71\%$, $R_p=2.85\%$, $R_{exp}=3.02\%$ and $\chi^2=1.51$) indicate the fine agreement between the calculated and the observed XRD patterns. The lattice parameters of $\text{Ba}_3\text{SrLa}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$ are calculated as $a=b=12.4918(2)$ Å, $c=3.9309(1)$ Å, $V=613.395(20)$ Å³. The selected bond lengths and angles are summarized in **Table 2**. Since three A2-sites are occupied by Ba ions, the rest of A-sites might be occupied disorderly by Sr and La ions considering the small difference of ion radius between them, which results in a disordered cation distribution. In tetragonal tungsten bronze ferroelectrics, the cation distribution has significant influence on the ferroelectric transition, and the relaxor behavior is closely associated with the disordering in cationic repartition [18]. **Fig.3** shows a view of the crystallographic structure along the c axis of $\text{Ba}_3\text{SrLa}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$. Fe^{3+} and Nb^{5+} statistically occupy the B1 and B2 sites in a ratio of 1:4. Ba^{2+} , Sr^{2+} and La^{3+} occupy the A2 sites. The residual Sr^{2+} and La^{3+} occupy the A1 sites. The cross occupancy occurs between Sr^{2+} and La^{3+} since the small radius difference. Microstructures of the thermally etched surfaces of $\text{Ba}_3\text{SrLn}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$ (Ln=La, Nd, Sm) ceramics are showed in **Fig.4**, where one can see that the dense ceramics have been obtained.

3.2 Dielectric characteristic analysis

The dielectric characteristics of $\text{Ba}_3\text{SrLn}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$ (Ln=La, Nd, Sm) ceramics as the functions of temperature at various frequencies are shown in **Fig.5**. Broad dielectric peaks with strong frequency dispersion are observed in the curves of dielectric constant vs temperature for all compositions. The dielectric peak temperature T_{max} shifts to a higher temperature with decreasing ionic radius of rare earth, ~104 K for La, ~175 K for Nd, and ~233 K for Sm at 100 kHz. The dielectric loss peak of $\text{Ba}_3\text{SrLn}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$ (Ln=Nd and Sm) ceramics, which appears at lower temperature than that of dielectric constant, also exhibits strong frequency dispersion.

These behaviors are typical characteristics for relaxor ferroelectrics with a diffuse phase transition. The diffuse degree of these transitions can be evaluated by the modified Curie-Weiss law [19].

$$1/\varepsilon - 1/\varepsilon_{max} = (T - T_{max})^\gamma / C_I \quad (1)$$

where ε is the dielectric constant, ε_{max} is the peak value of ε at T_{max} , C_I is the modified Curie-Weiss constant and γ is the diffuseness exponent. The variation of $\log(1/\varepsilon - 1/\varepsilon_{max})$ with $\log(T - T_{max})$ at 100 kHz for $\text{Ba}_3\text{SrLn}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$ is shown in **Fig.6**. The γ values obtained from the slope of the curve are 1.63, 1.51 and 1.71 for Ln=La, Nd and Sm, respectively. The γ value is between 1 (normal ferroelectric and obeying the Curie-Weiss law) and 2 (relaxor ferroelectric and completely disordered system), implying a diffuse ferroelectric nature of $\text{Ba}_3\text{SrLn}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$ (Ln=La, Nd, Sm) ceramics.

In order to further investigate the ferroelectric nature, the dielectric maximal temperature as a function of frequency is analyzed using the Vogel-Fulcher relationship [20-22].

$$f = f_0 e^{-E_a/k_B(T_{max} - T_f)} \quad (2)$$

where f_0 is the Debye frequency, E_a is the activation energy of the polar clusters, k_B is the Boltzmann constant, T_{max} is the permittivity maximum temperature, and T_f is the static freezing temperature. The relationship represents the polar glasslike behavior with thermally activated polarization fluctuations above a static freezing temperature existing in the relaxor ferroelectrics [23-25]. Because of the narrow measured frequency range (100 kHz-1 MHz), data of $\text{Ba}_3\text{SrLa}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$ ceramics is not fitted. As shown in **Fig.7**, the dielectric maximal temperature as a function of frequency well follows the Vogel-Fulcher relationship. The fitting parameters are $E_a=0.080$ eV, $T_f=106.73$ K, $f_0=6.39 \times 10^{10}$ Hz for Ln=Nd; and $E_a=0.182$ eV, $T_f=104.61$ K, $f_0=8.00 \times 10^{12}$ Hz for Ln=Sm. The good agreement of experimental data with Vogel-Fulcher relationship indicates that the interaction of dipoles is analogous to the spin-glass-like thermal activated process.

Extensive studies of the ferroelectricity of tungsten bronze structured compounds

have confirmed the dominating role of off-center displacement of B-site ions [26-28]. As shown in **Fig.3** and **Table 1**, both A and B sites show the random occupation of ions. The occupation of ions with different valence and ionic radius at the same site result in the local compositional fluctuation and unequally distributed local electric field and strain field. Therefore, the long-range ordering of $\text{Ba}_3\text{SrLn}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$ (Ln=La, Nd, Sm) at low temperature owing to off-center Fe/Nb dipoles is destroyed by the distributed fields. The large ferroelectric domains are broken into polar nano-domains which contribute to the dielectric relaxation [28].

3.3 Magnetic analysis

Fig.8 shows the magnetization-field (M - H) curves for $\text{Ba}_3\text{SrLn}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$ (Ln=La, Nd, Sm) ceramics at 300 K. The classical magnetic hysteresis loops are obtained in the present ceramics. The remanent magnetic moments (M_r) of Ln=La, Nd and Sm are 0.43 emu/g, 0.64 emu/g and 0.46 emu/g, respectively, which are much larger than those in $\text{Ba}_4\text{Ln}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$ compounds. At the same applied field of 20 kOe, the magnetization (M_{2T}) is 1.23 emu/g for Ln=La, 1.55 emu/g for Ln=Nd, and 1.10 emu/g for Ln=Sm. The coercivity (H_C) of Ln=La, Nd and Sm are 1340 Oe, 1847 Oe and 1911 Oe, respectively. In comparison to $\text{Ba}_4\text{Ln}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$ compounds, $\text{Ba}_3\text{SrLn}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$ exhibits an enhanced M_r and M_{2T} . It is supposed that A-site cations occupation have a significant influence on the magnetic properties, which needs further investigation. Now, we can discuss the effects of LnNbO_4 secondary phases on the magnetic properties. While the content of NdNbO_4 in $\text{Ba}_3\text{SrNd}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$ ceramics is less than that of SmNbO_4 in $\text{Ba}_3\text{SrSm}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$ ceramics, the magnetic saturation and remanent moments of $\text{Ba}_3\text{SrNd}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$ ceramics are larger than $\text{Ba}_3\text{SrSm}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$ ceramics. So the magnetism can be ascribed to the tungsten bronze structure but not the secondary phase. As the enhanced magnetism has been achieved in the present system, new multiferroic materials can be expected if the relaxor nature can be tuned towards normal ferroelectric.

4. Conclusions

The single phase tetragonal filled tungsten bronze structure in space group $P4/mbm$ is obtained for $\text{Ba}_3\text{SrLa}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$, while such tungsten bronze major phase is determined together with minor amount of secondary phases in $\text{Ba}_3\text{SrNd}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$ and $\text{Ba}_3\text{SrSm}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$. The obvious magnetic hysteresis loops with enhanced M_r are obtained in the present ceramics at room temperature comparing to the $\text{Ba}_4\text{Ln}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$ (Ln=La, Nd, Sm) ceramics and they can be ascribed to the tungsten bronze structure. Meanwhile, the typical relaxor behaviors are determined: a broad dielectric peak with strong frequency dispersion and the peak temperature following well with the Vogel-Fulcher relationship. If the relaxor nature can be tuned towards normal ferroelectric, new multiferroic materials can be expected in the present system.

Acknowledgements

This work is supported by National Science Foundation of China under Grant Nos. 51332006, 51102210, and 51202215.

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Table 1 Refined structural parameters for $\text{Ba}_3\text{SrLa}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$ from X-ray powder diffraction data

Atom	Wyckoff position	x/a	y/b	z/c	$B_{\text{iso}}(\text{\AA}^2)$	Occupies
La(1)	2a	0	0	0	0.270(63)	0.103(1)
Sr(1)	2a	0	0	0	0.270(63)	0.022(1)
Ba(1)	4c	0.1708(1)	0.6708(1)	0	2.501(69)	0.188
La(2)	4c	0.1708(1)	0.6708(1)	0	2.501(69)	0.022(1)
Sr(2)	4c	0.1708(1)	0.6708(1)	0	2.501(69)	0.041(1)
Fe(1)	2b	0	0.5	0.5	1.280(82)	0.025
Nb(1)	2b	0	0.5	0.5	1.280(82)	0.1
Fe(2)	8d	0.0753(1)	0.2151(1)	0.5	0.342(44)	0.1
Nb(2)	8d	0.0753(1)	0.2151(1)	0.5	0.342(44)	0.4
O(1)	2b	0	0.5	0	0	0.125
O(2)	4c	0.2821(6)	0.7821(6)	0.5	0	0.25
O(3)	8d	0.0579(7)	0.2117(7)	0	0	0.5
O(4)	8d	0.3427(8)	0.0070(6)	0.5	0	0.5
O(5)	8d	0.1361(6)	0.0608(8)	0.5	0	0.5

Lattice parameters: $a=b=12.4918(2)$ $\text{\AA}$, $c=3.9309(1)$ $\text{\AA}$, $V=613.395(20)$ $\text{\AA}^3$, space group $P4/mbm$.

Agreement indices: $R_{\text{wp}}(\%)=3.71$, $R_p(\%)=2.85$, $R_{\text{exp}}(\%)=3.02$, $\chi^2=1.51$.

Table 2 Selected bond distances (Å) and angles (°) of Ba₃SrLa₂Fe₂Nb₈O₃₀ ceramics

Bond distances (Å)		Bond angles (°)	
La(1)/Sr(1)-O(3)	2.742(9)×4	O(1)-Fe(1)/Nb(1)-O(1)	180.0(0)
La(1)/Sr(1)-O(5)	2.707(5)×4	O(1)-Fe(1)/Nb(1)-O(4)	90.0(3)×4
	2.707(7)×4		90.0(4)×4
Ba(1)/ La(2)/Sr(2)-O(2)	2.781(5)×2	O(4)-Fe(1)/Nb(1)-O(4)	180.0(7)
Ba(1)/ La(2)/Sr(2)-O(4)	2.842(7)×2		180.0(9)
	2.842(6)×2		95.1(6)×2
Fe(1)/Nb(1)-O(1)	1.965(0)×2		84.9(6)×2
Fe(1)/Nb(1)-O(4)	1.967(8)×2	O(2)-Fe(2)/Nb(2)-O(3)	96.2(4)×2
	1.967(10)×2	O(2)-Fe(2)/Nb(2)-O(4)	97.7(6)
Fe(2)/Nb(2)-O(2)	1.968(8)	O(2)-Fe(2)/Nb(2)-O(5)	93.6(6)
Fe(2)/Nb(2)-O(3)	1.978(1)×2		175.0(7)
Fe(2)/Nb(2)-O(4)	1.896(8)	O(3)-Fe(2)/Nb(2)-O(3)	167.2(0)
Fe(2)/Nb(2)-O(5)	2.072(10)	O(3)-Fe(2)/Nb(2)-O(4)	87.6(4)×2
	1.966(8)	O(3)-Fe(2)/Nb(2)-O(5)	91.2(5)×2
			83.9(4)×2
		O(4)-Fe(2)/Nb(2)-O(5)	168.7(8)
			87.3(6)
		O(5)-Fe(2)/Nb(2)-O(5)	81.4(6)

List of Figure Captions

Fig.1. XRD patterns of crashed powders of $\text{Ba}_3\text{SrLn}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$ ceramics: a) $\text{Ln}=\text{La}$, b) $\text{Ln}=\text{Nd}$, and c) $\text{Ln}=\text{Sm}$.

Fig.2. Observed (open circles), calculated (solid lines), and difference (bottom line) of X-ray powder diffraction patterns for $\text{Ba}_3\text{SrLa}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$ ceramics. The tick marks present the position of all possible Bragg reflections of $\text{Ba}_3\text{SrLa}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$.

Fig.3. Polyhedral representation of tungsten bronze structure of $\text{Ba}_3\text{SrLa}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$ along c-axis.

Fig.4. SEM micrographs of (a) $\text{Ba}_3\text{SrLa}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$, (b) $\text{Ba}_3\text{SrNd}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$, and (c) $\text{Ba}_3\text{SrSm}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$.

Fig.5. Dielectric constant and dielectric loss as a function of temperature for (a) $\text{Ba}_3\text{SrLa}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$, (b) $\text{Ba}_3\text{SrNd}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$, and (c) $\text{Ba}_3\text{SrSm}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$.

Fig.6. $\text{Log}(1/\varepsilon - 1/\varepsilon_{\max})$ as a function of $\text{log}(T - T_{\max})$ at 100 kHz for (a) $\text{Ba}_3\text{SrLa}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$, (b) $\text{Ba}_3\text{SrNd}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$, and (c) $\text{Ba}_3\text{SrSm}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$.

Fig.7. Log of relaxation time as a function of dielectric constant peak temperature, where the solid dots are experimental data and the curve is the Vogel-Fulcher fitting for (a) $\text{Ba}_3\text{SrNd}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$ and (b) $\text{Ba}_3\text{SrSm}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$.

Fig.8. Magnetic hysteresis loops of (a) $\text{Ba}_3\text{SrLa}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$, (b) $\text{Ba}_3\text{SrNd}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$, and (c) $\text{Ba}_3\text{SrSm}_2\text{Fe}_2\text{Nb}_8\text{O}_{30}$ ceramics at 300 K.

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