Multi-susceptible Single-Phased Ceramics with Both Considerable Magnetic and Dielectric Properties by Selectively Doping

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Materials with extraordinary susceptibilities coexisting are vitally important for the multi-functionality and integration of electronic devices. However, multiferroic composites, as the most potential candidates, will introduce inevitable interface deficiencies and thus dielectric loss from dissimilar phases. In this study, single-phased ferrite ceramics with considerable magnetic and dielectric performances appearing simultaneously were fabricated by doping target ions in higher valence than that of Fe\(^{3+}\), such as Ti\(^{4+}\), Nb\(^{5+}\) and Zr\(^{4+}\), into BaFe\(_{12}\)O\(_{19}\). In terms of charge balance, Fe\(^{3+}/Fe^{2+}\) pair dipoles are produced through the substitution of Fe\(^{3+}\) by high-valenced ions. The electron hopping between Fe\(^{3+}\) and Fe\(^{2+}\) ions results in colossal permittivity. Whilst the single-phased ceramics doped by target ions exhibit low dielectric loss naturally due to the diminishment of interfacial polarization and still maintain typical magnetic properties. This study provides a convenient method to attain practicable materials with both outstanding magnetic and dielectric properties, which may be of interest to integration and multi-functionality of electronic devices.

Results and Discussion

Formation of Fe\(^{2+}\) ions and Fe\(^{3+}/Fe^{2+}\) pairs in the ferrites with doping Ti\(^{4+}\). Fig. 1 illustrates the XRD patterns of the barium ferrite ceramics. As can be seen, only M-type barium ferrite phase is detected in all the samples. The
structural parameters ‘a’ and ‘c’ as well as cell volume of barium ferrite ceramics are listed in Table 1. It is found that lattice constants and cell volume both decrease initially from x = 0 to x = 0.4 and then increase slowly with Ti4+ ions increasing. The smallest “a” and “c” are obtained when x = 0.4.

Fig. 2 shows SEM photographs of the barium ferrite ceramics. It is seen that the ceramics with or without Ti4+ doping all form typical hexagonal plate-like particles. The grain size seems to become larger when Ti4+ content increases from x = 0 to x = 0.8 as shown from Fig. 2(a) to Fig. 2(d), which indicates that the formation and grain growth of barium ferrite ceramics are promoted apparently with increasing Ti4+ content.

Table 1 | Structural parameters and volume of BaFe12-xTixO19 ceramics (x = 0, 0.4, 0.6 and 0.8) sintered at 1200°C for 3 h

| x     | a[Å]    | c[Å]    | V[Å³]   |
|-------|---------|---------|---------|
| 0     | 5.904   | 23.251  | 701.889 |
| 0.4   | 5.858   | 23.121  | 687.125 |
| 0.6   | 5.859   | 23.126  | 687.509 |
| 0.8   | 5.864   | 23.182  | 690.385 |

Figure 2 | SEM photographs of barium ferrite ceramics of (a) BaFe12O19, (b) BaFe11.6Tix0.4O19, (c) BaFe11.4Tix0.6O19 and (d) BaFe11.2Tix0.8O19 sintered at 1200°C for 3 h.

The lattice constants of the ferrites decrease initially and then increase slowly with addition of Ti4+. As is known, Ti4+ ions added into the ceramic matrix probably substitute for constituent Fe3+ ions in barium ferrite due to the close ionic radius of Ti4+ (0.605 Å) and Fe3+ (0.645 Å)12, which has been evidenced by Mössbauer spectroscopy elsewhere13. Meanwhile, defect reaction as following will be triggered.

\[
Ti + Fe_{Fe^*} \xrightarrow{BaFe_{12}O_{19}} Ti^{4+} + Fe_{Fe^*} \quad (1)
\]

It is seen that some Fe3+ ions will be transferred to be Fe2+ ions in terms of charge balance. In fact, there are many deficiencies such as oxygen vacancies with positive charge exist in barium ferrite ceramics originally14. When Ti4+ ions are initially doped into barium ferrites, for electronic balance, the oxygen vacancies may be restrained to form dominantly rather than the transformation of Fe3+ into Fe2+. Then, as oxygen vacancies reach smallest, Fe3+ ions will start to be doped into the ferrites initially due to the smaller size of Ti4+ compared with Fe3+. Then, a large number of Fe2+ ions are generated in BaFe12-xTixO19 with x ≥ 0.6.

Moreover, Fig. 3 demonstrates the XPS spectra of Fe 2p for BaFe12.4Tix0.6O19 (Fig. 3a) and BaFe11.2Tix0.8O19 (Fig. 3b) after subtracting baseline, in which C1s peak at 285 eV was used for charge correction and the peaks at ~709.3 eV and ~722.8 eV are belong to Fe2+ and Fe3+, respectively.
Fe\textsuperscript{2+} 2p\textsubscript{3/2} and Fe\textsuperscript{2+} 2p\textsubscript{1/2}, respectively\textsuperscript{15}. It is apparent that Fe\textsuperscript{2+} ions are formed in the ferrites with Ti doping of x \(\geq 0.6\) which is in fact related with the transformation between Fe\textsuperscript{3+} and Fe\textsuperscript{2+} ions based on Eq. (1). For clearly understanding, it can be confirmed as shown in Fig. 4, in which there are two resonance peaks with two Landé factors (g) of 2.0 and 2.3 appearing in imaginary part (\(\mu''\), the magnetic loss) of the relative complex permeability of BaFe\textsubscript{12-x}Ti\textsubscript{x}O\textsubscript{19} (x = 0.5, 0.6, 0.7 and 0.8) over 26.5–40 GHz\textsuperscript{10}. As a matter of fact, the g of around 2.3 is from the exchange coupling between Fe\textsuperscript{3+} and Fe\textsuperscript{2+} ions (Fe\textsuperscript{3+}/Fe\textsuperscript{2+} pairs) which is different from 2.0 of Fe\textsuperscript{3+} in the ferrites\textsuperscript{10}. The intensity of the peak about Fe\textsuperscript{3+}/Fe\textsuperscript{2+} pairs increases apparently with increasing Ti\textsuperscript{4+} ions. So in this work, Fe\textsuperscript{3+}/Fe\textsuperscript{2+} pairs with Fe\textsuperscript{2+} ions generated by doping Ti\textsuperscript{4+} are clearly formed in BaFe\textsubscript{12-x}Ti\textsubscript{x}O\textsubscript{19} especially with x \(\geq 0.6\).

Magnetic properties kept high effectively in the doping ferrite. Fig. 5 displays hysteresis loops of the BaFe\textsubscript{12-x}Ti\textsubscript{x}O\textsubscript{19} ferrites (x = 0, 0.4, 0.6 and 0.8). The data of coercive force (\(H_c\)), anisotropic field (\(H_a\)), saturation magnetization (\(M_s\)) and residual magnetization (\(M_r\)) deduced from Fig. 5 are summarized in Table 2. It is seen that the maximum \(H_c\), \(H_a\), \(M_s\) and \(M_r\) of all the ferrits are obtained through the law of approach to saturation (LAS), which can be expressed as Eq. (2)\textsuperscript{16}:

\[
M = M_s \left(1 - \frac{A}{H} - \frac{B}{H^2}\right) + x_p H
\]

where A is the inhomogeneity parameter, B is the anisotropy parameter and \(x_p\) is the high-field differential susceptibility. B of hexagonal symmetry can be expressed as Eq. (3).

\[
B = \frac{H_a^2}{15}
\]

It is shown in Table 2 that \(H_a\) of BaFe\textsubscript{12-x}Ti\textsubscript{x}O\textsubscript{19} decreases dramatically from 15.43 kOe to 10.43 kOe as x varies from 0 to 0.8. In fact, grain size increases and thus the amount of grain boundaries will decrease with increasing Ti\textsuperscript{4+} ions in the ferrites as shown in Fig. 2. Fe\textsuperscript{3+} ions which in general contributes \(H_a\) in barium ferrite is substituted by non-magnetic Ti\textsuperscript{4+} ions in this case and hence \(H_a\) will decrease. The more the Fe\textsuperscript{3+} ions are replaced, the weaker the \(H_a\) is Ref. 16. The \(H_a\) of BaFe\textsubscript{12-x}Ti\textsubscript{x}O\textsubscript{19} decreases therefore with increasing Ti\textsuperscript{4+} content to x = 0.8 although the reduction in grain boundaries will increase \(H_a\). In addition, coercivity which represents the ability to retain magnetization.
to resist magnetization reversal process under a reverse magnetic field is mainly controlled by the hindrance for nucleation of reverse domain, domain wall motion and spin rotation in magnetic materials. As \( H_c \) which impedes spin rotation decreases with increasing Ti\(^{4+} \) ions in the ferrites, the coercivity decreases hence with increasing Ti\(^{4+} \) ions. Moreover, nuclei of reverse domains are easily formed around the deficiencies such as grain boundaries and dislocations, while the deficiencies can also act as pinning centers to hinder domain wall motion. In fact, grain boundaries have a marked influence on pinning domain wall instead of inducing nucleation of reverse domains in barium ferrite. The coercivity of barium ferrite will decrease significantly with decreasing grain boundaries (increasing grain size). Due to the decrease in both \( H_c \) and grain boundaries, the coercivity decreases eventually from 3.04 kOe to 1.06 kOe with increasing Ti\(^{4+} \) content from \( x = 0 \) to \( x = 0.8 \), which is about 65% lower than that of undoped one. It implies that the Ti\(^{4+} \) doped barium ferrite ceramics will be a good candidate with low energy consumption applied in devices.

Meanwhile, \( M_s \) and \( M_r \) still maintain high values in BaFe\(_{11.2}\)Ti\(_{0.8}\)O\(_{19}\). In fact, Fe\(^{3+} \) ions of BaFe\(_{12}\)O\(_{19}\) in 12k, 2a, and 2b sites are up-spin and 4f\(_1\) and 4f\(_2\) are down-spin. In this case, the net magnetization of the ferrites is contributed by excess of up-spin magnetic moments. That is to say, if Fe\(^{3+} \) ions are substituted by non-magnetic Ti\(^{4+} \) ions in 12k, 2a, and 2b sites, magnetization will decrease. Conversely, magnetization will be improved if Fe\(^{3+} \) ions which are in 4f\(_1\) and 4f\(_2\) sites are substituted. As is reported, Ti\(^{4+} \) substitutes preferably for Fe\(^{3+} \) in 12k, 2b, and 4f\(_1\) sites with both states of up spin and down spin. It leads to only a gentle decline of both saturation and residual magnetization in the ferrites with increasing Ti\(^{4+} \). Apparently, \( M_s \) and \( M_r \) of BaFe\(_{11.2}\)Ti\(_{0.8}\)O\(_{19}\) of about 60 emu/g and 29 emu/g, which are only 20 ~ 30% lower than that of undoped one, are still high enough in practical use in devices keeping typical magnetic properties.

Fig. 6(a) and 6(b) shows the permeability and magnetic loss tangent of the BaFe\(_{12-x}\)Ti\(_{x}\)O\(_{19}\) (\( x = 0, 0.4, 0.6 \) and 0.8) ceramics as a function of frequency respectively. It is seen that permeability of all the samples is almost independent of frequency, except for a little bit decrease in BaFe\(_{11.2}\)Ti\(_{0.8}\)O\(_{19}\) above 70 MHz. Meanwhile, the frequency independent permeability increases rapidly from about 1.5 to 5.1 with \( x \) varying from 0 to 0.8. The magnetic loss tangent of the ferrites depends on frequency and Ti\(^{4+} \) content. It decreases from \(~0.35 \) to \(~0.07\) at low frequency of 1 MHz and increases from \(~0.1\) to \(~0.4\) around 100 MHz respectively with increasing content of Ti\(^{4+} \) ions from \( x = 0 \) to \( x = 0.8 \). While it is as low as \(~0.1\) at moderate frequency.

Obviously, the permeability improves with increasing Ti\(^{4+} \) ions in barium ferrites. As is known, domain wall motion and domain rotation are two dominant magnetization processes for polycrystalline ferrites. For the BaFe\(_{12-x}\)Ti\(_{x}\)O\(_{19}\) polycrystalline ceramics, grain boundaries abating due to larger grain size in high Ti\(^{4+} \) doped samples promotes domain wall motion. Furthermore, anisotropic field and demagnetizing field impeding domain rotation in the ferrites decreases with increasing Ti\(^{4+} \) content. Consequently, controlled by enhancing both domain wall motion and spin rotation, the permeability of BaFe\(_{11.2}\)Ti\(_{0.8}\)O\(_{19}\) ceramic is improved to 5.1, which is 3 ~ 4 times of BaFe\(_{12}\)O\(_{19}\) ceramic.

Except for the permeability, magnetic loss is also related naturally to the doping content in the ferrites. Considering the high electrical resistivity of barium ferrite, eddy loss can be neglected. Magnetic loss of BaFe\(_{12-x}\)Ti\(_{x}\)O\(_{19}\) over the frequency range between 1 MHz and 100 MHz is contributed dominantly by hysteresis loss and residual loss. Actually, hysteresis loss is predominant over lower frequency range, while residual loss takes charge in higher frequency range. As can be seen in Fig. 5, area of hysteresis loops reduces with increasing Ti\(^{4+} \) ions content. The magnetic loss decreasing with Ti\(^{4+} \) content at low frequency is thus controlled by hysteresis loss, which is \(~0.35\) of BaFe\(_{12}\)O\(_{19}\) to \(~0.07\) of BaFe\(_{11.2}\)Ti\(_{0.8}\)O\(_{19}\) at ~1 MHz. However, the residual loss is contributed in general by domain wall resonance which occurs at frequency above 100 MHz in BaFe\(_{12}\)O\(_{19}\). As shown in Fig. 6(b), it seems that the resonance peak moves toward lower frequency range with doping Ti\(^{4+} \) in the ferrites. It implies that the magnetic loss of the BaFe\(_{12-x}\)Ti\(_{x}\)O\(_{19}\) ceramics is importantly controlled by the residual loss, which increases a little from \(~0.1\) to \(~0.4\) with doping Ti ions from \( x = 0 \) to 0.8 at ~100 MHz. At moderate frequency, magnetic loss is as low as \(~0.1\), which is a little bit decrease with doping Ti in the ferrites due to a little reduction of hysteresis loss. Obviously, the magnetic loss of BaFe\(_{12-x}\)Ti\(_{x}\)O\(_{19}\) ceramics is diminished to be lower than \(~0.1\) with increasing Ti\(^{4+} \) content to \( x = 0.8\), especially at frequencies below 70 MHz.

| $| x \text{ (Ti^{4+} content)} | H_c (KOe) | H_r (KOe) | M_s (emu/g) | M_r (emu/g) |
|---|---|---|---|---|---|
| 0  | 15.43 | 3.04 | 72.38 | 37.43 |
| 0.4 | 12.72 | 3.03 | 58.36 | 28.41 |
| 0.6 | 11.38 | 1.93 | 64.09 | 31.42 |
| 0.8 | 10.34 | 1.06 | 59.82 | 28.60 |

Colossal permittivity and low dielectric loss of the doped single-phased ferrites. Fig. 7(a) and 7(b) shows the permittivity and dielectric loss of the BaFe\(_{12-x}\)Ti\(_{x}\)O\(_{19}\) (\( x = 0, 0.4, 0.6 \) and 0.8) ceramics as a function of frequency respectively. The permittivity of the ceramics increases significantly at low frequency range with doping Ti\(^{4+} \). Unlike the permittivity of barium ferrite without doping which is almost independent of frequency, the permittivity of the Ti\(^{4+} \) ions doped barium ferrites decreases rapidly with increasing frequency and the decreasing speed becomes slow as the content of Ti\(^{4+} \) is high from \( x = 0.4 \) to 0.6 to \( x = 0.8 \). A steplike shoulder appears typically in permittivity at moderate frequency with Ti content of \( x \geq 0.6\). Colossal
permittivity which is about 100 k below 100 KHz and 20 k above 1 MHz appears in BaFe$_{11.2}$Ti$_{0.8}$O$_{19}$ ceramic. The dielectric loss tangent of BaFe$_{12-x}$Ti$_{x}$O$_{19}$ ceramics decreases dramatically initially and then keeps stable with increasing frequency and it reduces accordingly with rising Ti$^{4+}$ ions in the ferrites.

It is known that space charge polarization contributes most probably high permittivity especially at low applied frequencies.$^{28}$ Free electric charges may easily increase with doping and move freely in ceramics without localization. It will contribute the permittivity to the ceramics due to the charge response and the permittivity thus decreases with frequency. Such as in the ceramics with Ti$^{4+}$ content of x = 0.4 ~ 0.6, the permittivity is apparently higher than that of the undoping one and decreases rapidly with increasing frequency at low frequency range, because the un-localized charges form in the barium ferrits with initially doping Ti$^{4+}$ ions. Higher permittivity and more rapid decline at low frequency are exhibited with Ti$^{4+}$ content increasing from x = 0.4 to x = 0.6. However, Fe$^{3+}$ will most probably transform into Fe$^{2+}$ to keep charge balance in the ferrits with high doping of Ti$^{4+}$ ions. It implies that the electric charges generated will be localized between the two ions to form Fe$^{3+}$ and Fe$^{2+}$ pairs. Thus, as revealed in Fig. 6(a), the decline of permittivity at low frequency range becomes smoother with increasing Ti$^{4+}$ content from x = 0.4 ~ 0.6 to x = 0.8.

In fact, the step-like shoulder at middle frequency range in BaFe$_{12-x}$Ti$_{x}$O$_{19}$ with x $\geq$ 0.6 is based on Fe$^{3+}$/Fe$^{2+}$ pair dipoles. As analyzed above, Fe$^{3+}$ ions are supposed to be abundantly produced in the ferrits since x reaches 0.6, Fe$^{3+}$ and Fe$^{2+}$ pairs make most probably electron pair dipoles in the ferrits. The higher the Ti$^{4+}$ content is in the ceramics, the more the amount of pair dipoles is. So the step-like shoulder which is generated generally by relaxation dipoles such as Fe$^{3+}$ and Fe$^{2+}$ pairs appears initially in BaFe$_{12-x}$Ti$_{x}$O$_{19}$ with x $\geq$ 0.6 and becomes more apparent in BaFe$_{11.2}$Ti$_{0.8}$O$_{19}$ ceramic reasonably. Hence, the high permittivity is probably dominantly contributed by the Fe$^{3+}$/Fe$^{2+}$ pair dipoles in BaFe$_{12-x}$Ti$_{x}$O$_{19}$ with x $\geq$ 0.6 instead of by charge response in the ferrits with x < 0.6. Moreover, considering polycrystalline ceramics in this case, conductivity inhomogeneity will appear in the ferrits due to the different electron hopping styles or hopping species in grains compared with those in grain boundaries. According to the Koop’s opinions, the conductivity inhomogeneity contributes importantly the high permittivity.$^{11}$ The colossal permittivity which is about 100 k below 100 KHz and 20 k above 1 MHz appears hence in high Ti$^{4+}$ doped ferrite ceramics of BaFe$_{11.2}$Ti$_{0.8}$O$_{19}$ based on these two important contributions.

Furthermore, besides of high permittivity, the dielectric loss tangent of the ferrits decreases attractively with doping Ti$^{4+}$ ions. The smallest dielectric loss of 0.2 is obtained in BaFe$_{11.2}$Ti$_{0.8}$O$_{19}$ at ~10 kHz. It is much lower than that of percolative ferroelectric/ferromagnetic composite ceramics with both extraordinary dielectric and magnetic properties. In fact, in percolative ferroelectric/ferromagnetic composite ceramics, a great deal of space charges and other deficiencies will be produced in the interfaces between ferrite phases and ferroelectric phases due to the two different lattice structures. However, these deficiencies can be effectively eliminated in the single-phased ceramics as the grain boundaries are relatively perfectly matched among the identical lattice structures. Thus, the part of dielectric loss contributed by interfacial polarization is significantly decreased and low dielectric loss of only 0.2 appears in the single-phased barium ferrite ceramics. Apparently, single-phased ferrite ceramics doped with Ti$^{4+}$ ions are potential multifunctional ceramics with both impressive magnetic and dielectric properties.

**Dual properties appearing universally in the single-phased ferrits with Fe$^{3+}$/Fe$^{2+}$ pairs.** As a matter of fact, colossal permittivity as well as excellent magnetic properties appearing simultaneously in the single-phased ferrite ceramics is not due to Ti element itself but due to its higher valence state than that of Fe$^{3+}$ in the ferrites. As is shown in Fig. 8, plots of the permittivity and dielectric loss tangent of Nb$^{5+}$ and Zr$^{4+}$ doped barium ferrite BaFe$_{11.7}$Nb$_{0.3}$O$_{19}$
A series of BaFe_{12-x}TixO_{19} ceramics with x varies from 0 to 0.8 were synthesized by a known sol-gel process. The Fe^{3+} and Fe^{2+} pair dipoles are produced by the substitution of high valence ions, such as Ti^{4+}, Nb^{5+} and Zr^{4+}, for Fe^{3+} based on charge balance in the ferrites. As Ti^{4+} substitutes preferably for Fe^{3+} in the sites with two compensated spin directions in the barium ferrite, the saturation magnetization and residual magnetization of the Ti^{4+} doped ferrites still keep high values to be practically used. Controlled by hysteresis loss, the magnetic loss of the Ti^{4+} doped ferrite ceramics diminishes effectively. Following the electron hopping between Fe^{3+} and Fe^{2+} ions and conductivity inhomogeneity between grains and grain boundaries in the ferrites, giant permittivity appears. Eliminating completely the interfaces between dissimilar phase structures, the dielectric loss tangent of the single-phased ferrites reduces significantly compared with that of the extensively interested percolative ferroelectric/ferromagnetic composite ceramics. Obviously, the single-phased ferrite ceramics doped by target ions in higher valence than that of Fe^{3+} reveal both extraordinary magnetic and dielectric properties simultaneously, which are even more competitive compared with the known systems such as multiferroic composites because of lower dielectric loss and thus become the most promising multifunctional materials in application of electronic devices for integration and multi-functionality.

Methods

A series of BaFe_{12-x}Ti_xO_{19} ceramics with x varies from 0 to 0.8 were synthesized by a sol-gel process. Firstly, barium nitrate (Ba(NO_3)_2), ferric nitrate (Fe(NO_3)_3·9H_2O), citric acid (C_6H_5O_7·H_2O) were weighed, and dissolved in deionized water to obtain solutions A. Solutions B containing Ti^{4+}, Nb^{5+}, and Zr^{4+} were prepared by dissolving Ti(OC_2H_5)_4, C_6H_5O_7·H_2O into anhydrous ethanol. According to stoichiometric proportion, the solutions A and B are mixed to get solutions C, ammonia was used to adjust the PH value to about 7. The solutions C were dried at 120°C to form fluffy dry gels, the gels were then further calcined at 800°C for 3 h and red-brown powders were achieved. Finally, the powders mixed with appropriate amount of 5% PVA were molded into a ring shape under a pressure of 10 Mpa and then sintered at 1200°C to form BaFe_{12-x}Ti_xO_{19} ceramics.

Figure 8 | plots of the dielectric constant and dielectric loss of (a) BaFe_{11.7}Nb_{0.3}O_{19} and (b) BaFe_{11.5}Zr_{0.5}O_{19} as a function of frequency.

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Author contributions
P.Y. and N. lead the project and designed work progress. P.Y. and C.Y. analysed the results and conceived the central idea. J.G. and C.Y. performed the most experiments. Y.J. and S.Q. partially supported the experiments. C.Y., Y.J. and P.Y. wrote the manuscript and all authors discussed the results.

Additional information
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