

# Electromagnetic properties of NiZn ferrite/SrTiO<sub>3</sub>/HDPE composites for high-frequency applications

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Low loss dielectric and magnetic composite with SrTiO<sub>3</sub> (STO) ultrafine particles and NiZn ferrite (NZO) ultrafine particles embedded in a high density polyethylene (HDPE) matrix was fabricated using the extrusion technology. The dielectric and magnetic properties of the as-prepared composites with different volume fraction of ceramic fillers were investigated. The results indicate that as the volume of the ceramic filler was fixed, with the increasing of the ratio of STO to NZO, the permittivity of the composites increases while the dielectric loss, permeability and magnetic loss decrease. The cut-off frequencies of the composites are all above 1 GHz.

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## 2. Introduction

In recent years, with the rapid developments of electronics industry, electronic components have been required with smaller size and, higher performance. It is desired that the materials for these electronic components exhibit multifunctional properties, such as magnetic-electric [1, 2], magnetic-optical [3], flexibility [4] and other properties. Among them, dielectric-magnetic composites have recently received much attention. Much work has been carried out to synthesize dielectric-magnetic composite materials to meet the requirement for multifunctional components [5-9]. These composite materials can show both high inductive and capacitive properties, which has been identified to be an enabling solution to fabricate miniature filter and antenna, electro-magnetic interference (EMI) devices and so on. For the ceramic-based composites, however, the magnetic phase and the dielectric phase need to be cofired at high temperatures; moreover, the shrinkage mismatch between the two ceramic phases could seriously influence the performance of the final products. Alternatively, polymer-based composites with high permittivity [10-15] or high permeability [16-19] have been proposed due to their light weight, chemical stability, compatibility with printed wiring board (PWB), and ability to be easily fabricated into various shapes.

According to effective medium theory, the high permittivity of the polymer-based composites with high permittivity ceramic or metal particles can be easily obtained, whereas high initial permeability can be obtained by dispersing ferrite particles into the polymer matrices. So far some studies [20-22] have been carried out about this kind of composites. However, the composites obtained have the deficiency of possessing very high loss and low cut-off frequency below 1GHz, which limits the practical high-frequency application.

In this paper, a novel high-frequency electromagnetic composites were achieved by introducing the SrTiO<sub>3</sub> (STO) and NiZn ferrite (NZO) fillers into a high density polyethylene (HDPE) matrix. The electromagnetic properties and the

mechanical properties of the composites were investigated in detail. And such magnetic-dielectric three-phase composites, possessing very low loss, could be used in high-frequency communications for the capacitor-inductor integrating devices such as electromagnetic interference filters and antennas.

## 2. Experimental

The HDPE (density: 0.95 g/cm<sup>3</sup>, M<sub>w</sub>:100000). The ceramic filler was fabricated by conventional oxide mixing method and ground into powder with grain size of about 500 nm. The permittivity of STO and permeability of NZO are 250 and 35, respectively. To make the powders possess an active surface, they was fully mixed with 2 % oleic acid solution. The surface modified ceramic filler and the HDPE were mixed for 12 min in a Rheomix600p system (Rheomix600p, HAAKE Co., Germany) operated at 60rpm and 180 °C. Then, the mixture was put into a disk mold and hot-pressed under stress of 10 MPa at a temperature of 180 °C for 5 min. The dielectric and magnetic measurement in the high frequency range of 100 MHz ~ 1 GHz were carried out by HP4291B impedance analyzer with HP16453A dielectric material test fixture and HP16454L magnetic material test fixture, respectively.

Fig.1 shows the magnetic hysteresis loops of the composites with the same matrix volume but with different volume ratio of NZO to STO. It can be seen that the saturation magnetization (M<sub>s</sub>) and the remnant magnetization (M<sub>r</sub>) decrease with increasing STO content, as expected, since these parameter depends on the total mass of the magnetic material. The reduction of the values may be caused by nonlinearity of the magnetic moments at the surface of the NZO particles, resulting in a decrease of the saturation magnetization for a higher STO content [23]. The coercivity (H<sub>c</sub>) keeps constant as the variation of NZO content, which shows all the samples have very similar microstructure.

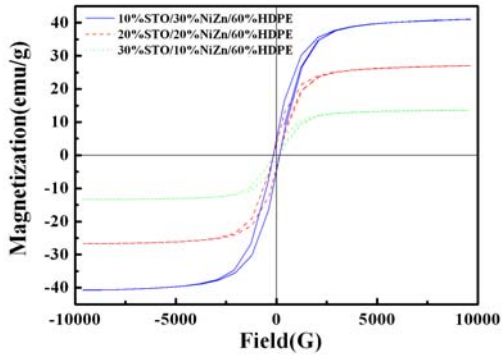


Fig. 1. Magnetic hysteresis loops of the composites with different volume ratio of NZO to STO.

The frequency dependence of the magnetic properties of the composite with different volume ratio of NZO to STO is shown in Fig. 2. It can be seen that with the increasing the volume of NZO the initial permeability and magnetic loss increase. According to Rikukawa [24], for either domain wall movement or spin rotation, the initial permeability is proportional to  $M_s^2$ . As has been explain above, with increasing NZO added into the composites,  $M_s$  increases. With the increasing of frequency the permeability of all the composites nearly keeps constant and the magnetic loss presents a dispersion feature and increases slightly only in high frequency range. It also can be found that the cut-off frequencies (i.e., the frequency where the  $\mu''$  value is maximal) of all the composites are above 1 GHz. And we can also assume that with the decreasing of the volume of NZO, the cut-off frequency increases. According to Snoek's law [25], the product of the initial susceptibility and resonance frequency is a constant for a ferromagnetic material, i.e.,  $(\mu_0 - 1)f_r = \gamma/2\pi M_s$ . The decreasing of the volume of NZO may cause the decrease of  $M_s$ . Accordingly, the increase of cut-off frequency can also be expected. Additionally, the magnetic loss of all the composites are all very low, which is due to the insulating matrix surrounding the NZO particles may drastically increase the resistivity of materials and reduce the eddy-current loss [26].

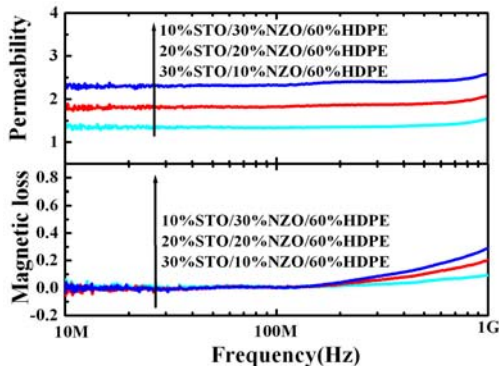


Fig. 2. Frequency dependence of the magnetic properties of the composite with different volume ratio of NZO to STO.

Fig. 3 shows the frequency dependence of the magnetic properties for the bulk NZO ceramic and the 0.1STO/0.3NZO/0.6HDPE composite. Comparing with the bulk NZO ceramics, it shows a resonance at about 100 MHz, the resonance frequency for the three-phase composite shifts to a much higher frequency beyond our measurement range. The three-phase composites possess an advantage of much wider working frequency range.

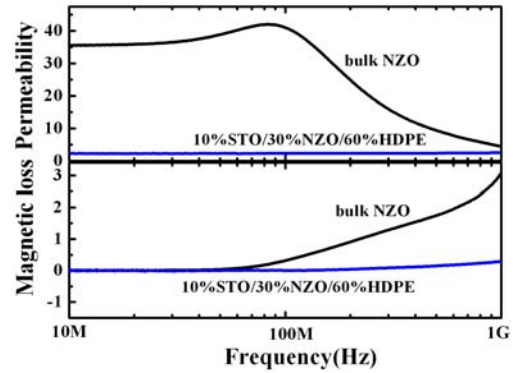


Fig. 3. Frequency dependence of the magnetic properties of the 0.1STO/0.3NZO/0.6HDPE composite and the bulk NZO.

Fig. 4 presents the frequency dependence of the magnetic properties for the NiZn/HDPE composites with different volume ratio of NZO. It can be easily found that with the increasing of NZO and the increasing of frequency they have the same tendency of the STO/NiZn/HDPE composites with the same volume of NZO. The magnetic loss of the two kinds of composite are very similar, which implied that both STO and polymer matrix have very high resistivities. The difference is the initial permeability of the STO/NiZn/HDPE composites is a little higher than those of the NiZn/HDPE composites possessing the same volume of NZO. For illustration and comparison, the calculations using well-known Maxwell-Garnett (MG) formula are also shown. For the two-phase composite of spherical ferrites and nonmagnetic host matrix, the MG formula for the effective permeability of the composite is

$$K = K_1 \left( 1 + \frac{3f_{NZO}\beta}{1 - f_{NZO}\beta} \right) \quad (1)$$

where  $\beta = (K_2 - K_1)/(K_2 + 2K_1)$ ;  $K$  is the effective permeability of the composite;  $K_1 = 1$  and  $K_2 = 35$  are the initial permeability of the polymer matrix and NZO, respectively;  $f_{NZO}$  is the volume fraction of the NZO particles. As shown in Fig. 5, for the NiZn/HDPE composites, when  $f_{NZO} < 0.40$ , the experimental permeability are in good agreement with the calculations by using the MG formula. However, for the STO/NiZn/HDPE composites, the experimental permeability are a bit larger than the corresponding the calculations by using the MG formula, which may due to the STO/NiZn/HDPE composites have more

ceramic fillers than those of the NiZn/HDPE composites and ceramic filler clusters easily occurred.

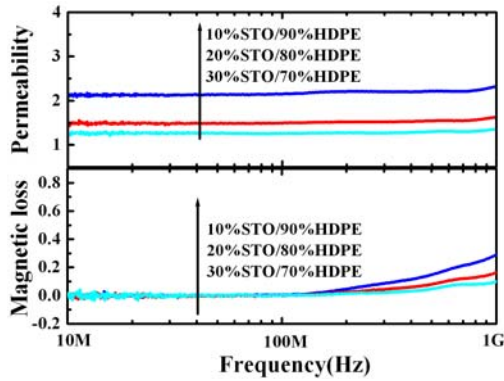


Fig. 4. Frequency dependence of the magnetic properties for the NiZn/HDPE composites with the different volume ratio of NZO.

Fig. 6 shows the dielectric properties of the composites with the same matrix volume but with the different volume ratio of NZO to STO. As expected, the permittivity increases while the dielectric loss decreases with the increasing of STO volume fraction. Notably, the dielectric loss of all the composites is very low in high frequency range. The permittivity presents good stability within a wide range of frequency. It also can be found that in low frequency range the dielectric loss of the composites are relative high, with the increasing of frequency, the dielectric loss decreases first and then

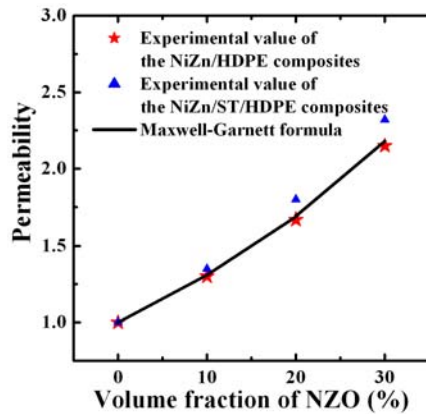


Fig. 5. Variation of the experiment and calculated values of the composites with the volume fraction of NZO.

increases. This is attributed to the low resistance of NZO and can be explained by Debye formula [27]. When an alternating electric field is applied on it, not only polarization loss but also leakage loss generated. Dielectric loss could be divided into two fractions.

$$D = D_p + D_G = \frac{(\epsilon_s - \epsilon_\infty)\omega\tau}{\epsilon_s + \epsilon_\infty\omega^2\tau^2} + \frac{\gamma}{\omega\epsilon_0} \left( \frac{1}{\epsilon_\infty + \frac{\epsilon_s - \epsilon_\infty}{1 + \omega^2\tau^2}} \right) \quad (2)$$

where  $D$  denotes dielectric loss,  $D_p$  denotes  $\tan\delta$  of polarization loss,  $D_G$  denotes  $\tan\delta$  of leakage loss. It can be observed that at a certain temperature as frequency ( $\omega$ ) go to 0, i.e., state electric field,  $D_p$  go to 0. That is to say, dielectric loss is almost attributed to leakage loss. In this case, the frequency is very low,  $\omega\tau \ll 1$ , so dielectric loss could be described approximately as below.

$$D \cong \frac{\gamma}{\omega\epsilon_0\epsilon_s} \quad (3)$$

Dielectric loss is inverse proportional to frequency. As frequency increasing, polarization loss gradually increases and become predominant. It can be assumed that a resonant peak would occur with the further increasing of frequency due to the LC resonance in the measurement circuit.

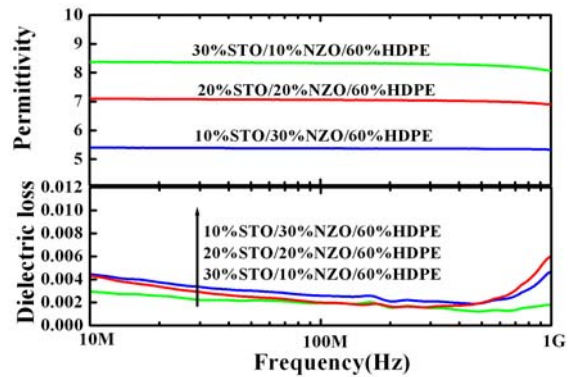


Fig. 6. Frequency dependence of the dielectric properties of the composite with different volume ratio of NZO to STO.

#### 4. Conclusions

STO/NZO/HDPE composites with various volume fractions of ceramic filler were prepared by using the extrusion technology. As the volume of the ceramic filler was fixed, with the increasing of the ratio of STO to NZO, the permittivity of the composites increase while the dielectric loss, permeability and magnetic loss decrease. The cut-off frequencies of the composites are all above 1 GHz. The permittivity and permeability of all the composites have shown good stability and low dielectric and magnetic losses has been obtained within the measurement range from 10 MHz to 1 GHz. For the 0.1STO/0.3NZO/0.6HDPE composite, the permittivity, dielectric loss, permeability and magnetic loss were 5.6, 0.0016, 2.5 and 0.003 at 100 MHz, respectively. Such magnetic-dielectric three-phase composites could be used in high-frequency communications for the capacitor-inductor integrating devices such as electromagnetic interference filters.

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### References

- [1] C. W. Nan, *Phys. Rev. B*, **50**, 6082 (1994).
- [2] M. Fiebig, *J. Phys. D*, **38**, R123 (2005).
- [3] T. Kanai, S. I. Ohkoshi, A. Nakajima, T. Watanabe, K. Hashimoto, *Adv. Mater (Weinheim, Ger.)* **13**, 487 (2001).
- [4] K. Y. Kim, H. S. Tae, J. H. Lee, *Microw. Opti. Technol. Lett.* **35**, 102 (2002).
- [5] J. V. Mantese, A. L. Micheli, D. F. Dungan, *J. Appl. Phys.* **79**, 1655 (1996).
- [6] T. Yamamoto, M. Chino, R. Tanaka, *Ferroelectrics* **95**, 175 (1989).
- [7] K. K. Patankar, V. L. Mathe, R. P. Mahajan, *Mater. Chem. Phys.* **72**, 23 (2003).
- [8] X. W. Qi, J. Zhou, Z. X. Yue, Z. L. Gui, L.T. Li, S. Buddhudu, *Adv. Funct. Mater.* **14**, 920 (2004).
- [9] B. W. Li, Y. Shen, Z. X. Yue, C. W. Nan, *J. Appl. Phys.* **99**, 123909 (2006).
- [10] Y. Bai, Z. Y. Cheng, V. Bharti, H. S. Xu, Q. M. Zhang, *Appl. Phys. Lett.* **76**, 3804 (2000).
- [11] C. Huang, Q. M. Zhang, *Adv. Funct. Mater.* **14**, 5001 (2004).
- [12] J. Y. Li, C. Huang, Q. M. Zhang, *Appl. Phys. Lett.* **84**, 3124 (2004).
- [13] Z. M. Dang, Y. Shen, C. W. Nan, *Appl. Phys. Lett.* **81**, 4814 (2002).
- [14] Z. M. Dang, Y. H. Lin, C. W. Nan, *Adv. Mater.* **15**, 1625 (2003).
- [15] Y. Shen, C. W. Nan, M. Li, *Chem. Phys. Lett.* **396**, 420 (2004).
- [16] J. Slama, R. Dosoudil, R. Vicern, A. Giruskova, V. Olah, I. Hudec, E. Usak, *J. Magn. Magn. Mater.* **254–255**, 195 (2003).
- [17] R. Lebourgeois, S. Berenguerb, C. Ramiarinjaonab, T. Waeckerle, *J. Magn. Magn. Mater.* **254–255**, 191 (2003).
- [18] J. H. Paterson, R. Devine, A. D. R. Phelps, *J. Magn. Magn. Mater.* **196–197**, 394 (1999).
- [19] T. Tsutaoka, *J. Appl. Phys.* **93**, 2789 (2003).
- [20] J. Q. Huang, P. Y. Du, L. X. Hong, Y. Dong, M. C. Hong *Adv. Mater.* **19**, 437 (2007).
- [21] H. W. Zhang, H. Zhang, B.Y. Liu, Y. L. Jing, Y. Y. Liu, *IEEE T. Magn.* **41**, 3454 (2005).
- [22] Y. Shen, Z.X. Yue, M. Li. *Adv. Funct. Mater.* **15**, 1100 (2005).
- [23] K. H. Wu, Y. C. Chang, T. C. Chang, Y. S. Chiu, T. R. Wu, *J. Magn. Magn. Mater.* **283**, 380 (2004).
- [24] H. Rikukawa, *IEEE T. Magn.* **18**, 1535 (1982)
- [25] J. L. Snoek, *Physica (Amsterdam)* **14**, 207 (1948).
- [26] X. G. Lu, G. Y. Liang, Y. M. Zhang, W. Zhang, *Nanotechnology* **18**, 015701 (2007).
- [24] J. X. Fang , Z. W. Yin, *Dielectric Physics (Chinese)*, Science Press, Beijing (1989).

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