

Study on Properties of 0-3 Cement Based Piezoelectric Composites

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Abstract: The sulphoaluminate cement and a piezoelectric ceramic, $0.08\text{Pb}(\text{Li}_{1/4}\text{Nb}_{3/4})\text{O}_3 \cdot 0.47\text{PbTiO}_3 \cdot 0.45\text{PbZrO}_3[\text{P}(\text{LN})\text{ZT}]$, were used to fabricate 0-3 cement based piezoelectric composites. The piezoelectric and dielectric properties of the composites were mainly investigated. The results indicate that the piezoelectric strain factor d_{33} increases as the P(LN)ZT volume fraction increases, which well follows the cube model. The dielectric constant ϵ_r and dielectric loss $\text{tg}\delta$ show similar trends with the d_{33} . In the frequency range of 40~100 kHz, the dielectric constants of the composites decrease sharply, which mainly is attributed to interfacial polarization in the composite. Above 200kHz, the cement-based piezoelectric composites exhibit good dielectric-frequency stability.

Key words: 0-3 cement based piezoelectric composite; piezoelectric property; dielectric property; interfacial polarization;

1. Introduction

For some important modern structures, such as high rise buildings, large span bridges, dams and so on, severe vibration and significant internal damage may be caused by dynamic loading from different sources, such as strong wind and earthquake. This will pose a greatly threat to the safety of the structures. Therefore, in recent years, structural healthy monitoring has great attracted attention in the civil engineering [1-2]. Real-time structural health monitoring can provide instantaneous information on condition of a specified structure, which will result in a significant

increase of safety margin and reductions in maintenance cost. The essential components for such health monitoring are sensors and actuators which are made of the so-called smart materials.

However, the traditional smart materials, such as piezoelectric ceramics, shape memory alloys and optical fibers, may not be applicable in civil engineering due to the distinct differences in the properties between the smart materials and the concrete [3-5]. For example the single phase piezoelectric ceramic exhibit high acoustic impedance (30MRayl) compared to that of concrete (9MRayl); During the hydration of cement, some inner or outer factors, such as chemical reactions and

variations in relative humidity or temperature, will cause shrinkage or expansion of the concrete, while the traditional smart materials do not change synchronistically with the concrete. Such mismatching would not only degrade the energy transfer between the smart materials and the host concrete, but also cause considerable loss of the signal transmission at the boundaries. So a new kind of smart material should be developed to meet the requirement of the civil engineering applications.

To meet the requirements of civil engineering structures, 0-3 and 2-2 cement based piezoelectric composites have been developed [5-10]. The experimental results showed that the developed cement based piezoelectric composites have favorable piezoelectric properties as well as good compatibility with the concrete structures. The 2-2 cement based piezoelectric composite exhibit an obviously linear relationship between the amplitude of piezoelectric voltage and the frequency of the applied cyclic load in the range from 0.1 to 50 Hz, which covers the majority of fundamental frequencies for civil engineering structures. At the same time, it still exhibits a desirable converse piezoelectric effect. These results indicate that it is enough for the cement based piezoelectric composite to be utilized to used as sensing and actuating element in the sensor and actuator for applications in the civil engineering structures.

Although the study of the cement-based piezoelectric composites has made great progress, a lot of problems remain to be solved. In order to further explore the cement based piezoelectric composite with superior piezoelectric properties, in this study, 0-3 piezoelectric ceramic/sulphoaluminate cement composites with 50% to 75% P(LN)ZT volume fractions have been fabricated. The piezoelectric properties, dielectric properties and ferroelectric properties of the composites are studied especially.

2. Experimental Procedures

The 0-3 cement-based piezoelectric composites were fabricated by compressing technique. Initially, the P(LN)ZT and sulphoaluminate cement were ball-milled for 30min with ethyl alcohol in a resin mill. After drying, the mixed materials were pressed into disks of 15mm diameter and 2mm thickness under 80MPa. The specimens were put in a curing room with a temperature of 20 °C and relative humidity of 100% for 3d before measurements. After curing, the surfaces of the disks were polished and coated with a low temperature silver paint then the specimens were poled by applying an electric field of 4kV/mm for 30 min at 100 °C in a stirred silicone oil bath.

After poling, the composites were aged for 24h prior to the measurements. The piezoelectric strain factor d_{33} was directly measured

using a Model ZJ-3A d_{33} piezometer. The capacitances of the composites were measured as a function of frequency using an Agilent 4294A Precision Impedance Analyzer, from which the dielectric constant can be calculated. The impedance /phase spectra and dielectric loss were measured using the same apparatus. The planar electromechanical coupling coefficients were determined by measuring the resonant frequency and anti-resonant frequency of the planar mode resonant in the samples. The distribution and bond status of the P(LN)ZT particles in the composites were observed on a Hitach S-2500 Scanning Electro Microscope (SEM).

3. Results and Discussion

3.1 Piezoelectric properties

The properties of the piezoelectric composites depend, to a large extent, on the amount of piezoelectric ceramics they contain, which is an design parameter to be optimized for end application. Therefore, the effect of the P(LN)ZT volume fraction on the piezoelectric properties of the composites was studied in detail in this paper.

Fig.1 shows variation of the piezoelectric strain factor d_{33} with the P(LN)ZT volume fraction. Accordingly, the theoretical curves of different models are also provided in the figure. It is seen that the d_{33} value increases with the increase of P(LN)ZT volume fraction. The experimental results confirm that the d_{33} values of the 0-3 cement based piezoelectric composites are close to the theoretical values of the cubes model. The theoretical equations of the Furukawa model and the cubes model for the d_{33} value in two phase systems are as follow:

The theoretical equation given by Furukawa et al [11]:

$$d_{33} = \frac{15d'_{33}ne'_r}{(1+n)(3+n)e_r}$$

The theoretical equation of the cubes model[12]:

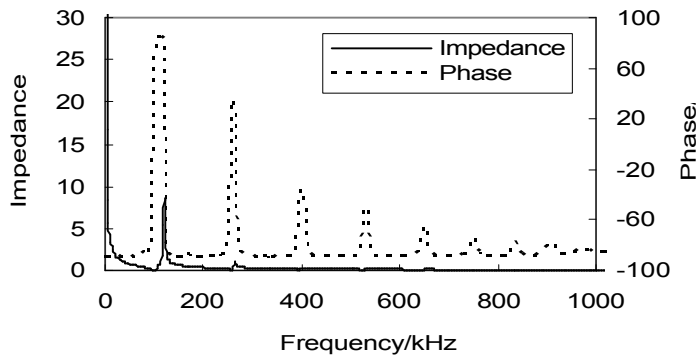
$$d_{33} = \frac{d'_{33}n}{[n^{\frac{1}{3}} + \frac{e_r}{e'_r}(1-n)^{\frac{1}{3}}]n(1+n)}$$

Where d_{33} and d'_{33} are the piezoelectric strain factors of the composite and the P(LN)ZT, n is the volume fraction of the P(LN)ZT, e_r and e'_r the relative dielectric constant of the P(LN)ZT and the matrix, respectively.

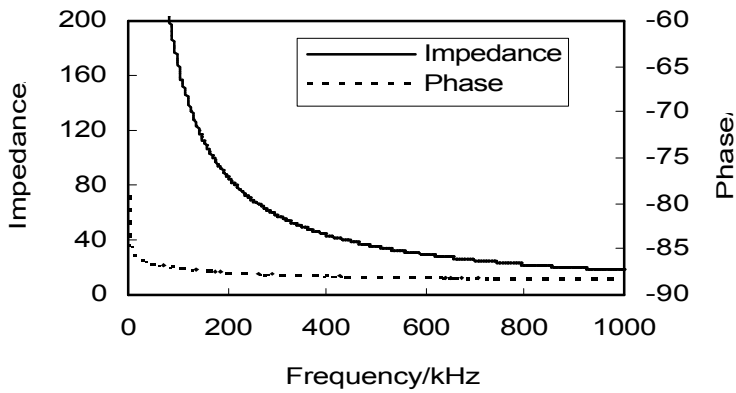
Fig.1 variation of the piezoelectric strain factor d_{33} with the P(LN)ZT volume fraction

Electromechanical properties of the composites were determined using an Agilent 4294A Precision Impedance Analyzer. Fig.2 shows the impedance and phase spectra of the P(LN)ZT, the sulphoaluminate cement and the composites. It can be seen that impedances of them reduce as frequency increases. The impedance of the sulphoaluminate cement is much higher than that of the P(LN)ZT and the composites. It can also be observed that the composites have planar resonance and other resonances are very weak, which is mainly attributed to the P(LN)ZT in composites. Whereas, for the sulphoaluminate cement, none planar resonance appears.

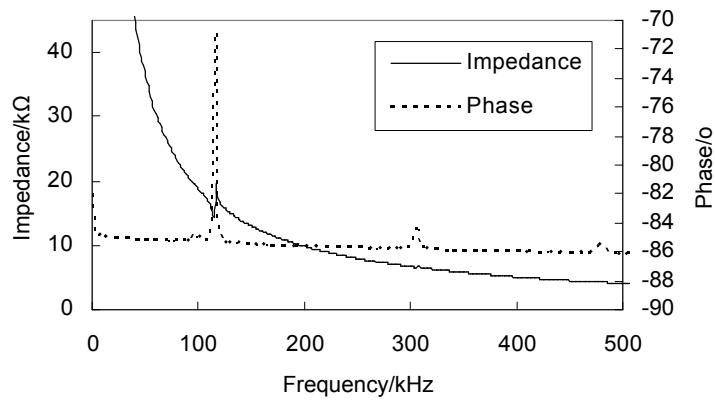
Fig.3 shows the impedance spectra of the composites with different the P(LN)ZT volume fractions. It is seen that the impedance of the composites decrease as the P(LN)ZT volume fractions increase. This is because the P(LN)ZT has lower impedance than that of the cement. Moreover, the P(LN)ZT volume fraction has also influenced on the resonance and anti-resonance frequency, and thus affects the planar electromechanical coupling coefficient K_p . From the resonance and anti-resonance frequency, the K_p was calculated. When the P(LN)ZT volume fraction varies from 50% to 75%, the K_p increases from 15.76% to 16.51%.



(a) P(LN)ZT



b Sulphoaluminate cement



(c) The composite with 65% P(LN)ZT volume fraction

Fig.2 Impedance and the phase spectra of the P(LN)ZT, Sulphoaluminate cement and composites

Fig3. Impedance spectra of the composite with different P(LN)ZT volume fraction

3.2 Dielectric Properties

The variations of dielectric constant ϵ_r of the composites and the cement with frequency are shown in Fig.4. and the variation of dielectric constant of the P(LN)ZT with frequency is shown in Fig.5 for comparison. It can be seen from Fig.4 that the dielectric constant of the composites increases with increasing the volume fraction of the P(LN)ZT. By comparing Fig.4 and Fig.5, it can be clearly seen that in the range of about 40~100kHz, the dielectric constants of the composites decrease sharply with increasing frequency, and the dielectric constant of the cement has little changes, while the dielectric constant of the P(LN)ZT is nearly independent of frequency. This means that the change of the dielectric constants of the composites is mainly caused by interface polarization of the composites and various polarization of the cement matrix.

It is well known that the cement is a porous inorganic material with a complicated microstructure. In brief, it is composed of an amorphous phase, crystallites in the micrometer range and bound water [13-14]. The various types of polarization may be excited in the presence of an applied electric field on the cement. When external electrical field acts on the cement matrix, many weak conducting ions (such as Ca^{2+} , OH^- , SO_4^{2-} and Al^{3+} and so on) [15] in the sulphoaluminate cement matrix begin to migrate besides polarization of electron, ion and dipole arising from hydrated products of cements, water and unreacted cement

particles etc. Commonly, after the cement mixes with the P(LN)ZT particle, many interface be formed, and at the same time, there is also many defects in cement matrix, such as impurities, lattice distortion and phase boundary etc .When the weak conducting ions reach these interface and defects areas , the ions will move slowly, leading to the accumulation of the ions near the interface and defect areas. Such accumulation effect will cause the polarization of space charge, that is, interfacial polarization. All the polarizations can follow the change of the electrical field at low frequency. Hence the dielectric constant of the composites are higher at low frequency. With increasing frequency, some polarizations, especially interfacial polarizations can not follow the change of electric field because of the long time for the construction of space charge polarization. Therefore, the dielectric constant of the composites is lower at high frequency.

It can also be seen that the higher the P(LN)ZT content is, the steeper the rate of variation of the dielectric constant with increasing frequency in the range of 40~100kHz. This is due to fact that with increasing the P(LN)ZT volume fraction , there are more interfacial polarizations in the composite, which are apt to be influenced by frequency. While the dielectric constants of the composites are almost not affected by frequency at high frequency. This indicates that at low frequency, the interfacial polarization is dominant in the composites.

About 120 kHz, some dielectric peaks appear in the dielectric constant-frequency curve of the composites. These peaks are mainly contributed by the dipole orientation polarization in the P(LN)ZT, which can be confirmed from Fig.5. It can also be observed that the higher the P(LN)ZT volume fraction is , the more the contribution of the dipole orientation polarization on the composites is, thus the dielectric peak of the composites will be also stronger.

Fig.4 Variation of dielectric constants of the composites and the cement with frequency

Fig.5 Variation of dielectric constant of the P(LN)ZT with frequency

The dependences of dielectric loss $\text{tg}\delta$ on the P(LN)ZT content are shown in Fig.6. It can be seen that with increasing the P(LN)ZT volume fraction, the dielectrics loss increases nonlinearly. When the P(LN)ZT volume fraction is less than 55%, the dielectric loss changes very small. But when the P(LN)ZT volume fraction exceeds 65%, the dielectrics loss increases remarkably. This is mainly because that the higher the P(LN)ZT volume fraction is, the worse the bond of the P(LN)ZT particle and the cement matrix, resulting in increase of porosity. This can be further confirmed by the results of SEM photographs for the composites. The SEM photographs of the composites with various P(LN)ZT volume fraction are showed in Fig.7, in which the white phases are P(LN)ZT particles and the black phases are the sulphoaluminate cement matrix. It can be seen that the P(LN)ZT particles disperse homogeneously throughout the cement matrix. When the P(LN)ZT volume fraction is low, The P(LN)ZT particles are closely packed by the cement matrix. There are not almost pores in the composites. But with increasing the P(LN)ZT volume fraction, the structure of the composites tends to be much less compact, leading to increase of dielectric loss.

Fig.6 Dependence of the dielectric loss on the P(LN)ZT volume fraction

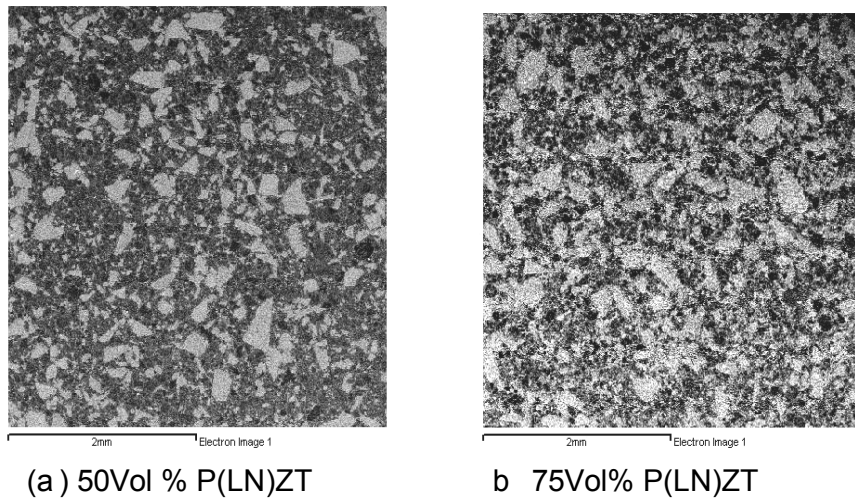


Fig.7 SEM photographs of the composites

4. Conclusions

The results obtained from the experiments are summarized as follows:

(1) The piezoelectric strain factor d_{33} increases as the P(LN)ZT volume fraction increases, which well follows the cube model. The dielectric constant ϵ_r and dielectric loss $\text{tg}\delta$ show similar trends with the d_{33} .

(2) There are obvious planar resonances in the impedance spectra of the composites. The impedance of the composites decreases as the P(LN)ZT volume fraction increases. When the P(LN)ZT volume fraction varies from 50% to 75%, the K_p increases from 15.76% to 16.51%.

(3) In the frequency range of 40~100 kHz, the dielectric constants of

the composites decrease sharply, which is attributed to interfacial polarization in the composite. Above 200kHz, the cement-based piezoelectric composites exhibit good dielectric-frequency stability.

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