Distribution of Ceramic Powder Dispersed in 0-3 Piezoelectric Ceramic–Polymer Composites and Their Properties

Wim Nhuapeng¹, Jerapong Tontrakoon¹ and Tawee Tunkasiri^{*1}

¹Department of Physics, Faculty of Science, Chiang Mai University, Chiang Mai 50200, Thailand *Corresponding author. E-mail: <u>tawee@chiangmai.ac.th</u>

ABSTRACT

The goal of this work was to study the relationship of physical properties such as acoustic impedance and density with distribution and volumetric fraction of ceramic powder in the 0-3 composites. The composites were fabricated using two methods, namely, calendering and centrifuge, with polyethylene and polyester resin as the matrix respectively and Lead Zirconate Titanate (PZT) as the filler. The morphology and distribution of the PZT particles were studied with scanning electron microscopy. The composite fabricated in a centrifuge was found to distribute more homogeneous and yielded higher physical values. The impedance and density...etc., were measured and found to increase with increasing of the volumetric fraction of the filler.

Key words: Piezoceramic-polymer 0-3 composite, distribution, physical properties.

INTRODUCTION

An important consideration for transducer performance is the physical properties, such as acoustic impedance, Bulk modulus, density...etc., between the transducer and its environment (Han et al., 1991). For example, if a ceramic transducer is employed in medical use, the coupling of the energy at the transducer-load interface is very poor (Gururaja et al., 1985) because of the mismatch of their physical properties. Therefore, attempt to reduce the such properties of the ceramic transducer is one of the goals in many works. Composites consist of piezoceramic and polymer seem to meet this desire, since they have advantages over single phase ceramics. This is due to their physical properties such as Bulk modulus, density,...etc., which are low compared to those of ceramics. This allows the composites to possess low physical values, such as, acoustic impedance, etc., but considerably high flexibility, which are able to suit for coupling with low impedance load and non-flat surface such as human tissue. Regarding to the connectivity of the piezoceramic and polymer phase, there are many types of mixing (Safari, 1994). Among these types the 0-3 connectivity is considerably easy to fabricate and this is the type that we employed for this work. However, distribution of the ceramic phase proved to have so much effect on its properties therefore, study of the distribution of the ceramic phase prepared by calendering (Fries and Moulson, 1994) and centrifuge (Nhuapeng and Tunkasiri, 2002) methods were carried out. Other physical properties such as density, acoustic impedance...etc., were also measured for comparison.

There are many methods of preparing these (0-3) composites, for examples, calendering and tape casting (Fries and Moulson, 1994), painting (Hanner et al., 1989), colloidal (Han et al., 1991) and centrifuge (Nhuapeng and Tunkasiri, 2002). The volumetric percentages of piezoceramic loading are 50, 60, 70, 70 and 65 respectively.

Another purpose of this work, therefore, was to study the physical properties of the piezoceramic-polymer composites at various fractions of filler. Polyethylene and polyester resin were employed as the matrix phases in fabricating of the composites using calendering and centrifuge methods respectively. Lead zirconate titanate (PZT) powder from Advanced Ceramic Limited type 40/30 was chosen as the filler.

MATERIALS AND METHOD

In order to obtain bigger powder of PZT the as-received powder was atmospherically sintered at 1200 °C, pulverized and sieved through nylon mesh (number 100). The distribution of the particle size of the sieved particles was scanned using a Shimazu sedimentograph (model Analysette 22). The arithmatic mean of the particle size was 150 μ m. The samples were prepared with various volumetric percentage of PZT 40, 50 and 60. Calendering and centrifuge methods were used to fabricate the composites with different matrix phases as mentioned before. Composites with 65% of the filler could not be achieved. This might be the particles being too big in packing. Therefore the powder was pulverized again and sieved through mesh number 300. The measured distribution of the particle size showed the mean size to be smaller (55 μ m). Fabrication of composites was carried out again with 65% of the filler. However, the results showed that only the centrifuge method produced the successful samples.

The acoustic impedance (Z) of a material is related to its mechanical properties by the equation

$$Z = c_{\rm L} \rho = ({\rm K} \rho)^{0.5}$$
 (1)

where c_L is the longitudinal wave velocity in the material, ρ and K are the material's density and Bulk Modulus respectively.

The acoustic impedance of the composites with various fraction of filler was measured using the method described by Bui (1988). Physical properties of the composites, such as density (ρ), longitudinal wave velocity (c_L) and Bulk modulus (K), were measured and calculated respectively. The distribution of ceramic phase in the composites was investigated using a scanning electron microscope (SEM).

RESULTS AND DISCUSSION

It can be seen that the composites fabricated by spinning in a centrifuge could load 65% of PZT particles which could not be achieved in the conventional calendering method. The density (ρ), longitudinal wave velocity (c_L), acoustic impedance (Z) and Bulk modulus (K) of the composites at various fraction of PZT are shown in Table 1., together with those of Grewe et al., (1989); Wersing (1986); Slayton and Setty, (1990). Furthermore the density and the acoustic impedance were found to vary as the ceramic fraction in the composites.

The Z values are in the range of 6.80-11.41 Mrayls and 7.43-12.43 Mrayls for composites prepared by calendering and centrifuge methods respectively. It is found that the results are comparable to that reported by Grewe et al., (1989); Wersing (1986); Slayton and Setty, (1990), though the fabrication routes are different. The Z values fabricated in the centrifuge show higher than that of the samples prepared by the

calendering method. This may be due to the density of the polyester resin higher than that of polyethylene. The Bulk modulus values show the same trend as that of Z, as seen in Table 1.

Table 1. shows the longitudinal velocity (c_L) obtained from the echo-shift method together with the Bulk modulus values calculated using equation (1). Both the longitudinal velocity and the Bulk modulus were found to increase with the increasing of the volumetric fraction of the PZT particles.

Samples	Vol%	ρ	cL	K	Z
		(g/cm^3)	(m/s)	$(kg/s^2.m)$	(Mrayls)
PE+PZT(40/30)	40	3.670	1852.8	12.5994	6.80
	50	4.516	2032.7	18.6608	9.18
	60	5.026	2270.2	25.9029	11.41
PZT(40/30)	100	7.890	3585.5	101.4352	28.29
Resin+ PZT(40/30)	40	3.690	1995.7	14.8280	7.43
	50	4.361	2219.6	21.4864	9.68
	60	5.045	2277.1	26.1633	11.49
	65	5.290	2349.7	29.2069	12.43
Grewe et al. (1989)	40	-	-	-	7.50
Wersing (1986)	100	-	-	-	30.00
Slayton and Setty (1990)	70	-	-	-	~8.00
	70	-	-	_	9.90

Table 1. Physical properties of the composites at various volumetric fraction of filler.

Figure 1 and 2 show scanning electron micrographs of the distribution of the PZT powder in the composites prepared by calendering and centrifuge methods respectively. The fabrication by the centrifuge method shows more homogeneous distribution of the ceramic particles with reasonably equal size in the polymer phases. Consequently, other preperties (as tabulated in Table 1.) shows considerably higher values. The smaller PZT-powder particles (55 μ m) seem to pack closer and denser than the larger PZT particles (160 μ m).



(a)



(b)

Figure 1. Scanning electron micrographs of the composites prepared by conventional method: (a) 50 vol%, (b) 60 vol%.



(a)



(b)

Figure 2. Scanning electron micrographs of centrifuging 40/30 PZT and resin composites: (a) 60 vol%, (b) 65 vol%.

CONCLUSIONS

Fabrication of 0-3 piezoceramic-polymer composites was carried out in two methods namely, calendering and centrifuge. The Z values obtained from both methods are close to each other and also close to that obtained by others, though prepared by different methods. The bulk modulus and the Z values obtained from different polymer phase composites increase with the increasing of the composite density. In the composites with smaller PZT particles fabricated through centrifuge method, the particles distribute more homogeneously and pack together closer and densers than that of the coarse ones which resulted in higher values of their physical properties.

ACKNOWLEDGEMENTS

The authors would like to express their sincere thanks to the National Metal and Materials Technology Center of Thailand for funding of this project. The authors would also like to thank Dr. L. D. Yu for his useful comments and correction of the manuscript.

REFERENCES

- Bui, T., and H.L.W.C.J. Unsworth. 1988. Specific acoustic impedances of piezoelectric ceramic and polymer composites used in medical applications. J. Acoust. Soc. Am. 83 : 2416-2421.
- Fries, R., and A.J. Moulson. 1994. Fabrication and properties of an anisotropic PZT/polymer 0-3 composite. *J.Mater.Sci.Mater.Electron.* 5 : 238-243.
- Grewe, M.G., T.R. Gururaja, R.E. Newnham and T.R. Shrout. 1989. Acoustic properties of particle/polymer composites for transducer backing applications. *IEEE Ultrasonic Symp.* p.713-716.
- Gururaja, T.R., W.A. Schulze, L.E. Cross, R.E. Newnham, B.A.Auld , and Y.J.Wang. 1985. Piezoelectric composite materials for ultrasonic transducer application-part 1. *IEEE Trans Sonics Ultrasonics* SU-32. p.489-497.
- Han, K.H., A. Safari, and R.E. Riman. 1991. Colloid processing for improved piezoelectric of flexible 0-3 ceramic-polymer composites. J.Am.Ceram.Soc.74 : 1669-1702.
- Hanner, K.A., A. Safari, R.E. Newnham and J. Runt. 1989. Thin film 0-3 polymer/piezoelectric ceramic composites: piezoelectric paints. *Ferroelectrics* 100: 255.
- Nhuapeng, W., and T. Tunkasiri. 2001. Properties of the 0-3 lead zirconate titanatepolymer composites prepared in a centrifuge. J. Am. Cer. Soc. (In press).
- Safari, A. 1994. Development of Piezoelectric Composites for Transducers. J.Phys. III France 4 :1129-1149.
- Slayton, M.H., and H.S.N. Setty. 1990. Single later piezoelectric-epoxy composite. *IEEE Inter. Symp. Appl. Ferroelectrics*. p.90-92.
- Wersing, W. 1986. Composite poezoelectrics for ultrasonic transducer. *IEEE Inter. Symp. App. Ferroelectrics.* p.212-13.