Asian Journal of Chemistry; Vol. 26, No. 17 (2014), 5883-5886

ASIAN JOURNAL OF CHEMISTRY

http://dx.doi.org/10.14233/ajchem.2014.18308

Effect of Carbon Particles on the Permittivity and Acoustic Performance of Epoxy Compounds†

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AJC-15814

The two-phase composite materials are fabricated by taking advantage of blending method, with the selecting substrate of epoxy 618 and carbon particles as the additive. Epoxy 618, curing agent and flexibility are mixed with the proportion of 10:1:1. The dibutyl phthalate is used as flexibility, ethylenediamine is selected as curing agent and the carbon particle is used as additives to improve the performance of epoxy compounds. Samples with a variety of mass ratios have been made. The dielectric properties and acoustic properties of the composites are measured. The results show that the dielectric constant, acoustic velocity and impedance of the composite material increases with the mass ratio of carbon.

Keywords: Composite materials, Dielectric constant, Acoustic velocity, Acoustic impedance, Carbon.

INTRODUCTION

When the epoxy resin (epoxy) is cured by the curing agent, it forms three-dimensional network structure inside, which has a good corrosion resistance, excellent mechanical strength, excellent processing performance, low shrinkage characteristics and so on, but also with lower permittivity, which is about 4.6-5.0[1] and hinders epoxy 's practical application of high dielectric constant. Two or more single-phase materials are composited to modify single-phase materials through taking advantage of the complementary of each material[2]. The piezoelectric composite material is such a successful example. The piezoelectric material's high impedance leads to the difficulty in the match with the air, water and human tissue and its hard brittle texture also makes itself difficult to be machined. By adding flexible polymer into piezoelectric material, researchers have gained the piezoelectric composite materials. This new composite materials has excellent piezoelectric properties, low acoustic impedance, easily researchers have gained the piezoelectric composite materials. This new composite materials have excellent piezoelectric properties, low acoustic impedance, easily machined and soft . Thus, the two-phase composite material can be acquired by adding the materials with high dielectric constant into epoxy, this helps to improve not just the dielectric properties of the epoxy, but its machinability. Therefore, this composite can be widely used in the high dielectric areas[3]. Maxwell-Wagner-Sills (MWS™) can be caused by adding the conductive particles into the polymer, thereby the dielectric constant of the composite material increases. The researchers have also studied other conductive particles, such as metal particles, carbon particles and other zero dimensional nanomaterials[4]; single-walled carbon nanotubes (SWNT™), carbon fiber and other one-dimensional nanomaterials[5,6]; graphene, graphite sheet and other two-dimensional nanomaterials[7]. In short, there are a variety of additives with better performance and high relative dielectric constant, but not suitable to be used as the additive material, such as iron, which is heavy and not well compatible with the epoxy, occurring severe delamination during curing if it is stirred sufficiently. With preferable properties, here carbon is selected as an additive material[8]. On one hand, different levels of samples of compound material are acquired. On the other hand, the influences of different weight fraction (Wt %) of several kinds of solid particles on the properties of composite dielectric and acoustic are presented in this paper.

EXPERIMENTAL

Fabrication of composite: The main materials that used for fabricating the composite are presented as follows, Epoxy 618 (Beijing Pulindaye Chemical Company™); ethylenediamine, AR (Xilong Chemical Company™); dibutyl phthalate,

†Presented at 2014 Global Conference on Polymer and Composite Materials (PCM2014) held on 27-29 May 2014, Ningbo, P.R. China

AR; carbon particles, chemically pure (Sinopharm Chemical Reagent™).

In this paper, blending method is adopted for the composite fabrication. Firstly, the epoxy and dibutyl phthalate are mixed at the weight ratio of 10:1 under the room temperature. Secondly, the carbon particles with different proportions (Table-1) are added into the mixture as needed. Thirdly, taking ethylenediamine as the curing agent, with the proportion of 1:10 to epoxy and fully stirring all the materials. So it is necessary to put the mixture into a vacuum oven (DZF-6020 type) for 5 min to remove air bubbleswhich affect the density of the composite materialgenerated due to the chemical reaction between epoxy and curing agent. Then pour the mixture into the mold. Especially, the composite material also needs to discharge a few bubbles during the pouring to achieve a better performance. Afterwards place it in the oven at 80 ºC for 5 h for curing. Grind the surface of the demould composite material and cut it into a rectangular with the width of 15 mm and the height of 5 mm by the cutting machine. Finally, sputter electrode at the surfaces of the sample and test the dielectric and acoustic performance of sample at room temperature.

Measurement of composite permittivity: The dielectric constant of the composite is calculated by the eqn. 1:

$$\varepsilon_{\gamma} = \frac{c_p d}{(s\varepsilon_0)} \quad (1)$$

wherein c_p stands for quasi-static capacitance, d represents the height of the sample, s is the sample cross-sectional area, ε_0 is the vacuum dielectric constant $\varepsilon_0 = 8.85 \times 10^{-12}$ F/m.

Measure the length (a), width (b), height (d) of the sample by vernier caliper with accuracy of ± 0.2 mm and then calculate the cross-sectional area (s = a * b). Use the precision impedance analyzer (Agilent 4294A) to obtain quasi-static capacitance C_p at 1 kHz and take these data into eqn. 1 to calculate the dielectric constant.

The dielectric constant characters the ability of bound charge of dielectric material. It has different requirements at different occasions. So it is necessary to selecting material with different dielectric constant according to the need in practical application. Dielectric constant is connected with the frequency. The data is measured at frequency of 1 KHz in this paper. It is the SEM image with 5000 Times (Fig. 1), which is composite added carbon particles, wherein bright particles are carbon and black part are epoxy. It showed that the average particle size of carbon particle is 8 μm and the additive does not appear a large agglomeration and substantially uniform distribute in the substrate from this picture.

They are 5000 times SEM images of C/Epoxy composite samples (Fig. 2). These pictures (Fig. 2) indicate that the density

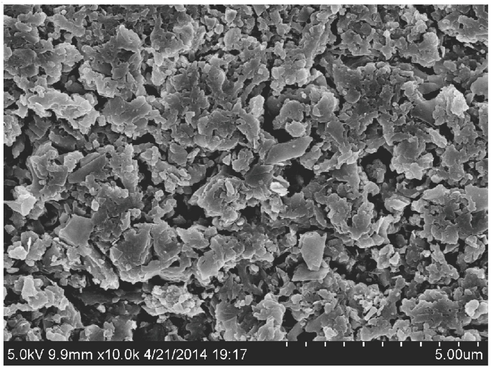

Fig. 1. SEM Images with 10000 times of composite added carbon

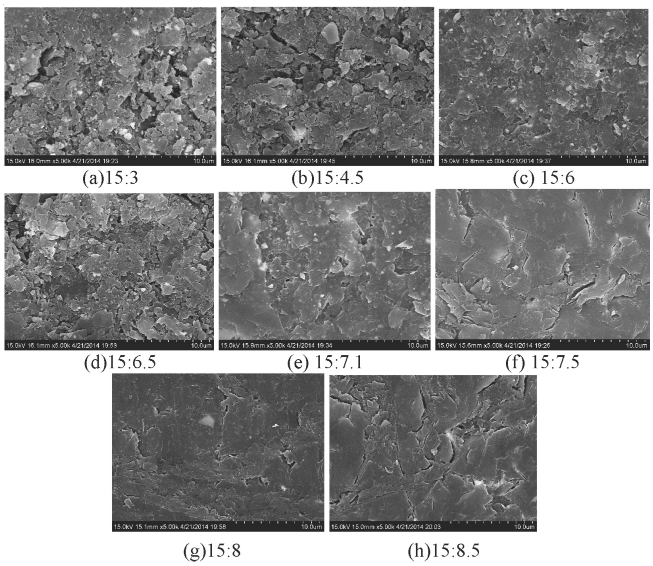

Fig. 2. SEM Images with 5000 times of polished surface of C/epoxy composites

of additions increases with the content of carbon particles. The carbon particles independently distribute in the epoxy when the weight fraction of carbon is small in Fig. 2(a-d). It can be seen that from Fig. 2(e-d), carbon particles connected with each other in epoxy substrate with relatively big weight fraction, which make the dielectric property of the composites increases suddenly (Fig. 3).

Fig. 3 indicates that the dielectric constant of carbon particles changes with weight fraction of carbon particle, the dielectric constant of the composite increases with the additive's weight fraction and the dielectric constant dramatically increases when the weight ratio of the additive reaches 30 % (Fig. 3). The dielectric constant of epoxy is less than 200 and slowly increase when the weight fraction is less than 30 % (Fig. 3). However the curve begins to rise sharply when the weight fraction reaches 30.23 %. And the dielectric constant

TABLE-1
WEIGHT RATIO OF RAW MATERIALS

Sample number	1#	2#	3#	4#	5#	6#	7#	8#
	Weight ratio							
C	3	4.5	6	6.5	7.1	7.5	8	8.5
Dibutyl phthalate	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5
Ethylenediamine	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5
Epoxy	15	15	15	15	15	15	15	15

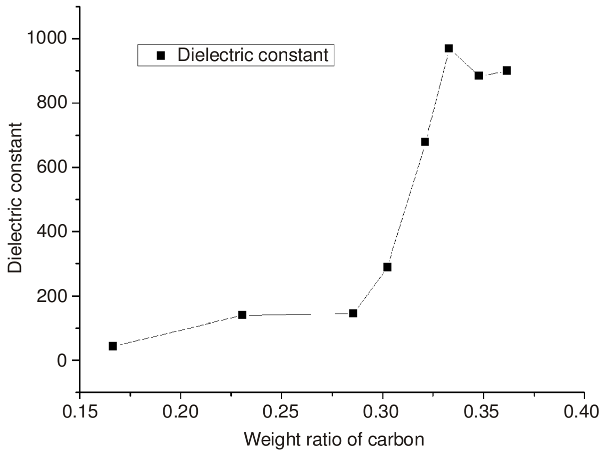

Fig. 3. ε_γ of C/epoxy composites

of composite increases from 288.29 to 677.26. The dielectric constant tends to maintain highly growth until the carbon weight fraction of 33.30 %. At the same time, the maximum dielectric constant is achieved 968.5, then the dielectric constant decreases with weight fraction increasing. In general, the permittivity of epoxy has greatly improved (Fig. 3). The dielectric constant of composite will decrease if continuing to increase the filling amount of the carbon particles, because that the carbon particles are prone to agglomeration and the bubbles may be introduced, which will affect the electrical and mechanical properties of composite. This study finds that the mixture is very dense and has reduced liquidity when the weight fraction of carbon powder is more than 40 %, which is not fit fill the infusion process that this study concerns. In a word, adding carbon particles into epoxy can improve the dielectric constant of the composite. There are two main reasons, firstly, the carbon particles as an additive added to the epoxy is equivalent to introducing a large number of tiny capacitors into the epoxy substrate according to the micro-capacitance theory. These tiny capacitors can increase the dielectric constant of the composite. Secondly, the carbon particles are added into the epoxy to gain the two-phase composite, the interfacial polarization occurs when charge accumulation appears on the interface of dissimilar materials, all of these results in dielectric constant increasing. It has been proved that the dielectric constant of the composites increases with the weight fraction of carbon particles in this experiment. The relative dielectric constant of the epoxy is about 700 when the carbon powder is about 30 Wt % and the dielectric constant of epoxy has improved hundred times. When the weight fraction is close to the percolation threshold, the dielectric constant has a mutation.

Testing the acoustic impedance of the composite: The acoustic impedance of the composite is determined by density and sound velocity, currently, sound velocity measurement methods used widely are mainly traditional pulse-echo method, resonance method and ultrasonic spectrum method. The pulse-echo method, which is selected to measure sound velocity in this paper, is widely used method to calculate the sound velocity through eqn. 2.

$$\nu = \frac{2d}{t} \tag{2}$$

wherein eqn. 2, d represents the height of the sample, t stands for the time lag of the two echo signals.

Specifically, the electronic scale with the precision of 0.1 g is used to gain the weight of each sample m and an accuracy of 0.2 mm vernier calliper is selected to measure samples' length (a), width (b), height (d), then obtain density, $\rho = m/(a \times b \times d)$. The pulse signal device (5072PR) and a digital oscilloscope (Agilent 54622A) constitute a pulse measurement system through the method of ultrasonic pulse echo. Generating a high frequency pulse signal (pulse signal of 5 MHz optional) by pulse signal device, an ultrasonic probe passes the sound signal to the upper surface of tested sample, the pulse signal is reflected by the lower sample surface, which forms the echo signal, the echo signal is received by the ultrasonic probe and displayed on an oscilloscope. The time lag between the two echo signals and the sample thickness are measured, then taking these data into the eqn. 2 to obtain the velocity.

The electrical impedance is defined as AC voltage dividing electric current in electricity, it is an analogy to the acoustics, the acoustic impedance is the ratio of sound pressure to vibration velocity of the particle in medium, composite amplitude of sound pressure is eqn. 3, the amplitude of the vibration velocity of the particle is the eqn. 4.

$$P = A\omega\rho c \tag{3}$$

$$V = A\omega \tag{4}$$

A represents amplitude, ω represents the angular frequency of vibration, ρ means medium density, c is expressed propagation velocity in the medium.

Acoustic impedance equation is obtained according to the definition:

$$Z = \frac{P}{V} = \rho c \tag{5}$$

The eqn. 5 indicates that the acoustic impedance is decided by the density of the medium and the acoustic velocity. The acoustic velocity is connected with itself, the media's state, the physical properties, the amplitude , the frequency and the vibration mode (longitudinal or transverse wave) of the sound wave, which will affect the acoustic velocity. The states of the medium include density, elasticity, temperature and pressure *etc*. The material properties are mainly determined by the mechanical properties.

RESULTS AND DISCUSSION

The acoustic performance of these samples was tested. In this paper and the test results of three samples shown in Fig. 4.

The acoustic velocity of C/epoxy composite increases with the weight ratio of C (Fig. 4). From the above analysis, it can be concluded that the acoustic velocity value depends on the density of the composite under the same conditions, such as the physical properties. The density of the composite decreases while the weight ratio of carbon particles increases in the C/epoxy composites. That is because the density of C is smaller than epoxy 618. At the same time of the acoustic velocity's increase, the changes between the minimum of 2.803 m/s and the maximum of 3.068 m/s indicate that the growth is not evident. Acoustic impedance synthetically decided by both

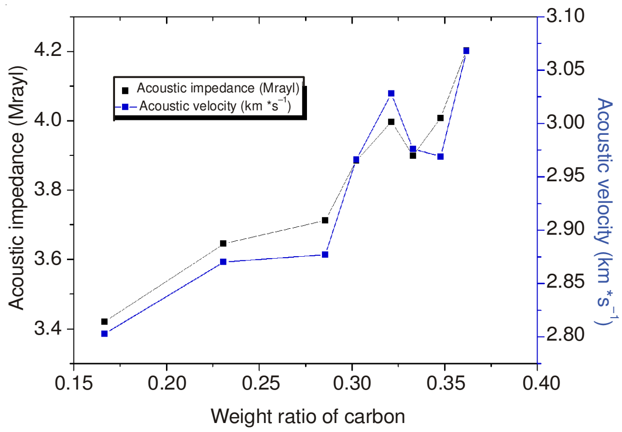

Fig. 4. Acoustic impedance and velocity of C/epoxy composite

the density and the sound velocity of the composite material. It can be concluded that acoustic impedance of the composite increases with the weight fraction of carbon particles in Fig. 4. Acoustic impedance has changed wildly while compared with the acoustic velocity. Acoustic impedance of C/epoxy composite increases from 3.42 to 4.203 Mrayl. Although the acoustic impedance of C/epoxy composite has shown up an increasing trend, the acoustic impedance and acoustic velocity have reduced at the point of the dielectric constant of C/epoxy composite's sudden increase. The reasons are not clear at present and need to be further studied. The epoxy is a substrate, preparing the composite material by adding carbon particles, the value of the composite acoustic impedance is determined by the weight ratio of the carbon particles and it increases with the ratio, which can be concluded from Fig. 4.

Conclusion

The dielectric constant of C/epoxy composite material changes a lot and the acoustic impedance variation is relatively small at the same time, while different types of carbon particles are added into epoxy. It's contrary to the C/epoxy composite, the BT/epoxy and W/epoxy composite changes greatly for acoustic impedance and is relatively small for dielectric constant. Three kinds of additives have little influence on the acoustic velocity of the composite.

ACKNOWLEDGEMENTS

This work was supported by the National Natural Science Foundation of China under Grant No.61302015; the Scientific Research Key Program under Grant No. KZ201110772040; General Program of Beijing Municipal Commission of Education under Grant No. SQKM201211232022; and the Open Fund of Beijing Key Laboray of Optoelectronic Measurement Technology under Grant No. GDKF2013004.

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