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Effect of Emulsion Temperature on Properties of Conductive Epoxy Porous Prepared by Single Emulsion Technique

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Abstract. Currently, conductive porous is used in composite to reduce the conductive filler loading and improve the conductive property. In this study, conductive epoxy porous (CEP) had been fabricated by single emulsion technique from the mixture of epoxy, polyamide hardener, carbon black filler and sodium bicarbonate. Epoxy mixture was dropped into a heated corn oil at different temperatures (140, 160 and 180 °C) under stirring of 1000 rpm. Because of the immiscibility of epoxy and corn oil, epoxy droplets were formed. Initial epoxy droplets were broken into smaller droplets due to applied stirring shear. In addition, the decomposed gas (carbonic gas and water vapour) from sodium bicarbonate also caused an internal expanding pressure to further break the epoxy droplet into many fine droplets. Curing reaction of epoxy and polyamide occur simultaneously formed CEP. It was found that all the CEPs exhibited higher conductivity compared to control epoxy and epoxy composite filled equivalent content of carbon black. Furthermore, emulsion temperature of 160 °C produced the smallest porous CEP, which performed the highest electrical property.

1. Introduction

Conductive microspheres have been paid excessive attention by researchers and industries due to many advantageous properties such as similar density to polymer matrix, high specific surface area, excellent thermal property and reasonable electrical conductive property [1,2]. Conductive microspheres can be obtained by coating conductive filler on the surface of non-conductive microspheres. The common microspheres used widely are glass microspheres, cenospheres, carbon microspheres and polymer microspheres [3, 4, 5]. Hence, conductive microspheres have been used to replace bare conductive filler in conductive polymer composites and applied in electronic, biomedical and sensor devices [6].

Numerous occasions researcher used emulsion technique to produce conductive microspheres. Emulsions are systems consisting of two or more liquid immiscible phases wherein one liquid is dispersed in another liquid phase [7]. Previous study reported fabrication of conductive epoxy porous by single emulsion method. The epoxy mixture was dropped and stirred in heated corn oil. Due to immiscible between epoxy mixture and corn oil, the epoxy mixture droplets form a spheres shape in heated corn oil. These epoxy mixture droplets contain blowing agent, thus, this epoxy droplets blown and simultaneously cured after receiving heat from corn oil [8]. To obtain good porous materials, the blowing agent has to be carefully selected depending on processing condition sort of temperature [9]. Elzbieta & Pawel stated that temperature has influence in liberation gas once blowing agent decompose resulted different expansion formed in polymer matrix [10]. In this study, conductive

epoxy porous prepared by single emulsion technique and the aim of this research was to discover the influence of emulsion temperatures of corn oil has an effect on the properties of conductive epoxy porous.

2. Experimental

2.1. Materials

Epoxy resin DER 331 and polyamide A062 was purchased from Euro Chemo-Pharma Sdn. Bhd. Epoxy resin DER 331 has epoxide equivalent weight of 182–192, density and viscosity at 25 °C of 1.16 g/cm^3 and 11–14 Pas. Polyamide A062 has equivalent weight per H active of 110, density and viscosity at 25 °C of 0.96 g/cm^3 and 35–45 Pas, respectively. Polyamine was purchased from Euro Chemo-Pharma Sdn. Bhd. White crystalline powder of sodium bicarbonate was purchased from HmbG Chemicals having density of 2.22 g/cm^3 was used as the blowing agent. Corn oil was purchased from Euro Chemo-Pharma Sdn. Bhd and light yellow in apperance. Corn oil has density and viscosity at 25 °C of 0.91 g/ml and 50 Poise. Carbon black N330 (HAF) was purchased from Mega Makmur Saintifik Sdn. Bhd. Carbon black has iodine absorption number of 82 ± 5 g/kg and CTAB surface area of 79~87 10^3 m^2/kg.

2.2. Preparation of Conductive Epoxy Porous (CEP)

The porous were prepared based on polyamide dominant which is polyamide-epoxy adducts. The weight ratio of epoxy and polyamide was 1E:2P. Epoxy was mixed with sodium bicarbonate by using over-head stirrer with the speed of 300 rpm for 3 minutes. Carbon black was added continuously and mixed at the speed of 300 rpm for another 2 minutes. Lastly, polyamide was added into the mixture and stirred for 3 minutes at the same speed. Epoxy mixture was continuously dropped into corn oil at 160 °C (weight ratio of epoxy mixture to corn oil of 1:9) and stirred at 1000 rpm for another 1 hour. The laboratory syringe was used as dropper with a nozzle diameter of 0.5 cm to drop epoxy mixture into corn oil. The porous was produced by allow it to cool down at 80 °C before filtered and washed five times with detergent solution at 60 °C. The ratio of detergent to water used is 1:20. The detergent solution was changed every 15 minutes. Drying and post-cure process were carried out at 80 °C for 4 hours in air-circulating oven.

2.3. Preparation of Epoxy filled CEP for Conductive Testing

The weight ratio of epoxy and polyamine was 2:1. 20 phr of CEP was mixed with epoxy by using over-head stirrer with the speed of 300 rpm for 3 minutes. Polyamine was added into the mixture and stirred for another 3 minutes at the same speed. The mixture was casted into PP mold with dimension of 24 x 20 x 4 mm and allowed to harden at room temperature for 24 hours.

2.4. Characterization

The morphology of CEP was carried out using a scanning electron microscope (SEM), model JSM 6260 LE JOEL. CEP was coated with gold/platinum layer to avoid electrostatic charging during examination. Bulk density of CBEP was determined according to ASTM D7481-09. Bulk density was carried out by filling 100 ml of CEP in 100 ml cylinder. The mass of 100 ml CEP was weight. Bulk density was calculated by using mass diving to volume. Pycnometer density of CEP was measured by using gas pycnometer density analyser, AccuPyc II 1340 V1.0.5. The 10 cm^3 of size chamber and Helium gas was used. Differential scanning calorimetry (DSC) of CEP was measured in temperature range from 25–200 °C with a heating rate 10 °C/min under nitrogen atmosphere. Thermo-gravimetric analyzer (TGA) of CEP was determined in temperature range from 25–600 °C with a heating rate at 20 °C/min under a nitrogen flow of 50 ml/min by using Pyris Diamond TGA from PerkinElmer. Electrical conductivity test of CBEP was obtained by using Keithley 4200 Semiconductor Characterization System.

3. Results and Discussion

3.1. Morphology Analysis

Epoxy droplets receiving heat from corn oil would induce two reactions such of curing reaction and decomposed reaction of sodium bicarbonate. These both reactions depend greatly on the heating temperature. Figure 1 displayed the morphology of CEP prepared in different emulsion temperatures. T140 showed bigger porous particles compared to these of T160 and T180. Furthermore, epoxy particles in T140 agglomerated to form large lumps. In expected, T180 possessed the smallest epoxy particles and the most separated epoxy groups compared to others (T140 and T160). It is because the fast decomposition of sodium bicarbonate at 180 °C produced carbonic and vapour too vigorously that low viscosity epoxy droplets could be burst into many small droplets.

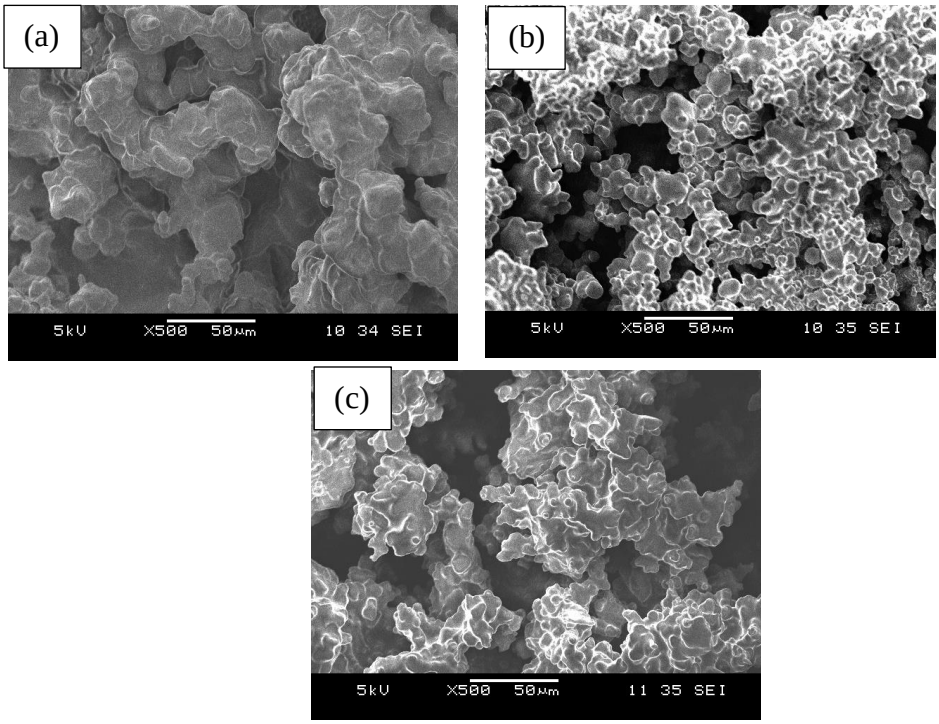

Figure 1: Scanning electron microscopy images of CEP for different emulsion temperatures at 500X magnification: (a) T140, (b) T160 and (c) T180.

3.2. Density Analysis

Figure 2 demonstrated the effect of different emulsion temperatures towards bulk density and pycnometer density of conductive epoxy porous. Figure 3 showed volume of CEPs in a same mass. Bulk density described as a direct measure of the packing closeness for particles in volume [11]. Surprisingly that T180 showed the highest bulk density while T160 possessed the lowest bulk density. It could be T180 had more small groups of epoxy particles, and then they can pack easier and occupy less spacing compared to others. T160 could have more bulky groups which composed from attaching epoxy particles and large pores. Therefore, same weight of T160 could occupy the highest volume compared to other hence exhibit the lowest bulk density as reported in Figure 3. This bulk density promised the potential of CEP dispersing well in epoxy composite and lead to better conductivity, which will be discussed in the next section. In contrast, pycnometer density decreases with increasing in emulsion temperatures. It could be some closed pore presenting among the packing epoxy particles in T160 and T180 so the Helium gas during pycnometer measurement could not able to enter. Hence,

the density of T160 and T180 were lower than T140. Especially in T180 which had more epoxy packing from small epoxy particles.

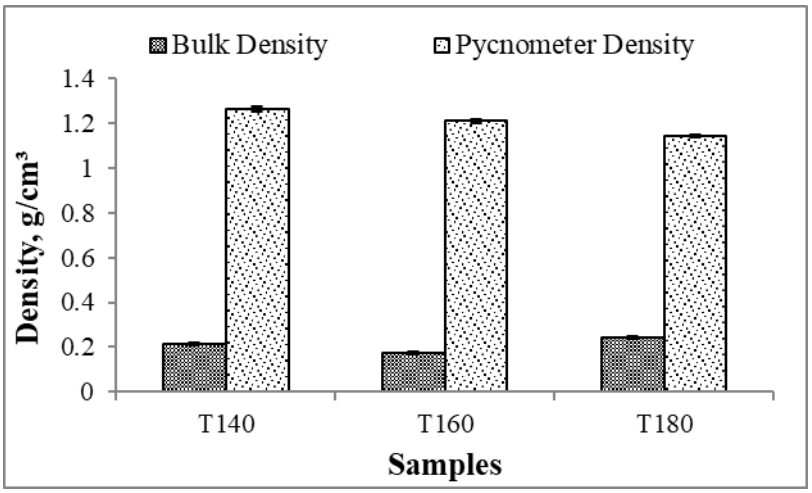

Figure 2: The effect of different emulsion temperatures of conductive epoxy porous towards bulk density and pycnometer density.

Figure 3: Volume of CEPs in a same mass: (a) T140, (b) T160 and (c) T180.

3.3 Thermo-gravimetric Analyzer (TGA)

The results of TGA weight loss and derivative thermograms of CEPs was reported in Figure 4.10(a) and 4(b). According to Figure 4(a), there is no decomposed peak occurred at before 160 °C so all sodium bicarbonate was decomposed completely. It was interested to find that the onset degradation was in well agreement to the bulk density results, where T160 started the earliest degradation following by T140 and lastly T180. Onset degradation depends strongly on the heat circulation in the sample. T160 had lowest bulk density, more volume occupying, larger open pores, hence more heat circulation and easier to be burnt. The best onset degradation of T180 was thanks to its closed pore, which was also proven from pycnometer density result. However, once the degradation start T180 exhibited the fastest rate of degradation because it had the smallest epoxy particles. After starting degradation, T140 had better heat conductivity hence it could be degraded faster than T160. These facts explained the results of temperature at 50% weight loss.

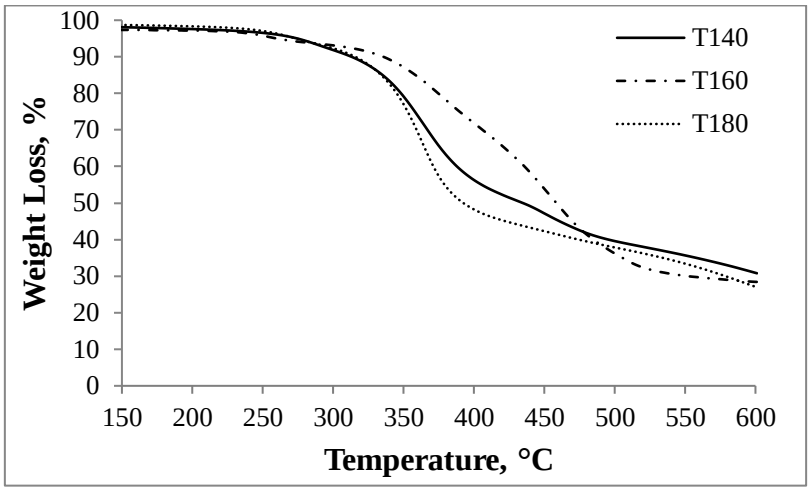

Figure 4(a): Weight loss of CEPs for different emulsion temperatures as a function of temperature.

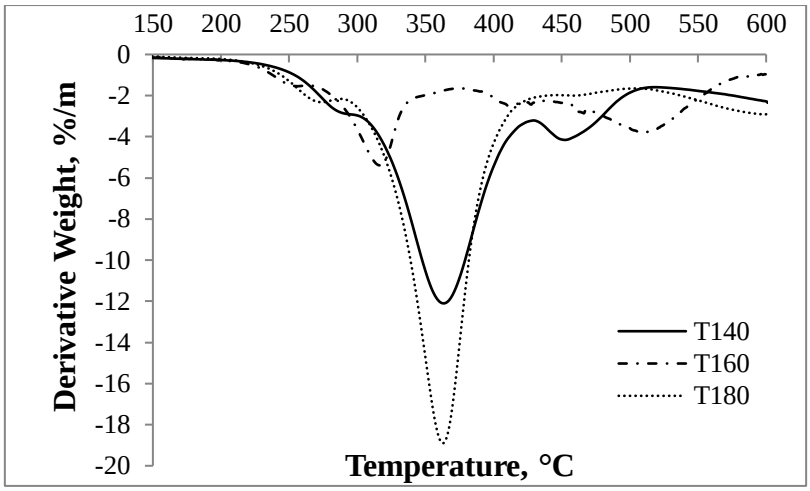

Figure 4(b): Derivative weight loss of CEPs for different emulsion temperatures as a function of temperature.

3.4 Conductive Behaviour

Figure 5 illustrated comparison of conductivity behaviour toward different epoxy composites. It was found that electrical conductivity of epoxy filled 20 wt% of CEP fabricated from different emulsion temperatures possessed highest electrical conductivity compared to control epoxy (control) and epoxy composite filled equivalent content of carbon black (CB). It can be implied that addition of 20 wt% of CEP into epoxy-polyamine composites exhibited better electrical conductivity compared to direct equivalent carbon black filler when same amount of carbon black (1.8 %) was used. This conductive behaviour proved the advantage of the proposed CEP, where the conduction was occurred via the connection of carbon black in composite. According to Figure 5, epoxy filled T160 indicates the highest electrical conductivity, while T140 exhibited the lowest electrical conductivity. This property reflected well the morphology of T160 and the bulk density. T160 exposed larger open pores and lowest bulk density, it means T160 could spread more throughout in epoxy composite. Therefore, the better carbon black dispersion in epoxy composite, and the higher conductivity is. The lower electrical conductivity of T180 could be due to its closed pore in the packing of small epoxy particles.

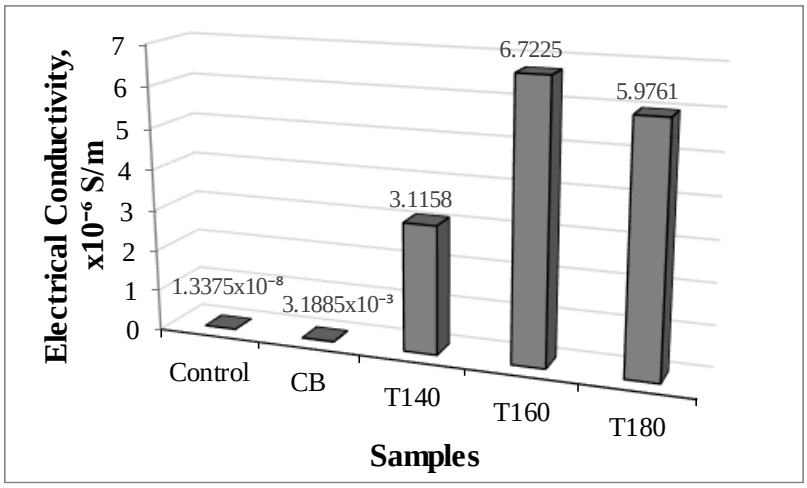

Figure 5. Electrical conductivity of epoxy composites.

4. Conclusion

Conductive epoxy porous was prepared by single emulsion technique. Emulsion temperatures are plays important role to determine properties of conductive epoxy porous. Epoxy mixture was dropped into a heated corn oil at different temperatures (140, 160 and 180 °C) under stirring. It was found that T140 showed bigger porous particles compared to T160 and T180. But, at higher emulsion temperatures of 180 °C, sodium bicarbonate decomposed produced carbonic and vapour too vigorously that low viscosity epoxy droplets could be burst into many small droplets were proved through the morphology. The use of 160 °C (T160) was optimum emulsion temperature offers better expansion behaviour. Besides, epoxy filled T160 exhibited the highest electrical conductivity compared to others because of T160 could spread more in which supported by bulk density result.

Acknowledgements

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