

A Comparison of Thermal Characteristics between Conventional and Electrospun Epoxy Composites

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Abstract—In this paper, a uniform dispersion of nanofillers in epoxy nanocomposites is achieved using electrospinning. Epoxy composites with various amounts of nano and micro aluminum fillers are prepared by conventional mechanical mixing and electrospinning techniques. An infrared laser-based thermal profile test is used to compare the thermal conductivity of the epoxy composites prepared with aforementioned methods. Thermal gravimetric analysis is also used to analyze the thermal stability of nanofilled epoxy composites. The results confirm that the thermal properties of the epoxy composites prepared by electrospinning are improved as compared to conventional sample. In addition samples containing both nano and micro fillers show higher improvement than those with only nanofiller.

I. INTRODUCTION

The further miniaturization of electronic devices and the compaction of electrical power equipment have created new requirements and new challenges for both packaging and electrical insulation. Materials for packaging electronic devices must be capable of conducting heat generated by the devices to avoid overheating [1-2]. Therefore, the most important characteristic of material used for this application is thermal conductivity. Epoxy resin is one of the common material that is used for electrical insulation and packaging [3]. In spite of the excellent performance of epoxy, its relatively low thermal conductivity cannot satisfy the increased demands of new electronic devices.

Fillers are used to enhance the specific properties of materials. Extensive research has been done on the application of microfillers. In some recent works large amounts of microfillers have compounded into polymers to improve the thermal, mechanical or electrical properties of base polymers [4-5]. However, a high level of filler generally degrades the mechanical properties and elaborates the processing. Recent studies have shown that the combination of micro and nano fillers not only can further improve the properties of only micro filled composites but also can obtain the desired properties at much lower filler loading [6-7].

Nanofillers agglomerate easily due to their high surface energy and the agglomeration cannot be avoided by conventional mechanical mixing. These agglomerations of filler negatively

affect the material properties. A uniform dispersion is necessary to realize improvements in the material properties [8-9]. In our previous study, a uniform dispersion of nanosilica in silicone rubber polymer was achieved by electrospinning allowing us to achieve higher loading of nanosilica compared to conventional mechanical mixing [10]. In this study epoxy composites are made using the same electrospinning technique with the addition of $5\mu\text{m}$ and 40nm aluminum filler which is commercial available.

II. EXPERIMENTAL

A. Epoxy Composite Preparation

The epoxy used in this study is epoxy resin 832C bought from MG Chemicals. This resin is a two-part cured epoxy compound, part-A is the epoxy resin, and part-B is the hardener. The mixing weight ratio of part-A to part-B is 2.31:1. It is a translucent and electric grade epoxy which insulates heat and conductivity efficiently. The fillers selected for investigation are 40nm and $5\mu\text{m}$ aluminum.

Viscosity and conductivity of polymers are the key parameters that determine whether the selected polymers can be electrospun [11]. The epoxy used in this study, epoxy resin 832C is hydrophobic, low viscosity and low conductivity. In order to electrospin this epoxy, the conductivity of this polymer has to be modified. Therefore, ethanol was added to the epoxy to increase its conductivity. As the volume of ethanol affects electrospinning process, addition of ethanol should be well controlled. As the epoxy has a low viscosity of 3300cps , only few milliliter of ethanol is enough to make epoxy performs electrospinning properly. Fig. 1(a) shows the schematic diagram of the electrospinning and (b) experimental setup.

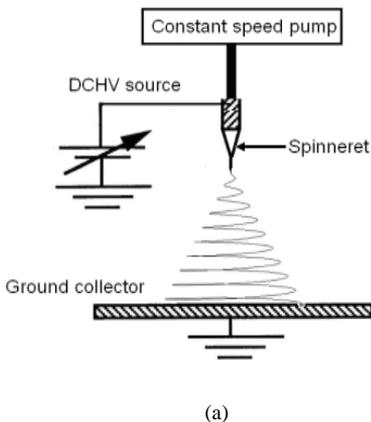


Fig. 1. (a) Electrospinning process and (b) experimental setup

Samples with different amounts of fillers were prepared by two methods, electro spinning and conventional mechanical mixing. Samples made by electro spinning method called electrospun sample (ES). In conventional mechanical mixing, samples prepared by a high shear force Ross laboratory mixer (model HSM-100LSK) and hereafter called con-

ventional sample (CS). In both methods, ingredient of the epoxy composite was calculated based on the weight percent (wt %).

For conventional sample, desired amount of nanofiller was gradually mixed with epoxy part-A in a small portion by the high shear force mixer until there is no visible lumps. The optimum mixing time is empirical based for different formulations. Next, micro-silica was mixed with aforementioned mixture in the same procedure. In order to reduce the heat generated by mixing, the beaker that containing compounds was steeped in cold water during mixing.

For electrospun sample, epoxy part-A and nanofiller were first mixed together in the same way as conventional sample, and then were mixed with a small amount of ethanol which increases the conductivity of the mixture to enable electrospinning of epoxy compound. Well mixed epoxy mixture was delivered to a high voltage energized needle through a connected syringe at a constant flow rate (here is 1mL/min). The applied high voltage varies between 12kV-20kV, and the distance between the energized needle and grounded plate changes between 10cm-25cm. The optimum voltage and distance are adjusted based on different formulations. The collected electrospun epoxy mixture was put in the oven at 65°C for 30mins to further evaporate the ethanol that may be remained in the compound, and then mixed with micro filler in the same way as conventional sample.

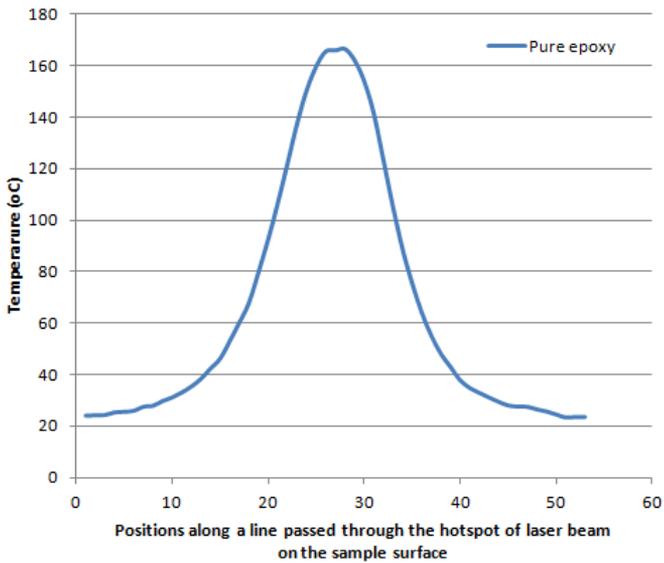
At last, epoxy part-B was added into the blending epoxy compound and was mixed together for about 30second by a low speed mixer. The final mixture was then degassed in the vacuum to get rid of the bubbles and poured into the moulds. The epoxy composites were cured in the room temperature for 24 hour, and post cured at 65 °C for 1hour. All samples prepared were 5cm×5cm with a thickness of 2mm.

B. Infrared Laser-based Thermal Profile Test

A near infrared laser (model FAP) operating in continuous wave (CW) mode at wavelength of 802nm was used to perform the thermal profile analysis and heat resistance testing to examine the local effects of nanofiller distribution. For the thermal profile analysis, the infrared laser power was set at 640 mW to irradiate the nanocomposite surface for 3 min. This power was selected to prevent burning the samples. The laser beam was incident normal to the sample's surface. For all tests, samples were located 5 cm away from the laser source. To ensure that all of the samples will absorb a sufficient amount of heat from the laser beam radiation and that the amount would be identical to that absorbed by the other composites, a thin of carbon black was coated onto the surface of the sample. For this test, it was assumed that the black slice can absorb the same energy from the laser source and also transfer an equal amount of energy to the composites. A FLIR-SC500 infrared camera was used to record the maximum temperature along a line passed through the hotspot on the surface of the samples at both sides. The recording time includes 3 min heating up and 3 min cooling down. The schematic diagram of the laser setup and maximum temperature distribution along a line pass through the hot spot were shown in Fig. 2 (a) and (b), respectively.



(a) Laser setup for thermal profile test



(b) Maximum temperature distribution along a line passed through the hot spot

Fig.2 Laser setup and maximum temperature distribution along a line pass through the hot spot.

After 3 min of laser radiation, the maximum temperature differences between two surfaces of samples were shown in Fig.3.

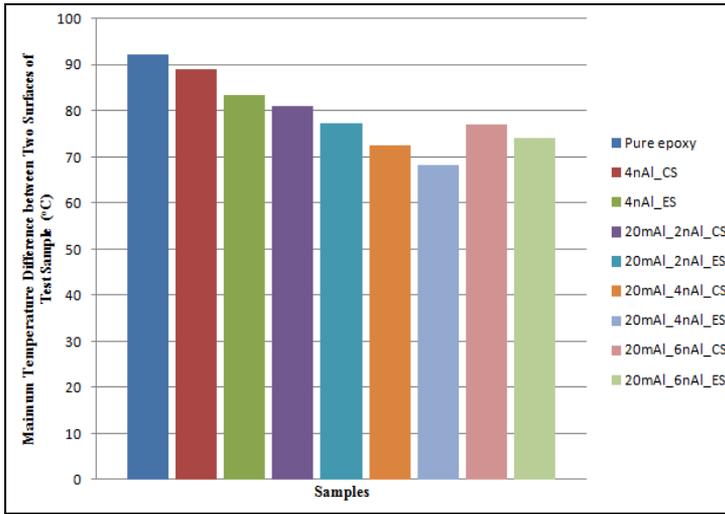


Fig.3 The maximum temperature difference between two- surface of different samples after 3mins laser heating up. CS=Conventional sample, ES=electrospun sample.

As thermal resistance

$$R = \frac{\Delta T}{Q} \quad (1)$$

where, Q is total heat flow through R, ΔT temperature drop across R.

Thermal conductivity is calculated from the thermal resistance

$$k = \frac{t}{R * A} = \frac{t * Q}{\Delta T * A} \quad (2)$$

where, t is the thickness of test sample; A is the heating area of the sample.

For this measurement, Q, A and t are assumed the same for all samples. Therefore, thermal conductivity k is proportional to $1/\Delta T$. Then any two of samples will have following relation:

$$\frac{k_1}{k_2} \propto \frac{\Delta T_2}{\Delta T_1} \quad (3)$$

The thermal conductivity for pure epoxy is 0.169 W/m K, so the conductivity of other samples can be deduced from equation (3). The thermal conductivity of all samples was calculated based on above equations shown in Fig. 4. The thermal conductivity of epoxy composites keeps increasing with the increased filler loadings. Electrospun sample containing 20wt% micro+ 4wt% nano has the maximum thermal conductivity. This maximum thermal conductivity is 35% higher than that of unfilled epoxy. However, there is a

reduction in thermal conductivity when the nanofiller increased to 6wt% for both electrospun and conventional samples, which may come from agglomeration of nanofiller.

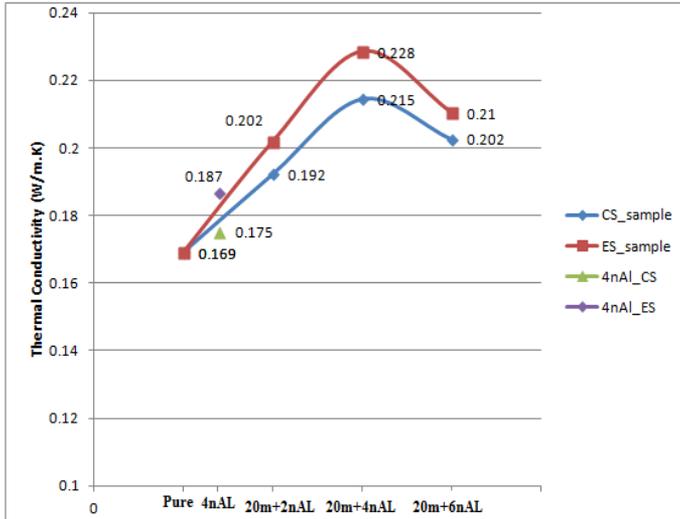
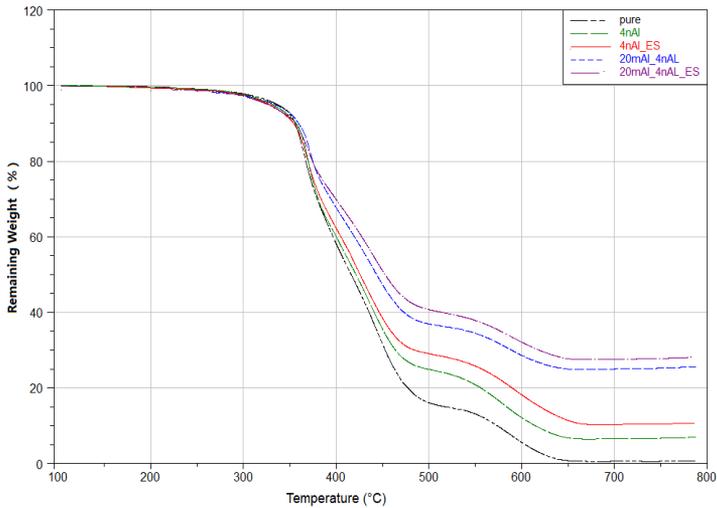


Fig 4. The calculated thermal conductivity of different epoxy samples. CS=conventional sample, ES=electrospun sample.

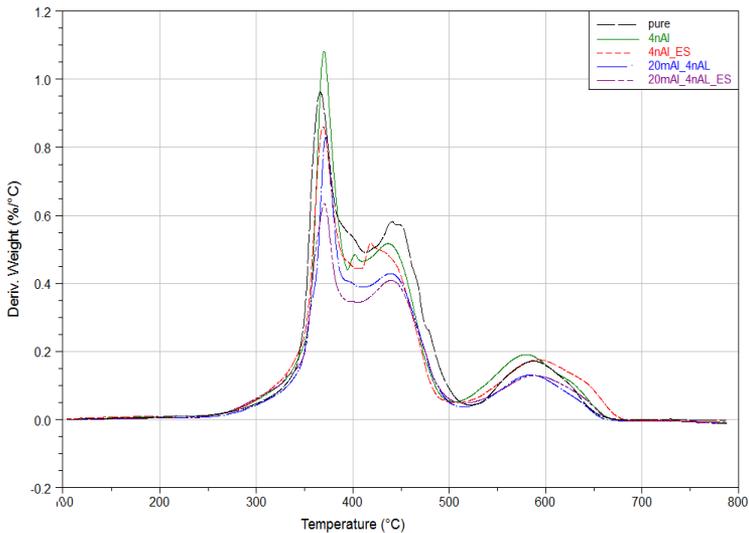
C. Thermo-Gravimetric Analysis (TGA)

Thermo-gravimetric analysis (TGA) is a test to study the weight changes of samples in relation to temperature under a controlled atmosphere, and gives useful information about thermal stability of samples. The TGA tests were conducted by using TA instruments SDT Q600 under ambient air condition, with a temperature rise rate of 25°C/min. The temperature was increased from 100°C to 800°C. The weight loss of samples as a function of time can be achieved by this test.

The test epoxy composites were pure epoxy, 4wt% nano aluminum filled epoxy composites which were prepared by two methods (electrospinning and high shear force mixer), and 20wt% micro+5wt% nano aluminum filled epoxy composites which were prepared by two methods as well. The TGA curves were shown in Fig. 5 (a) and (b).



(a) Weight loss as a function of temperature



(b) Derivative of weight loss as a function of temperature

Fig. 5. TGA curves of epoxy composites with different filler loadings. Pure means pure epoxy, 4nAl means 4wt% nano aluminum, 20mAl_4nAl means 20wt% micro aluminum and 4 wt% nano aluminum, CS= conventional sample, ES=electrospun sample.

From the TGA curves, Fig. 5(a), the initial decomposition temperature start around 350 °C for all samples while Fig. 5 (b) shows the precise different for all samples in terms of the

steepness corresponds to the rate of the weight loss. In Fig. 5(b), the first derivative of the TGA curves was used to precisely compare at what temperature sample decomposition started and the rate of weight change represented initial decomposing degree of samples. The rate of weight change is around 0.95 %/°C for pure epoxy. However, after adding fillers the rate of weight change for 4wt% nano electrospun sample is around 0.85 %/°C and 0.8 %/°C for 20wt% micro +4wt% nano filled conventional sample, respectively. The slowest weight change is about 0.65 %/°C for 20wt% micro+4wt% nanofilled electrospun sample. The only exception is 4wt% nano conventional sample where the rate of weight change is even faster than pure epoxy and it is around 1.5 %/°C. However, all first derivative peaks for composite samples in Fig. 5(b) are slightly moved backward to a higher temperature compared to pure epoxy sample showing improvement of the thermal stability. Moreover, the slow slopes and higher remaining weights of TGA curves for composite samples compared to that of pure epoxy in Fig. 5(a) confirm the improved thermal stability as well. The improved thermal stability is contributed to the increased bonding between fillers and base epoxy. In addition, the uniform dispersion could be the reason that the thermal stability of electrospun samples is always better than that of conventional samples.

CONCLUSION

With the addition of aluminum fillers, not only the thermal conductivity of epoxy composites were improved, but also the thermal stability of epoxy composites was enhanced. In addition, electrospun samples always showed better thermal properties than conventional samples, which may be attributed to the well dispersion of nanofillers. The sample filled with the combination of nano and micro fillers showed the best properties than any of samples filled only nano or only micro aluminum fillers. This confirmed the benefit of using the combination of nano and micro fillers in the base polymers.

ACKNOWLEDGMENTS

The authors thank the Natural Sciences and Engineering Research Council of Canada for financial support.

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