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Effects of Kingcure K-11 Hardener and Epoxidized Sunflower Oil on The Properties of Polymer Composite Material Based on Epoxy Resin Gelr 128

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ABSTRACT

Epoxidized sunflower oil (ESO) has been used to toughen epoxy resin GELR 128 cured with an accelerated aliphatic amine curing agent (Kingcure K-11) at room temperature. There was difference in the properties of the polymer composite materials based on epoxy resin GELR 128 cured by Kingcure K-11 between two processes: one-stage process and two-stage process for mixing ESO with epoxy resin GELR 128 at various content of ESO. The results showed that the two-stage process is considered to be more advantageous than the one-stage process. It can be concluded that the impact strength, critical stress intensity factor Kic and decomposition temperature of the polymer composite materials based on epoxy resin GELR 128 cured by Kingcure K-11 with content of ESO 5 phr in two-stage process was greater ones in one-stage process (impact strength: 35.012 kJ/m2, Kic: 2.72 MPa and decomposition temperature: 385.81 0C respectively).

Keywords*—accelerated aliphatic amine curing agent, epoxidized sunflower oil, epoxy resin GELR 128, toughness, two processes, polymer composite materials.*

I. INTRODUCTION

Epoxy resin was used as a glue agent from the mid 20thcentury with special initial properties such as high elasticity, mechanical properties, chemical inertia [1, 2]. However, when epoxy resins are used extensively in other different fields, they are denatured for increased brilliance due to the increased bonding of the polymer molecules [2-4]. This makes epoxy resin easy to peel as well as reduced impact resistance [5]. Nowadays, many research projects is aiming to reduce the brittle strength of epoxy resins, some of them was done by mixing carbon nanotube, oil, liquid rubber particles, or silica particles, ... [2, 6-11]. Furthermore, materials for these methods are non-renewable. Therefore, methods of using renewable natural resources are one of the best choose that attract the attention of many researchers [12-14]. We also used the vegetable epoxide oil as a blend of epoxy resins to increase the durability and strength of epoxy resins without compromising the modulus and thermal properties of the material.

This study investigated the effect of an accelerated aliphatic amine curing agent (Kingcure K-11) and epoxidized sunflower oil (ESO) on the tensile strength, flexural strength, modulus of elasticity, critical stress intensity factor and thermal property of the polymer composite materials based on epoxy resin GELR 128 in two processes: one-stage process and two-stage process at room temperature.

II. EXPERIMENT

A. Material

- Epoxy resin used in this study was DGEBA supplied by Epoxy Base Electronic Material Corporation Limited of China (GELR 128) which had an epoxide equivalent weight (EEW) of 184-190g eq-1 and a viscosity at 250C: 11-15 Pa.s;

- Epoxidized sunflower oil (ESO) was synthesized by using ion exchange method. The content of epoxy group is 16.45% with oxiran value 6.12.

- Kingcure K-11 (KK1) is an accelerated aliphatic amine curing agent in an amber-colored liquid form was purchased from Sanho Chemical Co.,LTD (Taiwan) (density 1.04 g/cm3 at 250C, viscosity at 250C: 1000-2000 mPa.s, amine value: 430±20 mgKOH/g and Active Hydrogen Equivalent Weight (AHEW): 93).

B. Modified epoxy resins

Modified epoxy resins were prepared by mixing DGEBA GELR 128 and epoxidized sunflower oil in two processes:
One-stage process: the various amount of ESO (0 - 30 phr) was added into 100 g of epoxy resin GELR 128 in 250 ml glass beakers, stirring for 45 minutes. Then KK1 was added with a pre-calculated ratio and stirred for 5 minutes. The all components were poured into a mold for curing at room temperature.
Two-stage process: Mixing ESO and KK1 in a 3-necked 250-well flask were stirred for 15 hours, adding epoxy resin GELR 128 and continued stirring for 5 minutes and then poured the mixture into the mold to cure at room temperature.

C. Research methods

- The morphologies at the fracture surfaces of the epoxy samples were evaluated from Scanning Electron Microscopy (SEM) on Hitachi (Japan) S4800 at the main laboratory, Institute of Materials Science - Vietnam Academy of Science and Technology
- Thermal stability was studied by simultaneous thermos gravimetric analyzer (TGA) by SETARAM TG under a heating rate of 100C/min. within a temperature range of ambient room temperature to 8000C at Hanoi University of Sciences, Hanoi National University.
- The tensile strength was determined according to ISO 527-2012 on the INSTRON 5582-100kN (USA) with the crosshead speed of 2mm/min at the Polymer Centre, Hanoi University of Science and Technology.
- The flexural strength was determined according to ISO 178 on the INSTRON 5528-100kN (USA) with the crosshead speed of 2mm/min at the Polymer Centre, Hanoi University of Science and Technology.
- Izod impact strength was determined according to ISO 180 on the Tinius Olsen Model 92T (USA) at the Polymer Centre, Hanoi University of Science and Technology.
- The critical stress intensity factor, KIC value was determined according to ASTM D5045-99 by three point sidedon a Lloyd 500N (UK) with the crosshead speed of 10mm/min at the Polymer Centre, Hanoi University of Science and Technology.

III. RESULTS

A. Morphological structure

Scanning electron microscopy of modified epoxy resin GELR 128 without ESO and with 15, 20 phr ESO are shown in Figure 1. From Figure 1 (a), the unmodified epoxy resin GELR 128 has smooth surface as a mirror with cracks in different surfaces characterize which is the lower impact strengths of thermoplastic resins. The fractured surface of the modified ESO epoxy resin GELR 128 consists of two distinct phases: spherical resins dispersed in a continuous epoxy network. The seeds are about 1-2 µm in diameter. KK1 is a polyamine that interacts with epoxy resin GELR-128 higher than ESO's epoxy group so that it forms a durable grid while epoxy sunflower oil decomposes into second phase as small liquid droplets (Figure 1b). The surface of epoxy resin GELR 128 with added ESO has an uneven roughness profile as unmolded epoxy resin GELR 128, which shows the durability of the material. For the two-stage process the appearance of seeds which show an fluctuate surface on the broken area (Fig 1c)

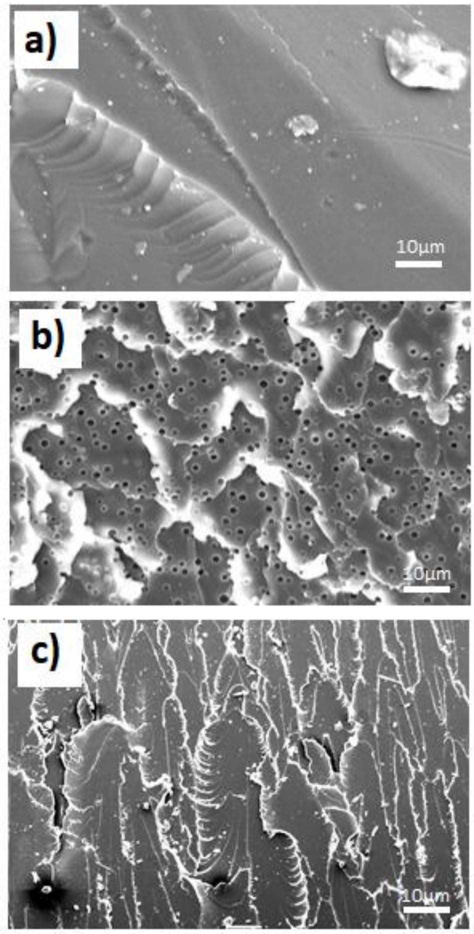

Figure 1SEM images of modified epoxy resin GELR 128 a) without ESO b) one- stage process with 15 phr ESO c) two-stage process with 20 phr ESO

B. Mechanical properties

The results of tensile and flexural properties of epoxy resin GELR 128 were denominated in two processes with and without epoxidized sunflower oil as shown in Table 1. The tensile strength of epoxy resin GELR-128 without ESO is 63.08 MPa with high flexural strength (103.4 MPa) and low modulus (2.94 GPa) which showed the hardness and brilliance of epoxy resin. When ESO is added, tensile strength and flexural strength decrease in proportion to the increase in the amount of ESO in the one-stage process. Phase separation also does not affect the reduction of tensile strength and flexural strength in the two-stage process due to the presence of low modulus phases in the epoxy network. This suggests that epoxy resin GELR 128 with ESO is more resilient than epoxy resin GELR 128 without the addition of a modified substance. In both process, tensile strength decreases gradually as increase the amount of ESO from 10-30 phr. This trend is similar to the flexural strength value, but

this significant decline occurs when ESO content is higher than 5 phr. Comparing with the one-stage process, the mechanical values of the epoxy resin GELR 128 after the denaturation by two-stage process are always lower, proving the superiority of the two-stage process, the material after the modify is more toughness with less amount of denaturation.
The relationship between the amount of ESO content and impact strength is shown in Figure 2. The impact strength of the epoxy resin GELR 128 after denaturation with ESO in two-stage process was significantly higher than one-stage process. However, the impact strength of the epoxy resin GELR 128 was made by one-stage process which is only moderately increased due to single-phase morphology (as SEM image results) which is the same trend in two-stage process. Therefore, the impact resistance value in two-stage process is always higher than one-stage process with all amount of ESO content. The maximum impact strength at ESO content is 5-10 phr and slightly decreases as ESO content increases

Table 1The effect of ESO to the tensile and flexural properties of epoxy resin GELR 128

	ESO (phr)	Tensile strength (MPa)	Elastic modular (GPa)	Flexural strength (MPa)	Flexural modular (GPa)
	0	63.08	1.71	103.4	2.94
One-stage process	5	53.48	1.58	94.5	2.62
	10	44.77	1.34	73.2	2.08
	15	38.25	1.33	67.5	1.86
	20	33.91	1.19	45.1	1.47
	30	33.39	1.06	44.2	1.33
Two-stage process	5	52.17	1.54	71.9	2.12
	10	43.20	1.29	62.3	1.87
	15	36.08	1.14	46.3	1.40
	20	27.19	0.98	39.2	1.27
	30	19.53	0.95	28.5	1.19

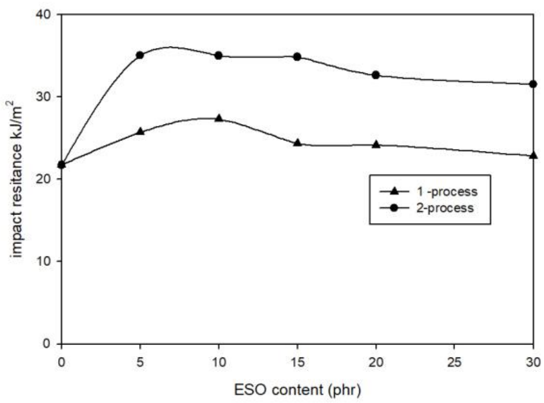

Figure 2: Impact strength of epoxy resin GELR 128 after denaturation at different content of ESO

The change in the durability deterioration was characteristic by the KIC-concentrated stress ratio by ESO content is shown in Figure 3. It can be seen that the addition of ESO to the epoxy resin GELR 128 which is increased the stress concentrate coefficient. From Figure 3, in the two-stage process, the stress ratio was higher than this in the one-stage process when the content of ESO is 5phr and reached the highest value of 2.58 MPa.m1/2 at the ESO level of 15 phr. Then it is tending to change the same strength but the Kic value in two-stage process is lower than Kic value in one-stage process. So the optimum content of ESO is 5phr. This is explained by the fact that the amount of epoxidised sunflower oil increases, the agglomeration of the oil particles occurs and this causes the material to lose its elongation.

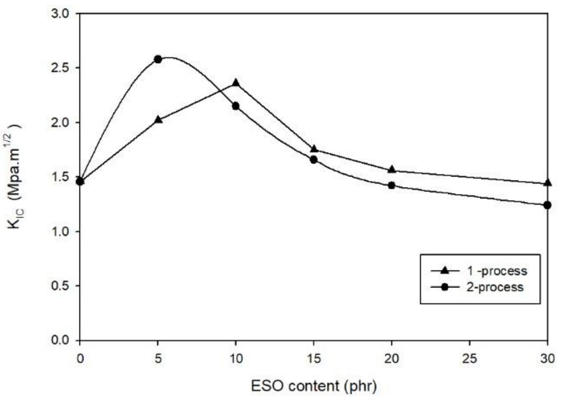

Figure 3Stress concentrate coefficient KIC of epoxy resin GELR 128 after modification at various ESO content

From the previous survey results, epoxy resin GELR 128 with curing by polyamine show the promised properties with the low content of nature oil. Our results show that the content of ESO 5phr was chosen as the suitable ratio to improve the mechanical properties of epoxy resin GELR 128

C. The thermal properties

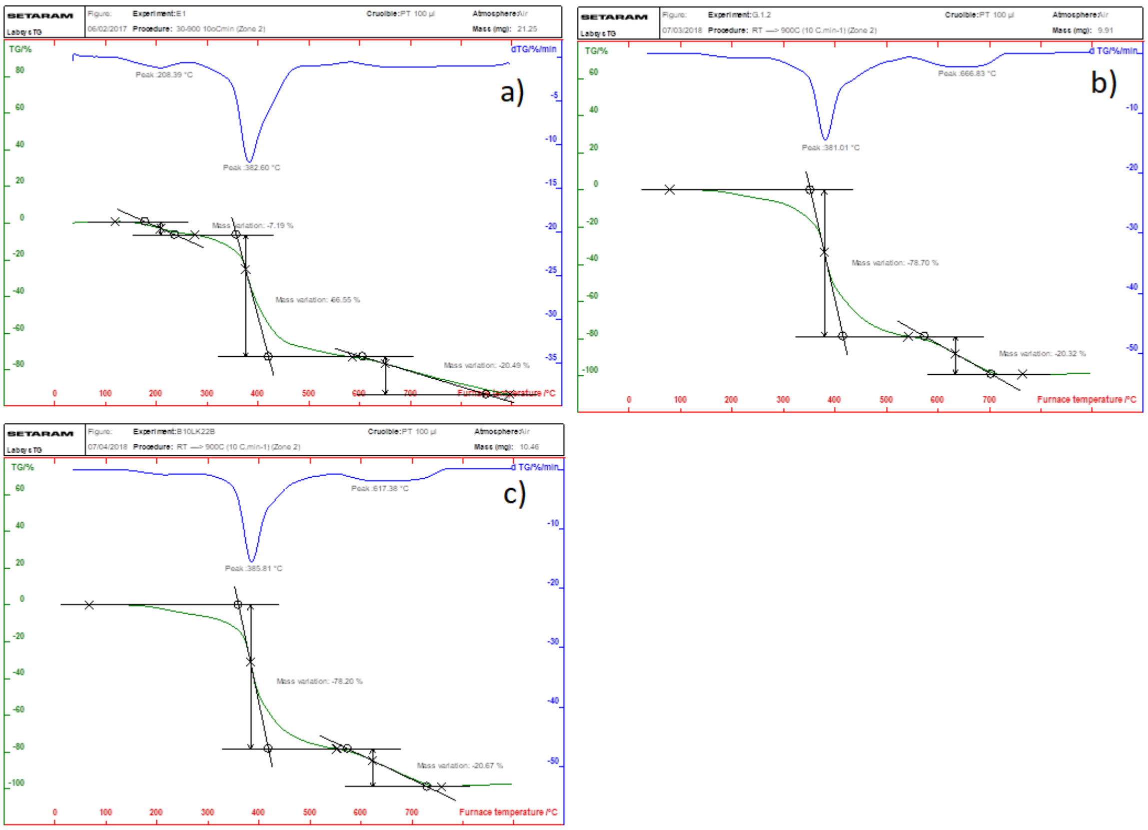

Figure 4The TGA of a) epoxy resin GELR 128, epoxy resin GELR 128 modified with ESO by b) one-stage process, c) two-stage process.

From the results of TGA (table 2 and figure 4), the opposite strength shows which are decrease the maximum decomposition temperature in one-stage process and increase in two-stage process compare with the neat epoxy resin GELR 128 sample. It can conclude that in two-stage process the reaction reach higher yield than in one-stage process In other way, compare with the SEM image (Fig 1), the content of the oil drop in one-stage process made the decompose temperature lower.

Table 2Effect of ESO content on the maximum decomposition (MDT) temperature of epoxy resin GELR 128

Number	ESO content (phr)	MDT, °C
1	0	382.6
2	10 (one-stage process)	381.01
3	5 (two-stage process)	385.81

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IV. Conclusion

The results of the toughen epoxy resin GELR 128 with epoxidized sunflower oil (ESO) cured by an accelerated aliphatic amine curing agent (Kingcure K-11) have shown some promised results. The two-stage process is considered to be more better than one-stage process in tough epoxy resin GELR 128. The impact strength, critical stress intensity factor Kic and decomposition temperature of the polymer composite materials based on epoxy resin GELR 128 cured by Kingcure K-11 with content of ESO 5 phr in two-stage process was greater ones in one-stage process

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References

[1] SOO-JIN PARK, H.-C.K., Thermal Stability and Toughening of Epoxy Resin with Polysulfone Resin. Journal of Polymer Science: Part B: Polymer Physics, vol.39. pp. 121-128, November 2000.

[2] Liu, S., X. Fan, and C. He, Improving the fracture toughness of epoxy with nanosilica-rubber core-shell nanoparticles. Composites Science and Technology, vol 125, pp. 132-140,March 2016.

[3] Benaniba, M.T., N. Belhaneche-Bensemra, and G. Gelbard, Kinetics of tungsten-catalyzed sunflower oil epoxidation studied by1H NMR. European Journal of Lipid Science and Technology, vol 109(2), pp. 1186-1193, December 2007.

[4] Ratna, D., Mechanical properties and morphology of epoxidized soyabean-oil-modified epoxy resin. Polymer international, vol 50(2), pp. 179-184January 2001.

[5] Ratna, D., R. Varley, and G. Simon, Processing and chemorheology of epoxy resins and their blends with dendritic hyperbranched polymers. Journal of applied polymer science, vol 92(3), pp 1604-1610 March 2004.

[6] Banthia2*, D.R.a.A.K., Toughened epoxy adhesive modified with acrylate based liquid rubber. Polymer International, vol. 49, pp. 281-287.February 2000.

[7] BROWN, S.C.K.H.R., Impact-modified epoxy resin with glassy second component. JOURNAL OF MATERIALSSCIENCE, vol. 22, pp 2587-2594.November 1987.

[8] Chen, Y., Z. Xi, and L. Zhao, New bio-based polymeric thermosets synthesized by ring-opening polymerization of epoxidized soybean oil with a green curing agent. European Polymer Journal, vol. 84, pp. 435-447, November 2016.

[9] G. Di Pasquale, O.M.o.t.t.a.a.A.R., J. T. Carter and P. T. McGrail, D. Acierno, New high-performance thermoplastic toughened epoxy thermosets. Polymer, vol. 38(17), pp. 4345-4348. August 1997.

[10] KING-FU LIN, Y.-D.S., Core-Shell Particles Designed for Toughening the Epoxy Resins. II. Core-Shell-Particle-Toughened Epoxy Resins. Journal of Applied Polymer Science, vol. 70, pp. 2313-2322. December 1998.

[11] 11. Yang, J.-P., et al., Superiority of nanosized over microsized hyperbranched polymer second phase in modifying brittle epoxy resin. Journal of Applied Polymer Science, vol. 119(2), pp. 863-870. July 2011.

[12] Zhou, H. and S. Xu, A new method to prepare rubber toughened epoxy with high modulus and high impact strength. Materials Letters, vol. 121, pp 238-240.April 2014.

[13] Zeng, R.-T., et al., Curing behavior of epoxidized soybean oil with biobased dicarboxylic acids. Polymer Testing, vol. 57, pp. 281-287. February 2017.

[14] Tan, S. and W. Chow, Biobased epoxidized vegetable oils and its greener epoxy blends: A review. Polymer-Plastics Technology and Engineering, vol. 49(15), pp 1581-1590. December 2010.