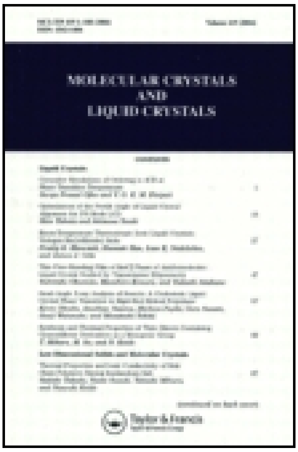

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Preparation and Mechanical Characterization of Tack-free Surfaced CSR/Epoxy Adhesive Films

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High performance epoxy systems have been widely used in electronic devices, aerospace structures, and automotive parts. In this study, introduction of both core-shell rubber (CSR) and thermoplastics into epoxy systems was considered to improve mechanical properties as well as to make tack-free surfaced epoxy films. CSR particles were very uniformly dispersed into epoxy by means of ultrasonic dismembrator and homogenizer at the presence of polymeric dispersion agent. Well-dispersed CSR containing epoxy systems exhibited higher peel strengths at room temperature. Furthermore, peel strength as well as impact peel strength at low temperature could be comprehensively improved.

Keywords tack-free surface; core-shell rubber particles; epoxy; impact peel strength; toughened epoxy

Introduction

High performance epoxy systems have been widely used in electronic devices (electronic packaging materials), aerospace structures (bonding film, carbon fiber reinforced epoxy composite), and automotive parts (structural adhesive) [1,2]. In these applications, epoxy systems are mostly used as a base material and higher bonding strength is needed. Among the possible approaches to improve the mechanical properties of epoxy systems, introduction of core shell rubbers (CSR) to epoxy systems can be considered [3–5]. By introducing liquid rubber and core-shell rubber (CSR) particles, mechanical characteristics such as peel and impact peel strengths can be improved [6,7]. In general, the diameter of CSR particles ranges from 30 to 250 nm. As the diameter of CSR particles is reduced, the thermodynamic and mechanical properties are improved. Furthermore, diallyl phthalate (DAP), multipurpose thermosetting resin for electronic applications, can be another solution to improve the mechanical and water absorption properties [8,9]. DAP possesses a capability of preventing water absorption by networking reaction in epoxy systems.Soluble thermoplastics such as polyamide and polycarbonate can be used to reduce the tackiness of epoxy systems. In this study, introduction of both CSR and thermoplastics into epoxy systems was considered to improve mechanical properties as well as to make tack-free surfaced epoxy films.

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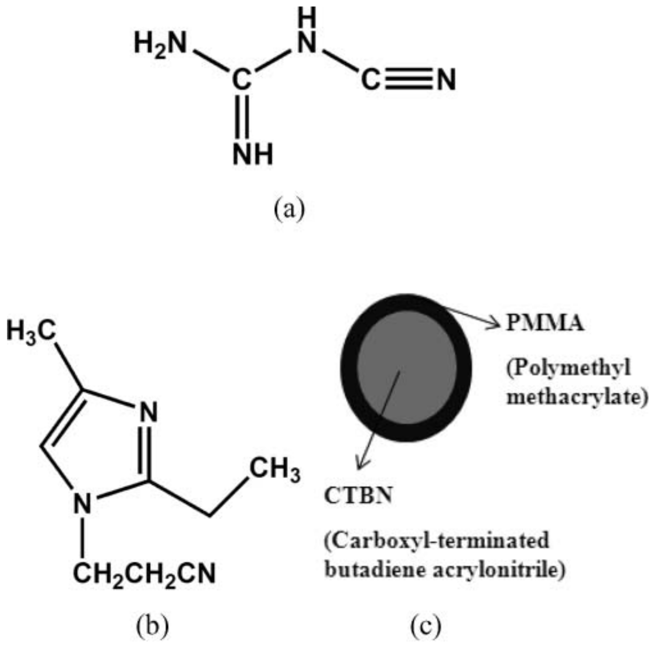


Figure 1. Chemical structures of Dicyandiamide (DICY) (a) and 1-(2-cyanoethyl)-2-ethyl-4 methylimidazole (2E4MZ-CN) (b), and core-shell rubber (CSR) structure.

Experimental

Diglycidyl ether bisphenol-A type (EEW: 184–190, Kukdo Chemical) as an epoxy resin,dicyandiamide (DICY, Hitachi Chemical) as a curing agent, 1-(2-cyanoethl)-2-ethyl-4-methylimidazole (TCI) as an curing accelerator, and 3-glydidyloxypropul trimethoxysilane (ShinEtsu) as a coupling agent are used as received. CSR particles (Rhom & Haas, Diameter:about 150 nm) and diallyl phthalate (DAP, TCI) as modifiers, polyether monoamine (Huntsman) as a dispersion agent were used, and the chemical structures of selected materials are shown in Fig. 1.

Epoxy resin was mixed with curing agent (equivalent ratio of 1:0.85), accelerator (2 phr) and coupling agent (0.5 phr) for 45minutes at 80°C. CSR particles (5, 10 and 15 wt%) with polyether monoamine were dispersed in the prepared epoxy systems using an ultrasonicdismembrator for 1 hr at 80°C. The loading percent of DAP was limited to about 5% in epoxy systems. Reactive polyamide and polycarbonate were added to epoxy systems to make adhesive films.Adhesive films were cured at 160°C for 20 min. T-peel and impact peel strengths were measured to mechanically characterize the prepared tack-free adhesive films. The specimen of impact peel strength test is shown in Fig. 2.

Results and Discussion

It is known that nano-sized CSR particles does not affect on the curing kinetics of epoxy systems [10]. However, if there exist micro-sized domains in epoxy systems, those domains could affect on the curing kinetics. At the presence of micro-sized domains in epoxy systems, in general, cure peak temperatures on the heat flow curves slightly increase since the cure reaction path could be hindered by micro-sized domains [11]. In this reason,

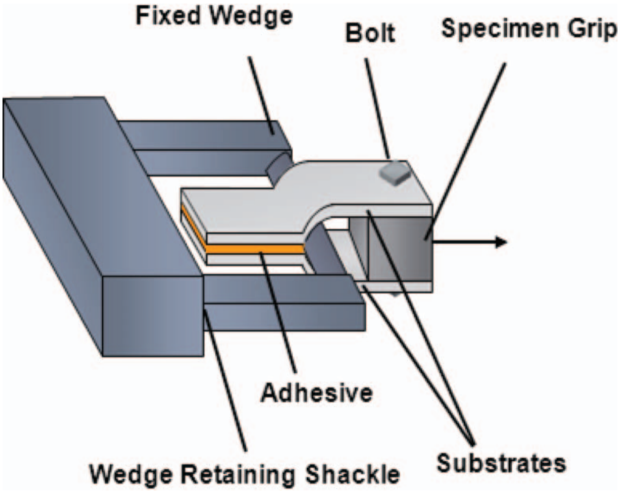
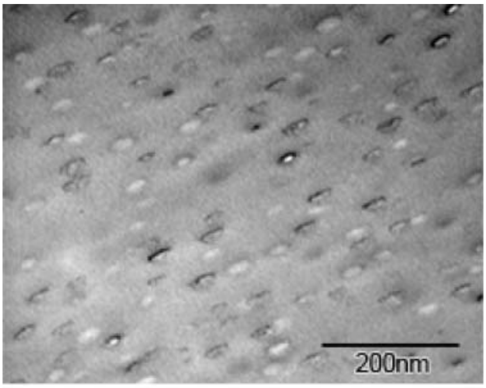
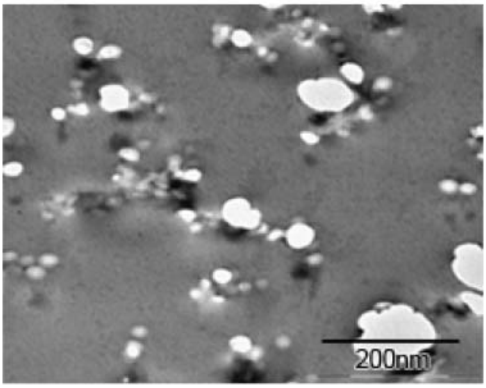


Figure 2. Specimen of impact peel strength test.

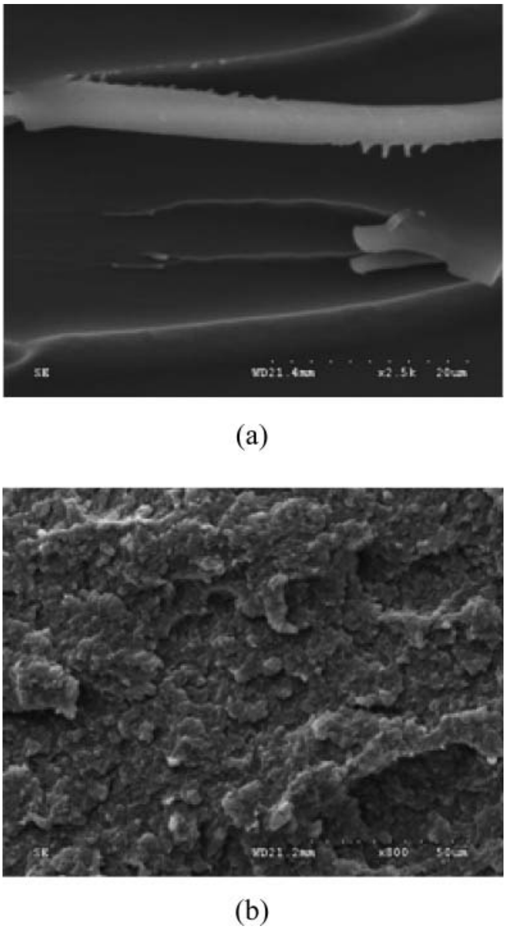


(a)



(b)

Figure 3. TEM images of well-dispersed CSR (dispersed by ultrasonicdismembrator and homogenizer) (a) and poorly-dispersed CSR (dispersed by using solvent or polymeric dispersion agent) (b).



(a)

(b)

Figure 4. SEM images of fractured surface: thermoplastics/epoxy (a) and thermoplastics/CSR/epoxy (b).

the size of selected CSR particles was about 150 nm. CSR particles were dispersed into epoxy in various ways. High frequency generator (ultrasonic dismembrator), homogenizer, polymeric dispersion agent, and solvent have been employed to disperse CSR particles into epoxy systems. Ultrasonic dismembrator and homogenizer were most effective on CSR dispersion at the presence of polymeric dispersion agent, as shown in Fig. 3. Aggregates were found when CSR particles were dispersed in epoxy systems by means of solvent or polymeric dispersion agent. Fractured surface of CSR/epoxy systems, as shown in Fig. 4, indicated that uniformly dispersed CSR particles could block micro crack propagations during fracturing process.

By introducing reactive thermoplastics, tack-free surfaced adhesive films were successfully prepared with a thickness of 5–50 micro meters. During curing process, these reactive thermoplastics are involved in epoxy crosslinking reactions.Well-dispersed CSR particles and reactive thermoplastics tremendously influenced on the peel strength of the prepared epoxy films, as shown in Fig. 5. Ultrasonic dismembrator was less effective because current epoxy systems contain soluble thermoplastics, leading to forming viscous

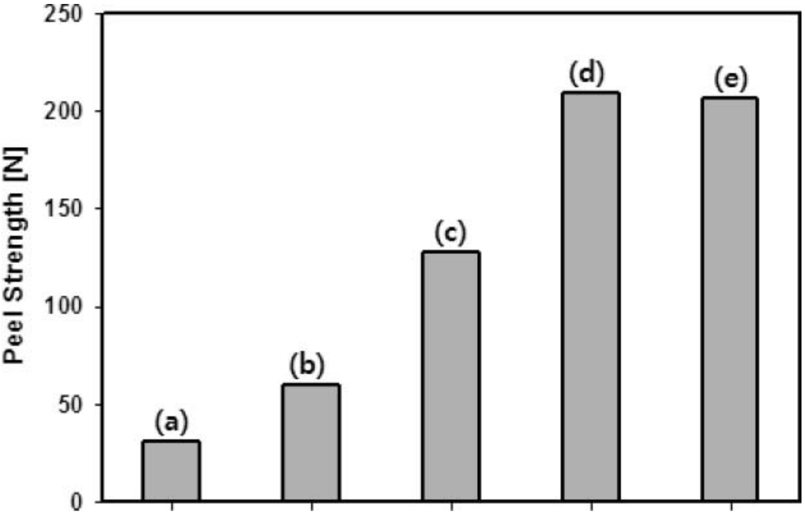


Figure 5. Peel strengths of epoxy systems: neat epoxy (a), thermoplastics/epoxy (b), thermoplastics/CSR(ultrasonic)/epoxy (c), thermoplastics/CSR(homogenizer)/epoxy (d), and thermoplastics/CSR(ultrasonic and homogenizer)/epoxy (e).

fluids. However, ultrasonic dismembrator can be used with homogenizer during the last part of CSR dispersion process in the present epoxy systems. Soluble thermoplastics were also effective to improve peel strength of neat epoxy at room temperature.

Impact peel strength tests were performed at room and low temperatures (23°C and −40°C, respectively) using the prepared epoxy films and test results are shown in Table 1. For neat epoxy, the values of impact peel strength were zero at both room and low temperatures. At the presence of thermoplastics, low value of impact peel strength was measured at room temperature and at low temperature, however, the value approached to zero. At 15% content of CSR in epoxy systems, higher values of impact peel strength were measured at room and low temperatures. As thermoplastics are incorporated into epoxy systems, the values of impact peel strength at room and low temperatures were significantly improved. It is recommended that both soluble thermoplastics and CSR particles should be simultaneously used to improve impact resistance properties especially at low temperature conditions.

Table 1. The values of impact peel strength measured at room and low temperatures

	Neat epoxy	Epoxy/ thermoplastics (10 wt%)	Epoxy/CSR (15 wt%)	Epoxy/thermoplastics (10 wt%)/CSR (15 wt%)
Impact Peel[N/mm] at 23°C	0.0	2.3	25.0	37.5
Impact Peel[N/mm] at −40°C	0.0	0.0	13.4	21.6

Conclusion

CSR particles and soluble thermoplastics were incorporated into epoxy systems to improve impact resistance properties and to form tack-free surfaced adhesive films. CSR particles were uniformly dispersed into epoxy by means of ultrasonic dismembrator and homogenizer at the presence of polymeric dispersion agent. Consequently, well-dispersed CSR containing epoxy systems exhibited higher T-peel and impact peel strengths. Prepared tack-free surfaced adhesive films can be applied to primary automotive structures in which higher peel strength as well as impact peel strength, especially at low temperature conditions, are exclusively required.

Acknowledgment

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References

[1] Gilbert, E. N., Hayes, B. S., & Seferis, J. C. (2003). *Compos. Part A.*, *34*, 245.
[2] Kang, S., Hong, S., Choe, C. R., Park, M., Rim, S., & Kim, J. (2001). *Polymer.*, *42*, 879.
[3] Nguyen, F. N., & Berg, J. C. (2008). *Compos. Part A-Appl. S.*, *39*, 1007.
[4] Gietl, T., Lengsfeld, H., & Altstadt, V. (2006). *J. Mater. Sci.*, *41*, 8226.
[5] Roberts, K. N., Simon, G. P., Cook, W. D.,& Burchill, P. J. (2000). *J Polym. Sci. Pol. Phys.*, *38*, 3136.
[6] Sue, H. J. (1992). *J. Mater. Sci.*, *27*, 3098.
[7] Becu, L., Maazouz, A., Sutereau, H., & Gerard, J. F. (1997). *J. Appl. Polym. Sci.*, *65*, 2419.
[8] He, S., Liang, G., Yan, H., Wang, J., & Yang, L. (2009). *Polym.Adv.Technol.*, *20*, 143.
[9] Shenoy, M. A., Patil, M., & Shetty, A. (2007). *Poly. Eng. Sci.*, *47*, 1881.
[10] Jo, Y. R., & Choe, Y. S. (2011). *Mol. Cryst. Liq. Cryst.*, *539*, 190.
[11] Song, H. W., Kang, A. S., Kim, W. H., Stephan, M., Kim, B. M., & Choe, Y. S. (2009). *Polymer (Korea).*, *33*, 290.