

Synthesis, Characterization, and Thermo Mechanical Properties of Siloxane-Modified Epoxy-Based Nano Composite

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ABSTRACT: In this study, polymer hybrid composites were synthesized by sol-gel process. 3-Amino-propyltrimethoxysilane [APTMS]/ γ -Glycidoxypropyl trimethoxy-silane (GPTMS); (4, 4'-Methylene-dianiline (DDM)] and 1,4-Bis(trimethoxysilylethyl) benzene (BTB) were added to DGEBA type epoxy resin for anticipated to exhibit excellent thermal stability. Boron trifluoride monoethylamine (BF₃MEA) was used as catalyst. The structure of nanocomposites was characterized by attenuated total reflectance (ATR) and solid-state ²⁹Si NMR which suggest EP-APTMS-BTB/EP-GPTMS-BTB possesses T³; T¹-T⁰, and T¹ structures when the BTB content was lower than 10 wt % and higher 20 wt %, respectively. BF₃MEA was proved to be an effective catalyst for the sol-gel reaction of APTMS, but it could not promote for GPTMS. From TEM microphotographs, EP-APTMS-BTB (10 wt %) possesses a dense inorganic structure (particle size around 5–15 nm) compare with the loose inorganic structure of EP-GPTM-/BTB (10 wt %). DSC, TGA were use to analyze the thermal properties of the nanocomposites and DMA was used to analyze the dynamic mechanical properties of hybrid composites. The T_gs of all nanocomposites decreased with the increasing BTB content. A system with BTB content lower than 10 wt % showed good dynamic mechanical property and thermal stability (Td₅ increased from 336°C to 371°C, char yield increased from 27.4 to 30.2%). The structure of inorganic network affects the Td₅ and dynamic mechanical properties of composite.

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KEYWORDS: composites; resins; thermal properties

Received 7 February 2014; accepted 7 May 2014

DOI: 10.1002/app.40984

INTRODUCTION

Nanopolymer composites, characterized by an ultrafine grain size, functional group effects, have stimulated much research interest by virtue of their unusual mechanical, electrical, optical, thermal, and magnetic properties.^{1–4} Epoxy resins are widely used as coating, adhesive, and laminate in the chemical and electrical industries^{5,6} due to their excellent chemical resistance, higher adhesive strength, good electrical properties, and ease processing.^{7–9} Many investigations have been reported for the past decades in the development of the high-performance organic/inorganic nanocomposites. From an industrial approach, owing to high costs of development, synthesis and commercialization of new polymers, most researchers look for new materials by bonding or functionalising polymers, so the tailor made properties of the materials can be achieved.^{10–13}

The increasing demands of the microelectronics and aerospace industry for new high-performance composite materials, many researchers have taken extensive efforts to explore new hybrid resins with high end-use temperatures.¹⁴ The unmodified epoxy

resins have limited application due to their inherent brittleness and relatively poor thermal stability, compared to the hybrid composite.¹⁵ The physical mixing of epoxy resins with polyimides is the most common approach used to prepare the polyimide-epoxy composites, however, this method suffers some drawbacks due to the problems of phase separation and lower thermal stability. This drawback solved by changing the monomer functionality or silanisation of the epoxy resin.

Silicone molecule usually used for the design of organic/inorganic hybrid. There are several synthetic routes to prepare epoxy/silica nanocomposites, sol-gel method is the most convenient and cost-effective method to prepare nanocomposites.¹⁶ Lee et al.¹⁷ reported that the epoxy/nanosilica hybrid materials could be prepared using bisphenol-A type epoxy resin and tetraethoxysilane (TEOS) as organic and inorganic sources, via sol-gel reaction of tetraethoxysilane. The epoxy-based hybrid materials exhibited good thermal stability. Ochi et al.¹⁸ demonstrated that the synthesis of epoxy-grafted silicone oligomer and the materials could be prepared using bisphenol-A type epoxy resin and epoxy-grafted silicon oligomer

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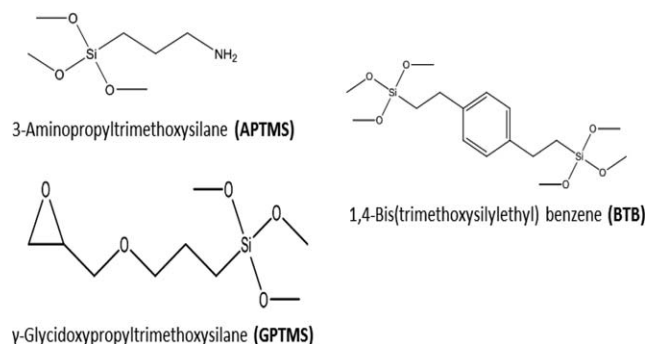


Figure 1. Structures of Silane coupling agents.

(ESO) as organic and inorganic source, although the mechanism and enhancing factor to enhance properties still focusing area of study for academia researchers. The epoxy/silica nano-hybrid materials exhibit good thermal stability and mechanical properties.^{2,4}

In this study, novel epoxy/polyorganosiloxane nanocomposites have been prepared, and the preparation of polyorganosiloxane in epoxy resin via non-aqueous sol-gel process was conducted. A series of Epoxy/BTB hybrid materials have been synthesized from 3-aminopropyltrimethoxysilane (APTMS) or γ -Glycidoxypropyltrimethoxysilane (GPTMS) and various percentages of (5–30 wt %) 1,4-Bis(trimethoxysilyl)ethyl Benzene (BTB) with the catalyst of Boron trifluoride monoethylamine (BF_3MEA) via the sol-gel reaction. BF_3MEA was used as a catalyst to catalyze sol-gel reaction. By adding the amino-polyorganosiloxane or epoxy-polyorganosiloxane to DGEBA type epoxy resin and 4, 4'-methylene-dianiline (DDM), respectively, EP-APTMS-BTB and EP-GPTMS-BTB hybrid materials have been prepared. ATR, ^{29}Si NMR, SEM, and TEM were used to characterize and observe the functional groups and the structure of inorganic structure of polyorganosiloxane. Thermal properties of EP-APTMS-BTB and EP-GPTMS-BTB hybrid materials were investigated by differential scanning calorimetry (DSC) and thermogravimetric analysis (TGA). The glass transition temperatures were analyzed by DSC. TGA was used to analyze the thermal stability. Dynamic mechanical analysis (DMA) was used to analyze the dynamic mechanical properties.

EXPERIMENTAL

Materials

Diglycidyl ether of bisphenol-A/DGEBA epoxy with an epoxide equivalent weight of 190 g/eq (Nan Ya Plastic, Taiwan) was obtained as commercial grade. 3-Aminopropyltrimethoxysilane/APTMS (Dow Corning, Midland, Michigan, USA), γ -Glycidoxypropyltrimethoxysilane/GPTMS (Acros, Janssens Pharmaceutica-laan, 3A 2440 Geel, Belgium), 1,4-Bis (trimethoxysilyl)ethyl Benzene/BTB (United Chemical Technologies, Bristol, PA, USA); which were used as a silane coupling agent. Boron trifluoride/ BF_3 MEA (TCI, Tokyo Chemical Industry, Tokyo, Japan) was used as a catalyst in the sol-gel reaction and curing reaction.

Preparation of Epoxy-APTMS-BTB and Epoxy-GPTMS-BTB Hybrid Materials

The Epoxy-APTMS-BTB (EP-APTMS-BTB) and Epoxy-GPTMS-BTB (EP-GPTMS-BTB) hybrid preparations were

obtained by mixing APTMS/GPTMS and BTB (5, 10, 20, 30 wt %) with stoichiometric ratios of BF_3MEA to achieve APTMS-BTB- BF_3MEA /GPTMS-BTB- BF_3MEA . Based on the experiments, the ratio of EP-APTMS-BTB- BF_3MEA is 1 : 0.3 : (0.05–0.3) : 0.05 and EP-GPTMS-BTB- BF_3MEA is 1 : 0.3 : (0.05–0.3) : 0.05. The chemical structures of silane agents are illustrated in Figure 1. Mixture stirred at room temperature (30°C) for 2 h, then a stoichiometric amount of DGEBA/DGEBA-DDM was added to APTMS-BTB/GPTMS-BTB solution, respectively, and stirred for 2 h to achieve uniform mixing. The Epoxy-APTMS-BTB solution and Epoxy-GPTMS-BTB solutions were poured into an aluminum pan, and cured at 180°C/12 h to fabricate EP-APTMS-BTB hybrid materials, while at 180°C/12 h to fabricate EP-GPTMS-BTB hybrid materials, respectively.

Instrumentation and Techniques

The FT-IR study was carried out using a FT-IR spectrophotometer (Perkin Elmer Spectrum one FTIR), On an attenuated total reflectance (ATR) in the range 4000–400 cm^{-1} with a resolution of 4 cm^{-1} to characterize the chemical structure of EP-APTMS and EP-GPTMS hybrid materials. The High-resolution solid-state ^{29}Si NMR experiment was conducted for characterizing the structure of the siloxane networks on a BRUKER DSX 400 MHz solid-state NMR. The scanning electron microscope (SEM; Hitachi S-4700 Type II, Japan) was used to analyze the morphology of fracture surfaces. The morphological properties of the nano siloxane structure in EP/APTMS/BTB and EP/GPTMS/BTB hybrid materials were analyzed by transmission electron microscopy (TEM, JEOL JEM-1230 at 100 kV voltage). The glass transition temperatures (T_g) of the hybrid materials were determined using a differential scanning calorimeter (DSC; TA instruments Q10, USA) under nitrogen with a heating rate of 20°C/min. The thermal decomposition of the hybrid materials was determined using a thermogravimetric analyzer (TGA; Perkin-Elmer 2400, USA) under nitrogen with a heating rate of 10°C/min to 800°C, to investigate the thermal decomposition temperatures and char yields. The Dynamic mechanical properties of hybrid materials were studied by a dynamic mechanical analyzer (DMA, TA instruments 2940, USA) in the range 35–210°C under nitrogen flow with a heating rate of 10°C/min to observe the storage modulus and $\tan\delta$.

RESULTS AND DISCUSSION

Characterization of Epoxy-APTMS-BTB and Epoxy-GPTMS-BTB Hybrid Materials

EP-APTMS-BTB/EP-GPTMS-DDM-BTB hybrid materials were prepared by mixing the component materials at room temperature and curing them by process of sol-gel. The mixing of the Epoxy and APTMS/GPTMS were characterized by the ATR spectra of EP-APTMS and EP-GPTMS-DDM are shown in Figure 2. The ATR spectra of EP-APTMS and EP-GPTMS-DDM displayed bands at 2972, 2922 and 2876 cm^{-1} in the –CH stretching vibration region as symmetric; symmetric and asymmetric stretching, respectively.¹⁹ A band at 1507 cm^{-1} and 1459 cm^{-1} were attributed to Ar (aromatic group) stretching vibration.⁶ Bands at 1231 and 1178 cm^{-1} were derived from Si-CH₂ stretching vibrations and C–N stretches vibrations, respectively.¹⁰ In the ATR-FTIR spectra, the alkoxy groups via sol-gel

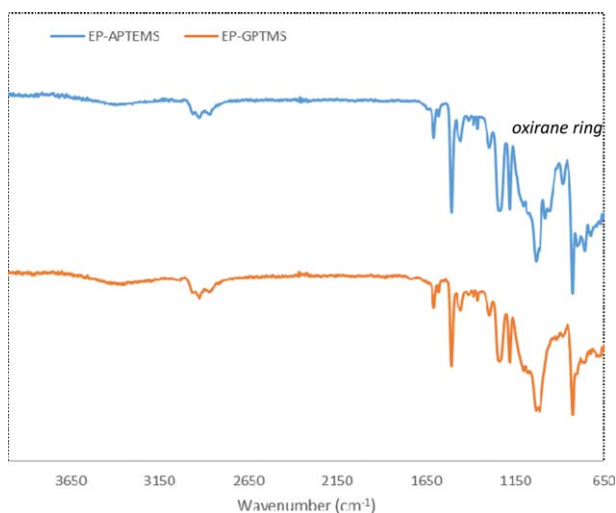


Figure 2. FTIR-ATR spectrum of (a) EP-APTMS and (b) EP-GPTMS hybrid materials. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

reaction demonstrated the presence of absorption peaks of Si–O–Si asymmetric and symmetric stretching at 1074 and 826 cm^{-1} . The absorption peaks of oxirane ring 913 cm^{-1} disappeared after curing process this is because of epoxy resin overlaps the ring.³ From this result, the presence of the APTMS and GPTMS on EP is going to be finalized, which is chemically bonded by polymer rather than physically present.

The structures of EP-APTMS-BTB and EP-GPTMS-BTB hybrid materials are characterized by ^{29}Si solid-state NMR. The Si NMR of BTB mixed with different (0, 5, 10, 20, 30 wt %) ratio to a hybrid of EP-APTMS and EP-GPTMS systems are shown in Figure 3(a,b). When the BTB content was less than 10 wt %, there were two peaks observed at -64 and -52 ppm which were attributed to trisubstituted group T^3 signal and trisubstituted group T^2 signal, respectively, shown in Figure 3(a). The results of solid-state ^{29}Si NMR indicate many silane alkoxy groups of APTMS and BTB are proceeding to the hydrolysis and condensation reactions, and some alkoxy groups which are located in the terminal of the network siloxane structure did not condense to form T^3 structures.⁷ When BTB content was higher than 20 wt %, there were three peaks appeared at -67 , -59 , and -50 ppm. The peak at -50 ppm was attributed to T^1 signal which is indicating that partial silanol or methoxyl groups of BTB and APTMS did not condense to form T^3 structure. This result suggested that the content of BTB increased, if one alkoxy group of a side of BTB reacting with another alkoxy group, it may interfere other two alkoxy groups of BTB to proceed condensation.^{3,6} In Figure 3(b), there are four peaks appeared at -67 , -59 , -49 , and -45 ppm which were corresponded to the T^3 , T^2 , T^1 , and T^0 , respectively. The results of solid-state ^{29}Si NMR were indicating that many alkoxy groups of GPTMS and BTB did not proceed to the condensation reaction. In other words, alkoxy groups of GPTMS and BTB may not be catalyzed by the addition of BF_3MEA as catalyst at room temperature. In the curing process, epoxide groups of GPTMS and epoxy resin would react with amine groups of DDM very

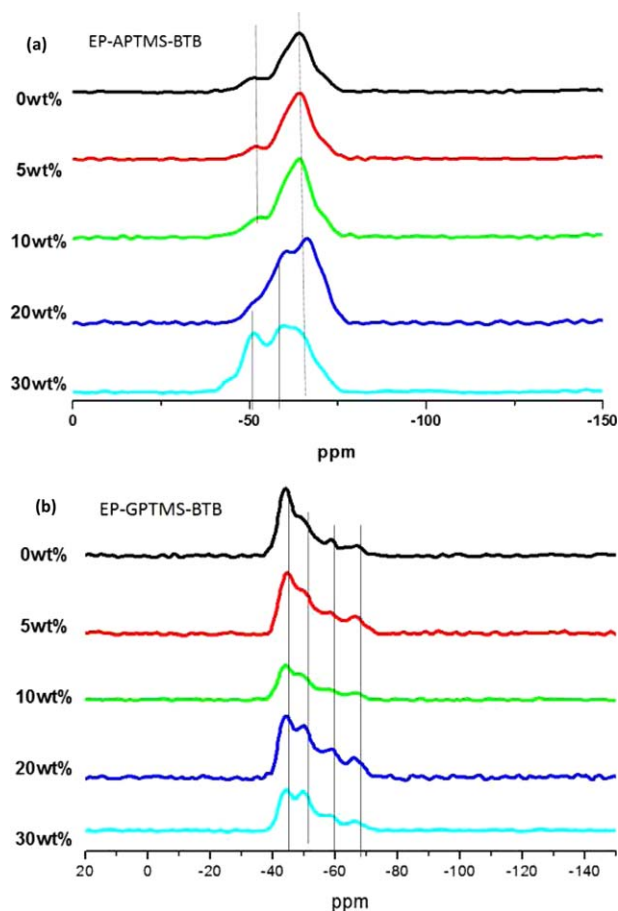
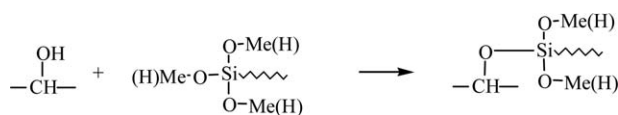


Figure 3. Solid-State ^{29}Si NMR spectra of (a) EP-APTMS-BTB and (b) EP-GPTMS-BTB hybrid system. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

rapidly to form a C–N covalent bond, and it may interfere alkoxy groups to generate Si–O–Si bond.^{6,11} The mechanism of possible bonding of BTB with APTMS and GPTMS are described in Scheme 1.

Scanning electron microscopy (SEM) and transmission electron microscopy (TEM) have been utilized to investigate the morphologies of the hybrid materials. Figure 4 is representing SEM image of EP-APTMS-BTB and EP-GPTMS-BTB materials. The hybrid composites exhibited a featureless morphology, and no discernable phase separation was observed after mixing. The fact that no dispersed materials were identified after the samples were etched with EP-APTMS. The nano APTMS/GPTMS siloxane domains are well dispersed in epoxy bridged siloxanes matrix. The cured network of epoxy-bridged siloxanes exhibits very uniform organic-inorganic hybrid structure. The uniform distribution is enhancing factor for mechanical properties of



Scheme 1. Schematic representation of possible bonding in BTB with APTMS and GPTMS based on NMR results.

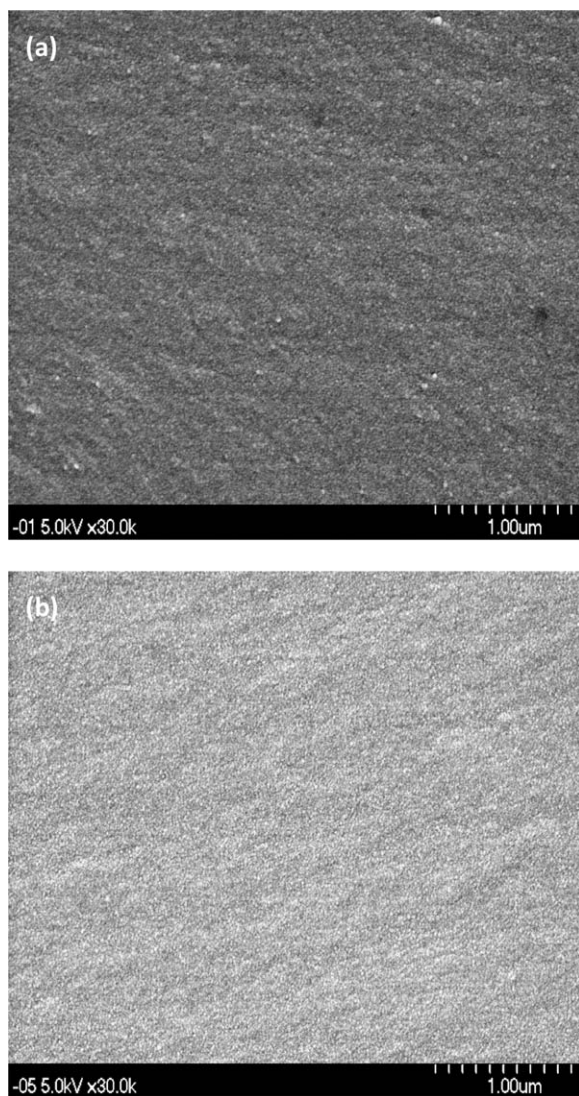


Figure 4. SEM micrographs of (a) EP-APTMS-BTB and (b) EP-GPTMS-BTB systems.

composite, which give uniform results and higher strength to the composite.²⁰ TEM microphotographs are shown in Figure 5. It can be seen that EP-APTMS-BTB 10 wt % (a) have clear dark points due to the dense siloxane domains around 5–15 nm. Siloxane domains are well distributed in the epoxy matrix. However, it can be seen that EP-GPTMS-BTB 10 wt % (b) have no clear dark points due to the loose siloxane domains. It is because alkoxy groups of GPTMS and BTB did not proceed to the condensation reaction by the addition of BF_3MEA at room temperature. Moreover, the hydroxy groups formed by curing reaction of epoxide groups can react with methoxy or hydroxyl groups of GPTMS or BTB through ether exchange reaction to form Si–O–C bonding during the thermal curing process as shown in Scheme 2. This indicated that Si–O–C bondings can improve the interaction between the organic and inorganic phases significantly could affect microstructures of EP-GPTMS-BTB system, which causes the materials possess good compatibility.³

Thermal Properties of Epoxy-APTMS-BTB and Epoxy-GPTMS-BTB Hybrid Materials

The DSC analysis is utilized to investigate the glass transition temperatures of hybrid materials. In Table I, when the BTB content was lowered than 10 wt % in EP-APTMS-BTB system, the T_g decreased from 167°C to 132°C with the increasing of BTB content. From the solid-state ^{29}Si NMR spectra, the hybrid materials have a complete inorganic network when BTB content was lowered than 10 wt %, and they possess uniform crosslinking density, it could restrict the vibrations of polymer chains.¹⁰ Moreover, the degree of crosslinking density decreased and free volume of polymer chains increased with the BTB content, which may cause decreasing of T_g . When the BTB content is higher than 20 wt %, the T_g could not be observed in DSC curves. From the Si NMR spectra, T^1 structures appeared when BTB content is higher than 20 wt %, and they possess disorderly crosslinking density, it could not restrict the vibrations of polymer chains effectively.⁶ In Table I, the EP-GPTMS hybrid materials showed T_g of 168°C. From the Si NMR spectra, the EP-GPTMS-BTB system possesses more T^0 and T^1 structures.

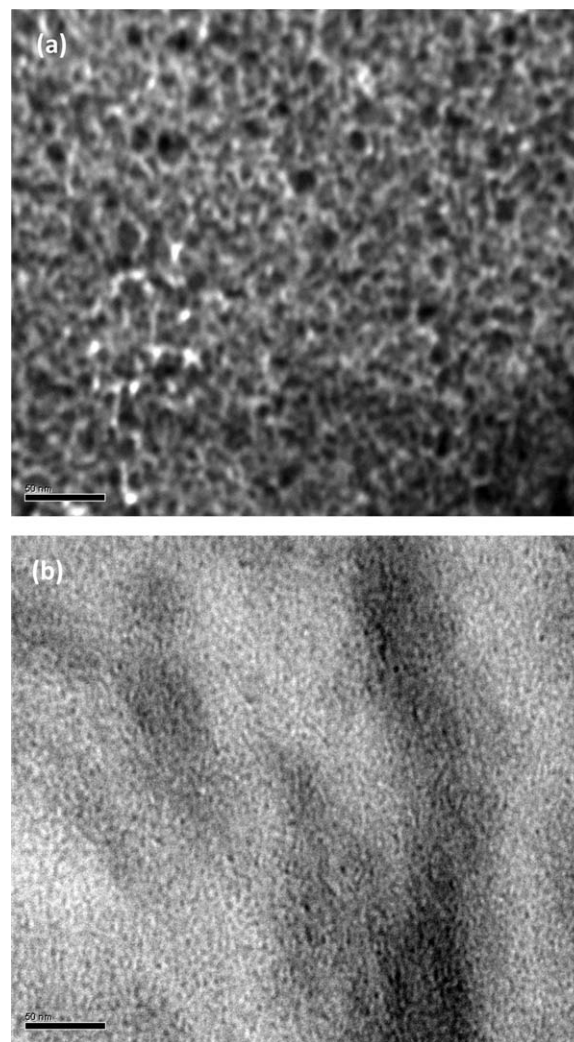
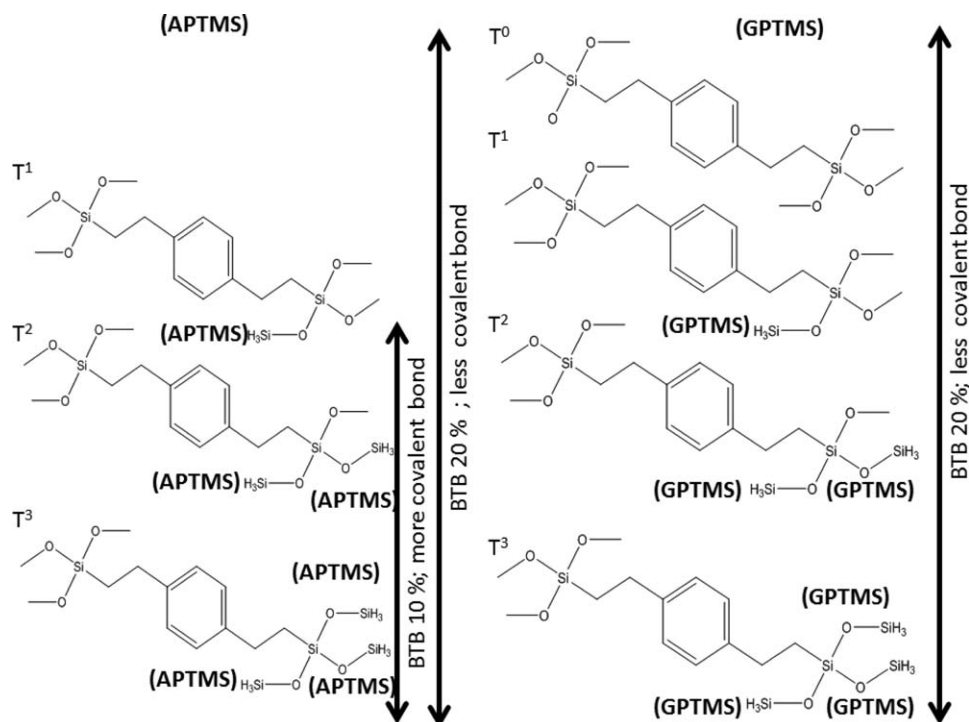


Figure 5. TEM micrographs of (a) EP-APTMS-BTB and (b) EP-GPTMS-BTB systems.



Scheme 2. Scheme of ether exchange reaction from the hydroxy groups reacted with methoxy or hydroxyl groups.

Incomplete inorganic network structures could not restrict the vibrations of polymer chains.³ Moreover, curing reaction proceed very rapidly to form a C–N covalent bond were restrict the vibrations of polymer chains effectively, which caused the T_g reached 168°C. By adding BTB, it would to interfere curing reaction, the crosslinking density became disorderly, which caused T_g could not observe in DSC curves.

TGA analysis is used to investigate the thermal decomposition behavior of materials. T_d . The T_{d5} and T_{d10} represent the thermal degradation temperature of the residue at 5 wt % and 10 wt % loss of the initial weight. In Table II, when EP-APTMS-BTB system with BTB content lower than 10 wt %, the T_{d5} increases from 336°C to 371°C with the increasing BTB content,

because the hybrid materials have complete inorganic network structures to protect polymer chains.³ When EP-APTMS-BTB system with BTB content is higher than 20 wt %, the T_{d5} decreases to 350°C. Since the nanocomposites possess T^1 structure in inorganic networks, some methoxy or hydroxy groups would be degraded earlier. Moreover, the crosslinking density was not uniform, the lower degree of crosslinking density of polymer chains would be degraded earlier, which causes T_{d5} decrease. The trend of T_{d5} and T_{d10} are similar. In Table II, the T_{d5} was in the 377°C–386°C temperature range due to similar inorganic structural distribution of EP-GPTMS-BTB system. The char yield at 800°C of EP-APTMS-BTB and EP-GPTMS-BTB systems increase with the increasing BTB content, because

Table I. T_g values of EP-APTMS-BTB and EP-GPTMS-BTB System with Various BTB Monomer Contents

Sample code	T_g (°C)
EP-APTMS-BTB 0 wt %	167
BTB 5 wt %	141
BTB 10 wt %	132
BTB 20 wt %	*a
BTB 30 wt %	*a
EP-GPTMS-BTB 0 wt %	168
BTB 5 wt %	*a
BTB 10 wt %	*a
BTB 20 wt %	*a
BTB 30 wt %	*a

*a Cannot be detected.

Table II. Thermal Decomposition Characteristics of EP-APTMS-BTB and EP-GPTMS-BTB System with Various BTB Monomer Contents

Sample code	T_{d5} (°C)	T_{d10} (°C)	Char yield (%)
EP-APTMS-BTB 0 wt %	336	374	27.4
BTB 5 wt %	355	390	28.1
BTB 10 wt %	371	397	30.2
BTB 20 wt %	366	397	34.5
BTB 30 wt %	350	380	37.8
EP-GPTMS-BTB 0 wt %	377	411	30.6
BTB 5 wt %	386	415	30.6
BTB 10 wt %	380	414	31.7
BTB 20 wt %	379	415	34.0
BTB 30 wt %	379	421	36.1

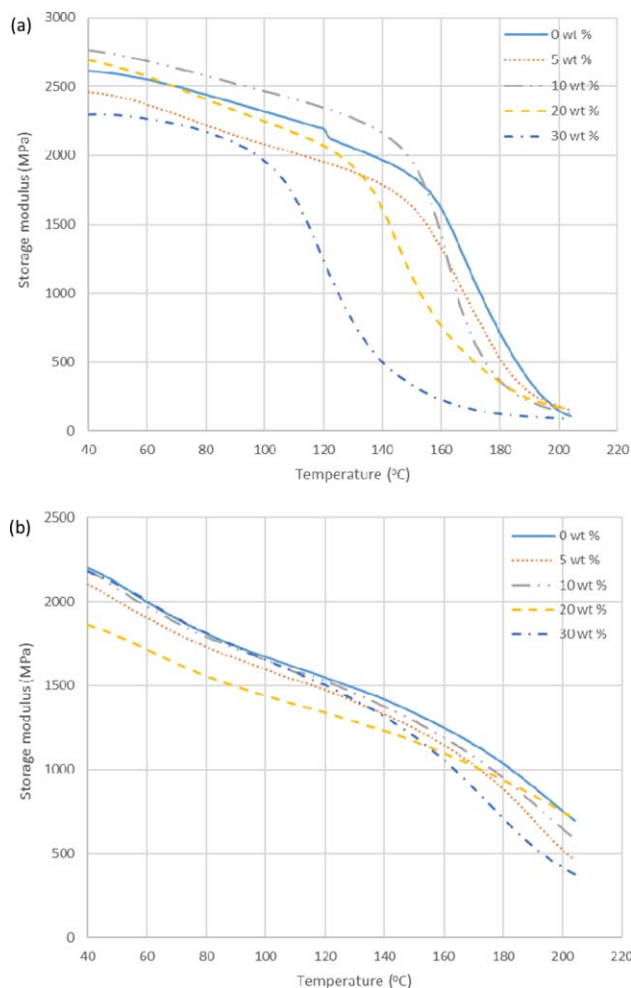


Figure 6. Storage modulus of (a) EP-APTMS-BTB and (b) EP-GPTMS-BTB with various BTB contents. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

the charge could isolate the thermal source, and it decrease the heat transfer rate and reduce combustible gas production, which provide good thermal stability and flame retardants.^{3,4}

The changes of storage modulus and $\tan\delta$ of the EP-APTMS-BTB system and EP-GPTMS-BTB system are illustrated in Figures 6(a,b) and 7(a,b). In Figure 6(a), all hybrid materials of EP-APTMS-BTB system possesses a clear glass-transition region. Glass transition as obtained by DSC is calculated from the absorbed heat or heat change. However, the value obtained by DMA takes into account the change in modulus due to the effect of temperature on the flexibility of the monomer chain. In such case, DMA gives the standard t_g while the results from DSC are affected by the hybrid material. When the BTB content is lower than 10 wt % in EP-APTMS-BTB system, the storage modulus clearly decreased below 140°C. When the BTB content is higher than 20 wt %, the storage modulus clearly decreased below 110°C. This result indicated when the BTB content is lower than 10 wt % that in the hybrid materials possess better dynamic mechanical property than BTB content higher than 20 wt %. It is because the hybrid materials when the BTB content is lower than 10 wt % possess a complete inorganic network

structure to provide good heat resistance. In Figure 6(b), the storage modulus decreased continuously with the temperature raise in EP-GPTMS-BTB system, and did not possess clear glass-transition region. This result indicated that this system possesses low dynamical mechanical property that is because the hybrid materials possess an incomplete inorganic network structure.³ In Table III represent data from Figure 7, the peak of $\tan\delta$ distinctly decreased with the increasing of BTB content in EP-APTMS-BTB system, because the degree of crosslinking density decreased, and free volume of polymer chains increased with the increasing BTB content, which may cause decreased of T_g . The peak of $\tan\delta$ slightly decreased with the increasing of BTB content in EP-GPTMS-BTB system [See in Figure 7(b)] due to similar slightly decreasing of crosslinking density, which may cause slight decreasing of T_g . DSC results support the T_g data obtained by DMA. Compared to DSC, heat change measured by DMA is less drastic because only the change in modulus against temperature is taken into account. Since this is a bulk phenomenon, the distribution is not uniform. In DSC measurements, if the distribution is non-uniform, the difference in heat loss changes drastically and provides an idea about the endothermic and exothermic reaction at each point. DSC data supports the DMA data and additionally, the NMR results.

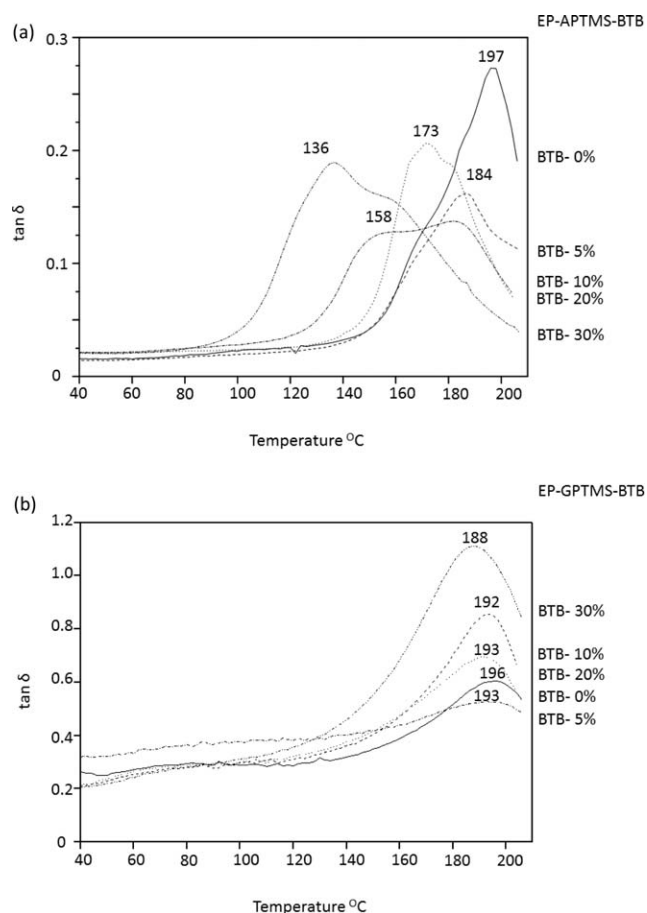


Figure 7. $\tan\delta$ of (a) EP-APTMS-BTB and (b) EP-GPTMS-BTB system with various BTB monomer contents.

Table III. $\tan \delta$ of EP-APTMS-BTB and EP-GPTMS-BTB System with Various BTB Monomer Contents

Sample code	T_g (°C)
EP-APTMS-BTB 0 wt %	197
BTB 5 wt %	184
BTB 10 wt %	173
BTB 20 wt %	158
BTB 30 wt %	136
EP-GPTMS-BTB 0 wt %	196
BTB 5 wt %	193
BTB 10 wt %	193
BTB 20 wt %	192
BTB 30 wt %	188

CONCLUSION

EP-APTMS-BTB and EP-GPTMS-BTB hybrid materials have been successfully prepared from epoxy resin. Tri-functional alkoxy silane was used as inorganic source via a sol-gel process. ATR-FTIR analysis results indicated that oxirane rings disappeared due to curing reaction, alkoxy groups decreased and Si–O–Si appeared due to sol-gel reaction in thermal curing process. Solid-state ^{29}Si NMR spectra indicated that the EP-APTMS-BTB system possesses more T^3 structures when the BTB content was lower than 10 wt %. T^1 structures appeared when BTB content was higher than 20 wt %. The EP-GPTMS-BTB system possesses more T^0 and T^1 structures. BF_3MEA was proved to be an effective catalyst for sol-gel reaction of APTMS in EP-APTMS-BTB system, but it can not promote the sol-gel reaction of GPTMS in EP-GPTMS-BTB system. TEM images that EP-APTMS-BTB hybrid materials exhibit dense siloxane domains around 5–15 nm. However, there is no phase separation in EP-GPTMS-BTB hybrid materials due to the Si–O–C bonds which can increase organic and inorganic phase interaction.

DSC analysis results indicated that the T_g decreased when BTB content is lower than 10 wt % in EP-APTMS system. However, EP-GPTMS-BTB system T_g cannot be found except EP-GPTMS in DSC curves. Thermal analysis confirms all hybrid materials possess good thermal stability. The hybrid materials possess good thermal mechanical properties when BTB content is lower than 10 wt % in EP-APTMS-BTB system. The T structure of inorganic network will affect the T_d5 and dynamic mechanical properties of nanocomposites. EP-APTMS-BTB system with BTB content lower than 10 wt % showed good dynamic mechanical property and thermal stability (T_d5 increased from 336°C to 371°C, char yield increased from 27.4 to 30.2%). EP-GPTMS-BTB system

exhibited similar thermal stability and lower mechanical property.

ACKNOWLEDGMENTS

The authors would like to thank the National Science Council of the Republic of China and National Taiwan University of Science and Technology for financially supporting this work with Grants NSC 100-2221-E-011-030-MY3 and 102H451201.

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