Processing – Morphology Regulation of Epoxy/Layered-Silicate Nanocomposites

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ABSTRACT: Polymer/layered-silicate nanocomposites have unique and hierarchical structures that can provide improvements to the properties of polymeric materials. Controlling the dispersion of the nanomaterials through processing greatly influences the resulting morphology and the resulting properties of the nanocomposite. In this article, the dispersion behavior of organic layered silicates (OLS) as a function of the processing procedure is reported. The behavior of the OLS in all stages of processing—in the solvent, the epoxy prepolymer, and in the epoxy through cure—is discussed. On the basis of understanding of the dispersion behavior of the OLS in the epoxy resin at each stage of processing, a different processing procedure can be designed and used so that the morphology of the epoxy/layered-silicate nanocomposite can be regulated. Mild low-shear processing resulted in an intercalated nanocomposite with large-size aggregates (> 10 μm), and high-shear processing resulted in an intercalated nanocomposite with relatively small-size aggregates (0.5–3 μm), whereas the high-shear and ultrasonication processing procedures gave rise to an exfoliated nanocomposite. © 2008 Wiley Periodicals, Inc. J Appl Polym Sci 108: 3324–3333, 2008

Key words: morphology; processing; SAXS; TEM; thermosets

INTRODUCTION

Polymer/layered-silicate nanocomposites incorporate two-dimensional silicate nanosheets within a polymer matrix. The nanosized filler provides the advantage that it creates an extremely large volume of interphase between the pure original polymer and the nanofiller. This provides great potential for tailoring the properties of the polymeric materials with the addition of only a small amount of nanosized filler. In comparison with traditional micrometer-sized fillers, the lower loading of the nanofiller also makes it possible to maintain processing and manufacturing procedures close to those of the unfilled original polymer materials. Thus far, polymer/layered-silicate nanocomposites have shown improved mechanical properties, thermal properties, flame retardancy, and ablation properties, and they have shown great commercial potential.[1–7]

Generally, the polymer/layered-silicate nanocomposite can be classified as having an intercalated or exfoliated morphology.[1,2,5] As the polymer chain penetrates the gallery of the layered silicate, the gallery height is increased. If the ordered structure of tactoids is maintained and the tactoids appear as islands in the matrix, this morphology of the nanocomposite is generally regarded as being intercalated. However, if the ordered structure of the tactoids is destroyed and the silicate nanolayers are randomly distributed throughout the entire polymer matrix, this morphology is regarded as being fully exfoliated. The morphology of the polymer/layered-silicate nanocomposite is typically determined with X-ray scattering (relatively global morphology information, reciprocal space) and transmission electron microscopy (TEM; relatively local morphology information, real space).

It is generally difficult for traditional fillers to have simultaneous improvements in modulus and toughness without the sacrifice of the working temperature. However, due to the extremely large interfacial areas in the polymer/layered-silicate nanocomposite, it is possible to have simultaneous increases in modulus and toughness.[2,5] Generally, the exfoliated morphology is favored for the optimization of the mechanical and barrier property benefits of nanosilicates. However, sometimes, the intercalated structure is favored, as is the case for the improvement of the cation's mobility in the gallery. In short,

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it is very important to control the morphology of the polymer/layered-silicate nanocomposites to achieve the desired improvement in the polymeric materials.

Epoxy resin is a very common thermosetting material and has wide applications in different fields, such as adhesives, coatings, electronic encapsulates, medical applications, sporting goods, automobiles, and aerospace. The properties of epoxy resin can be further tuned after the addition of the layered silicate. Although very extensive research on epoxy/layered-silicate nanocomposites has been carried out in the last decade,[8–29] the work has been focused on the preparation, characterization of the epoxy nanocomposite,[8–17,22–29] and the development of the interplanar spacing of the layered-silicate during cure.[18–22] A recent article[30] reported the effect of ultrasonication (US) dispersion methods on the properties of epoxy/layered-silicate nanocomposites, but all of the morphologies of the nanocomposites were still intercalated.[30] So the morphology control of the layered-silicate nanocomposite has proven somewhat harder and very challenging. In this article, the study of the dispersion behavior of organic layered silicates (OLS) through the processing procedure is reported. The behavior of the OLS in all stages of processing—in the solvent, the epoxy prepolymer, and in the epoxy through cure—are discussed. On the basis of understanding of the dispersion behavior of the OLS in the epoxy resin during processing, general-process morphology guidelines can be developed, and the morphology (intercalated or exfoliated) of the epoxy/layered-silicate nanocomposite can be regulated.

EXPERIMENTAL

Materials

Epon 862 (diglycidyl ether of bisphenol F epoxy) and the curing agent W (diethyltoluenediamine) were purchased from Miller-Stephenson Chemical Co. (Danbury, CT). *n*-Octadecylamine was obtained from Aldrich Chemical Co. (Milwaukee, WI). Hydrochloric acid was obtained from Fisher Scientific (Lawn, NJ). Sodium montmorillonite was obtained from Southern Clay Products (Gonzale, TX). The cation exchange capacity of the sodium montmorillonite was about 92 mequiv. The organoclay used in this study was SC18 ($C_{18}H_{37}NH_3^+$-montmorillonite) and was prepared with well-established ion-exchanged chemistry.[17]

Processing

Low shearing

The desired amount of organoclay and Epon 862 were mixed with a stirring bar at about 65°C for 3 h. After the mixture was degassed, the stoichiometric amount of curing agent W was added and mixed for another 15 min. The mixture was degassed again and cast in a silicone mold. The mixture was cured at 121°C for 2 h; this was followed by another 2-h postcure at 177°C.

High shearing

The organoclay and acetone were vigorously mixed with a high-shear mixer (IKA -Labortechnik Janke and Kunkel GMBh Co., Germany, 13,500 rpm) in a sonication bath. Then the Epon 862 and acetone mixture was added, and mixing continued by high-shear mixing in a sonication bath for 2 h. After the acetone was evaporated and the mixture was degassed, the stoichiometric curing agent W was added and mixed with a stirring bar. The mixture was degassed later. The mixture was cured in an oven at 121°C for 2 h and postcured at 177°C for 2 h.

High shearing and US

After the organoclay and acetone were vigorously mixed with a high-shear mixer in a sonication bath, the Epon 862 and acetone mixture was added, and mixing was continued by high-shear mixing in a sonication bath for 2 h. Then, the solvent was evaporated, and the organoclay/Epon 862 mixture was ultrasonicated (US probe, Fisher Scientific, Fair Lawn, NJ, 60 Sonic Dismembrator, frequency = 22.5 kHz, out power = 12 W) for 1 h. The mixture was degassed. A stoichiometric amount of curing agent W was added and ultrasonicated for 15 min. The resulting mixture was degassed, cast in a mold, cured in the oven at 121°C for 2 h, and postcured at 177°C for 2 h.

One-pot US

The mixture of SC18/Epon 862/W in the presence of acetone (volume = 1 : 1 for acetone/Epon 862) was mixed under stirring-bar mixing and US (US probe, Fisher Scientific, 60 Sonic Dismembrator, frequency = 22.5 kHz, out power ≈ 10 W) simultaneously for several hours. The acetone was evaporated, and the resulting mixture was degassed and cured in the oven at 121°C for 2 h and postcured at 177°C for 2 h.

Characterization

Wide-angle X-ray diffraction was performed in a Rigaku X-ray powder diffractometer (Tokyo, Japan). The generator power was 40 kV and 150 mA; scanning was from 2 to 10°, and the scan mode was continuous with a scanning rate of 0.8°/min. Small-angle X-ray scattering (SAXS) studies were performed at Beamline X27C with a one-dimensional detector at the National Synchrotron Light Source in Brookhaven National Laboratory. The sample for TEM was microtomed in a Reichert-Jung Ultracut Microtome (Phoenix, AZ) and mounted on 200-mesh

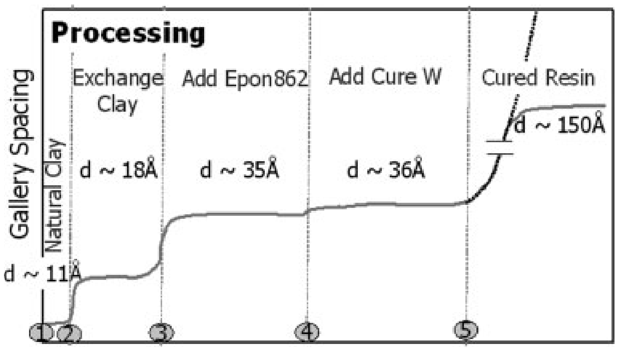

Figure 1 Interplanar spacing change as a function of processing procedure.

copper grids. TEM was performed with a Philips CM200 transmission electron microscope with a LaB_6 filament operating at 200 kV. Light microscopy was performed with a Nikon Microphot-FXL (Tokyo, Japan). Differential scanning calorimetry (DSC) was performed with a TA Instruments differential scanning calorimeter 2920 modulated DSC (Woodland, CA) at 2°C/min with air sweep gas.

RESULTS AND DISCUSSION

Behavior of the OLS in all stages of processing

The starting materials for the aerospace epoxy resin were composed of two components, including Epon 862 (diglycidyl ether of bisphenol F) and curing agent W (diethyltoluenediamine). Epoxy nanocomposites were made via the *in situ* polymerization methodology.

Sodium montmorillonite is hydrophilic in nature and is not compatible with hydrophobic polymers. The organotreatment of the surface of the silicate nanosheets can reduce the surface energy of the silicate nanosheets and, therefore, make the silicate nanosheets compatible with the polymer. The X-ray diffraction showed that the interplanar spacings were 18.0 Å for SC18 compared with 11.2 Å for original sodium montmorillonite (Fig. 1). When Epon 862 was mixed with SC18 via low shearing or high shearing in the presence of solvent, they appeared compatible and well dispersed. The X-ray diffraction showed that the 3.7 wt % SC18/Epon 862 mixture had an expanded gallery (~35 Å) due to the penetration of Epon 862 into the gallery (Fig. 1). Both the high-shear and low-shear processing procedures gave the same gallery height and an intercalated nanostructure.

The original particle size of the organoclay was in the range of several micrometers to about 70 μm. The images of light microscopy are shown in Figure 2. When the organoclay (SC18) was mixed with acetone under high-shear mixing, after 5 min the image indicated the particle size to be between 20 and 40 μm. After 30 min of high-shear mixing, the particles were further broken into smaller ones with sizes from several to 30 μm. After 110 min of

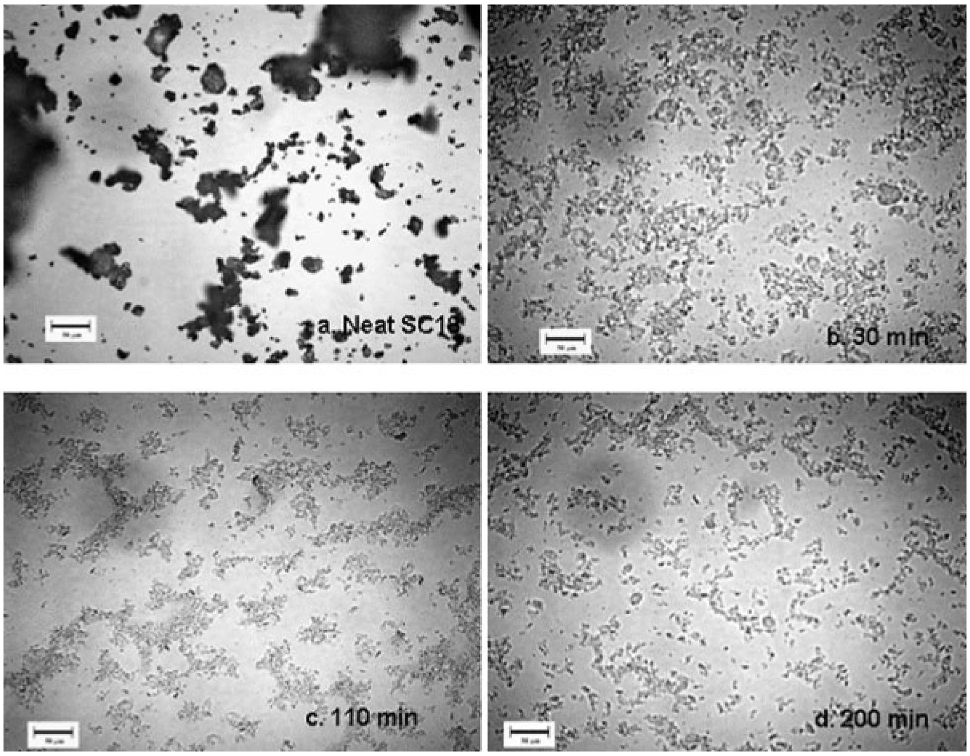

Figure 2 Light microcopies of (a) neat SC18 and SC18 in acetone after (b) 30, (c) 110, and (d) 200 min under high-shear mixing (bar size = 50 μm).

TABLE I
Summary of the Epoxy/Layered-Silicate Nanocomposites with Different Morphologies Through Different Processing Procedures

	a	b	c	d
Processing	Low-shear	High-shear	US (one-pot, acetone)	High-shear plus US
Interplanar spacing (Å)	~150	~ 150	~ 150	No aggregation
Aggregate size	> 10 μm	0.5–3 μm	100–500 nm lateral, 10s of nanometers thick, homogeneous, and random	100–500 nm lateral, nanometers thick, homogeneous, and random
Number of nanolayers per aggregate	Hundreds	30–200	2–8	1 or 2

mixing, the samples looked very homogeneous with typical particle sizes in the range of a few micrometers to 10 μm. After 135 min, the size distribution was in the range of tenths of micrometers to 10 μm. After 200 min, the size distribution was very uniform and in the range of a few micrometers. This size particle was expected because the high-shear mixing used is typically used for the separation of micrometer-size particles (Table I).

When Epon 862 was added to the dispersed organoclay–acetone mixture, the mixture was mixed under high shear in a sonication bath. Some images of light microscopy are shown in Figure 3. After 15 min of mixing, the light microscopy images indicated that the particles were separated and had particle sizes very similar to that before the addition of Epon 862. After 60 min, good dispersion was maintained with the same particle size. However, after 90 min, there appeared to be limited aggregation of small particles. At this stage, the clay particle size was in micrometers, and the distribution of the particle in the mixture was homogeneous.

Because the Epon 862 could penetrate into the gallery of the organoclay during the processing procedure of the epoxy nanocomposite at both low-shear and high-shear mixing, the silicate nanolayers were still stacked together. However, the particle sizes were different for these two processes. The large particles of the clay were broken into much smaller particles through high-shear mixing. The particle size in low-shear mixing was from several micrometers to about 40 μm, whereas the particle size was a

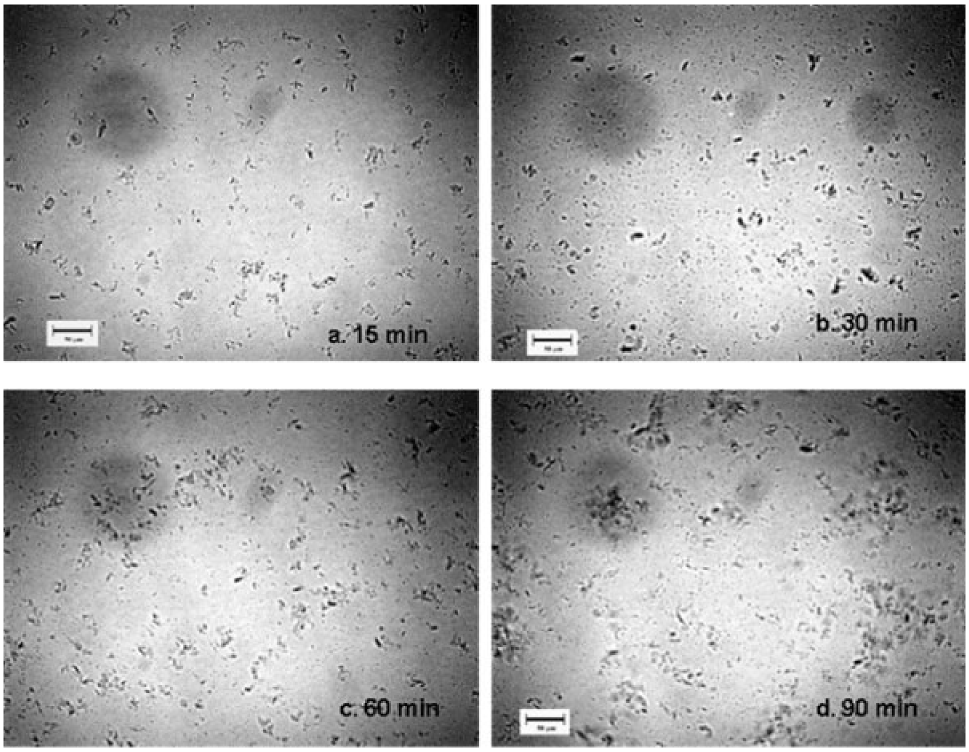

Figure 3 Light microcopies of the SC18/Epon 862 mixture with acetone after (a) 15, (b) 30, (c) 60, and (d) 90 min under high-shear mixing (bar size = 50 μm).

few micrometers after high-shear mixing. Although the clay was not in the exfoliated morphology after high-shear mixing, the organoclay was homogeneously dispersed in the Epon 862 in a much smaller size.

The morphology of the SC18/Epon 862 mixture typically had an intercalated nanostructure with an interplanar spacing of about 35 Å. When curing agent W was added, the SAXS data indicated an interplanar spacing of about 36 Å. There was almost no gallery expansion, which indicated that curing agent W was not well compatible with SC18. After that was the thermal curing step. Our previous research[22] on the study of morphology development during curing through *in situ* SAXS showed the following morphology development during cure. When the temperature was elevated from 60 to 98°C, the interplanar spacing was shifted from 38 to 42 Å. The increase in the gallery of the organoclay was very limited. At 104°C, the peak almost disappeared. This indicated that the ordered structure had almost collapsed. DSC studies showed that the onset temperature of curing for the 3 wt % SC18/Epon 862/W mixture was 100°C, much lower than that of pure Epon 862/W (∼120°C). The catalytic effect was due to the acidic organic pendent group ($C_{18}H_{37}NH_3^+$) inside the gallery of SC18. When the temperature was higher than 100°C, the curing took place for the epoxy resin inside the gallery of the organoclay, whereas there was little polymerization occurring for the epoxy resin outside the gallery. The consumption of the epoxy monomer inside the gallery was the driving force for the migration of the epoxy resin from outside the gallery to inside the gallery. The migration of the epoxy resin through the edges caused the clay nanolayers to curve to create a more disordered structure. With more migration of epoxy resin into the gallery, the curved sheets gradually became flat and ordered. The (001) peak of SAXS became more intense until the epoxy was almost fully cured, and the interplanar spacing ended with 150 Å at 200°C.

The morphology development of the mixture of 3.7 wt % SC18/Epon 862 with high shear with acetone and curing agent W was almost the same as that with curing agent W under low-shear conditions. The high-shear mixing procedure essentially broke the particles into much smaller ones with a relatively homogeneous dispersion. However, high shear affected the morphology on the micrometer level but had little effect on the nanoscale level.

A summary of the gallery expansion of the layered silicate during different processing procedures is shown in Figure 1. The original interplanar spacing between the galleries of the original sodium montmorillonite was 11.2 Å. After the sodium cation was exchanged with octadecylammonium cation, the interplanar spacing increased to about 18 Å. When SC18 was mixed with Epon 862, the compatibility and penetration of Epon 862 into the gallery expanded the gallery to about 35 Å. After the addition of curing agent W, the gallery expansion was very, very limited, with just about a 1-Å increase. However, a very significant expansion of the gallery took place during the cure.

Morphology regulation of the epoxy/layered-silicate nanocomposite

As discussed previously, the morphology development on the nanoscale had two stages. The first stage was the penetration of Epon 862 into the gallery of the organoclay (SC18). When equilibrium was reached, the interplanar spacing of the gallery expanded from the original 18 to 35 Å. The second stage was the large expansion of the gallery in the curing stage from 35 to about 150 Å. Apparently, these two stages affected the nanostructure more than the microstructure. Although both the low-shear and high-shear processing could sometimes cause some breaking of the aggregates, the aggregates were still micrometer sized. By controlling the size of the aggregate with processing before curing, we could obtain a controllable hierarchical intercalated nanocomposite. When mild low-shear processing conditions were applied for the mixing of SC18 with Epon 862, the original large aggregates were retained. More aggressive processing with high shearing and US should systematically decrease the aggregate size. The mildest processing procedure for mixing perhaps is mixing with a stirring bar. The stirring-bar mixing procedure was applied for the mixture of SC18 with Epon 862 at an elevated temperature (∼60°C) followed by a general cure procedure. The SAXS curve of the cured 3 wt % SC18/Epon 862/W nanocomposite is shown in Figure 4. The interplanar spacing was about 150 Å, which was

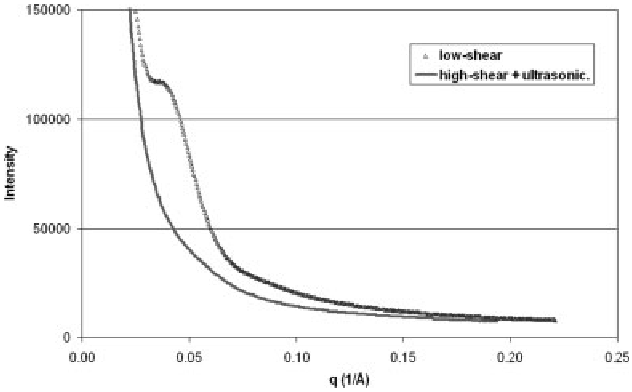

Figure 4 SAXS of the cured 3 wt % SC18/Epon 862/W nanocomposite through low-shear mixing and through high-shear and US processes.

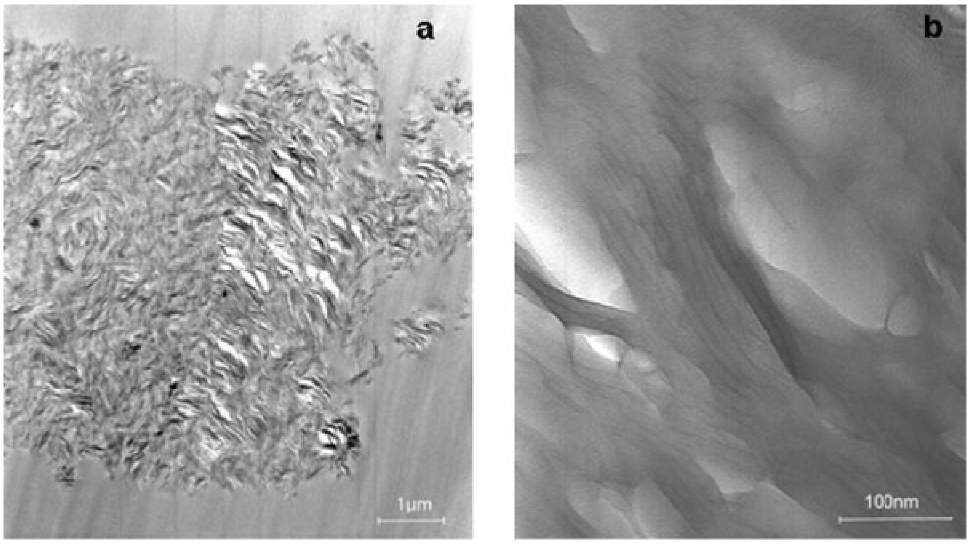

Figure 5 TEM images of the cured 3 wt % SC18/Epon 862/W nanocomposite through low-shear mixing.

substantially different from the original interplanar spacing of SC18 of 18 Å. TEM images at low and high magnification were taken, and representative images are shown in Figure 5. The dark lines were from silicate nanolayers and represent a two-dimensional projection of a three-dimensional material. As shown by the low magnification, the silicate nanolayers were in the aggregated state. The size of most aggregates was larger than 10 μm. There were a lot of domains of pure epoxy without silicate nanolayers. The aggregates appeared as islands in the sea. From the high magnification, it is clear that the silicate nanolayers were still aggregated and were in registry. The interplanar spacing measured from the TEM image was approximately 15 nm, which was very consistent with SAXS data. In short, the intercalated nanocomposite had very large interplanar spacing and relatively large aggregates.

For the intercalated nanocomposite with relatively small aggregates, we needed to break the aggregates into smaller ones when SC18 was mixed with Epon 862 with a more intense shearing. In the second processing scheme, high-shear mixing was applied. To increase the amount of shear, it was important to efficiently couple the energy with the right combination of applied shear and solution viscosity. In general, high-shear mixing was limited to breaking the aggregate into micrometer-sized particles. The SC18/Epon 862 mixing after high-shear mixing was also followed by the addition of curing agent W and subsequent curing. The SAXS of the cured 3 wt % SC18/Epon 862/W nanocomposite through high-shear mixing is shown in Figure 6. The interplanar spacing was about 150 Å, which was the same as that obtained through the stirring-bar mixing procedure. TEM images of this nanocomposite at low and high magnifications were also taken and are shown in Figure 7. As shown by the low magnification, the silicate nanolayers were still aggregated. The size of the aggregates ranged from 0.1 to 5 μm with most in the range 0.5–3 μm. The silicate nanolayer number in the aggregates was mostly in the range from about 30 to about 200. There were some domains of pure epoxy without silicate nanolayers. The aggregates appeared as islands in the sea (epoxy matrix). However, the distribution of the aggregates in the epoxy matrix was more homogeneous than those obtained through the low-shear mixing procedure. From the high magnification, it is clear that the silicate nanolayers were still in registry with the interplanar spacing of 15 nm, which was consistent with the SAXS data. This was the intercalated nanocomposite with very large interplanar spacing and relatively small aggregates.

The exfoliation of the layered silicate inside the epoxy matrix proved to be somewhat more difficult

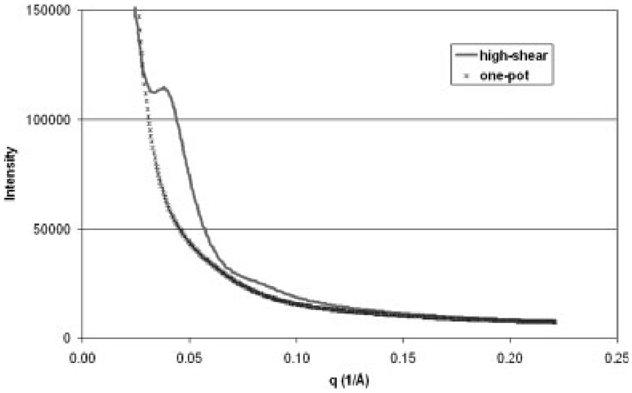

Figure 6 SAXS of the cured 3 wt % SC18/Epon 862/W nanocomposite through high-shear mixing and through one-pot processing with simultaneous US and stirring-bar mixing with acetone.

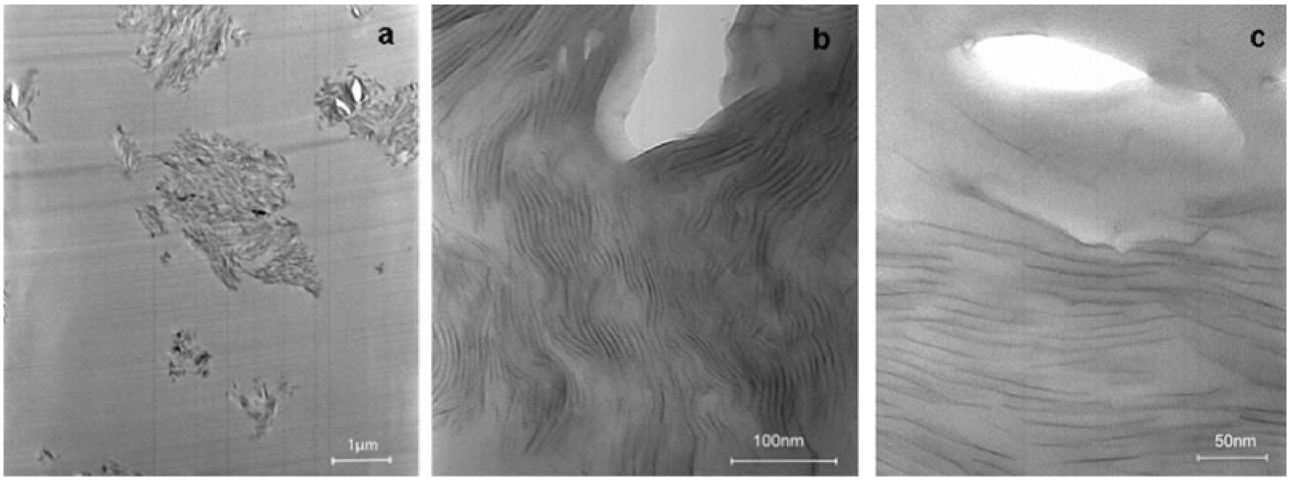

Figure 7 TEM images of the cured 3 wt % SC18/Epon 862/W nanocomposite through high-shear mixing.

than with other polymers, such as polyamide, due to the limits of the processing window during cure. The majority of the previously claimed exfoliated nanocomposites in epoxy have not ruled out the possibility of having an intercalated structure with an interplanar spacing larger than was measured by X-ray analysis. The preparation of the exfoliated epoxy/layered-silicate nanocomposites was a challenge but was very significant both for the academic interest and engineering applications. As discussed previously, the high-shear mixing procedure could help disperse the aggregates relatively homogeneously in the epoxy matrix. To achieve the exfoliated nanostructure, high-shear mixing was used first. US was applied to the SC18/Epon 862 mixture after high-shear mixing with curing agent W. US itself produces heat, which simultaneously causes partial curing of the epoxy resin. It is believed that heating can increase the interplanar spacing via curing, whereas US simultaneously provides energy to aid separation. Thus, better dispersion could be achieved. Increased separation of the silicate nanolayers made the migration of the epoxy resin from the extragallery to the intragallery easier as the barrier to entry was greatly reduced. US and curing had a somewhat synergic effect on the separation of the silicate nanolayers. Therefore, it should have been possible to achieve a truly exfoliated nanostructure for the epoxy/layered-silicate nanocomposites.

The SAXS of the cured 3 wt % SC18/Epon 862/W nanocomposite through the processing procedure of high shear and US is shown in Figure 4. There was no peak at the very low q (q is the momentum transfer wave number of scattering vector ($q = 2\,\pi/d = 4 \sin\theta/\lambda$), where λ is the wavelength of the incident radiation, θ is half the Bragg scattering angle, and d is the basal plane separation) region in the SAXS, which indicated that there was no ordered structure in this nanocomposite to within the measurable high spacing range of about 320 Å. This X-ray pattern was consistent with the exfoliated nanocomposite. To further confirm the morphology, the TEM images at both low and high magnification of the cured 3 wt % SC18/Epon 862/W nanocomposite through the preceding procedure of high shear and US are shown in Figure 8. As shown by the image at low magnification, there was no aggregate. The silicate nanolayers were dispersed randomly and continuously throughout the epoxy matrix. The distribution of the silicate nanolayers was a disordered exfoliation. The silicate nanolayers were mostly individual nanolayers, with a few doublets or triplets. The US time in this nanocomposite processing after the addition of curing agent W was 15 min. We also tried to extend the US time after the addition of curing agent W to 30 min. The TEM images showed an almost perfect exfoliated morphology.[26] Almost all of the silicate nanolayers were individual nanosheets with few doublets or triplets. However, the relatively lower degree of curing in the 15-min US procedure made the following procedure, such as degas, much easier, and the morphology was very close to a fully exfoliated morphology or could be considered as an exfoliated morphology.

To this point, three different morphologies of layered silicate epoxy nanocomposite have been achieved. They varied from the intercalated nanocomposite with large gallery height and large aggregates to the intercalated nanocomposite with large gallery height and small aggregates and finally to the exfoliated nanocomposite. However, there were still some interesting morphologies between the intercalated nanocomposites with small aggregates and the exfoliated nanocomposite. The typical size of the small aggregate in the intercalated nanocomposite varied from several hundred nanometers to several micrometers. On the basis of the interplanar spacing of 15 nm obtained from the SAXS data, the

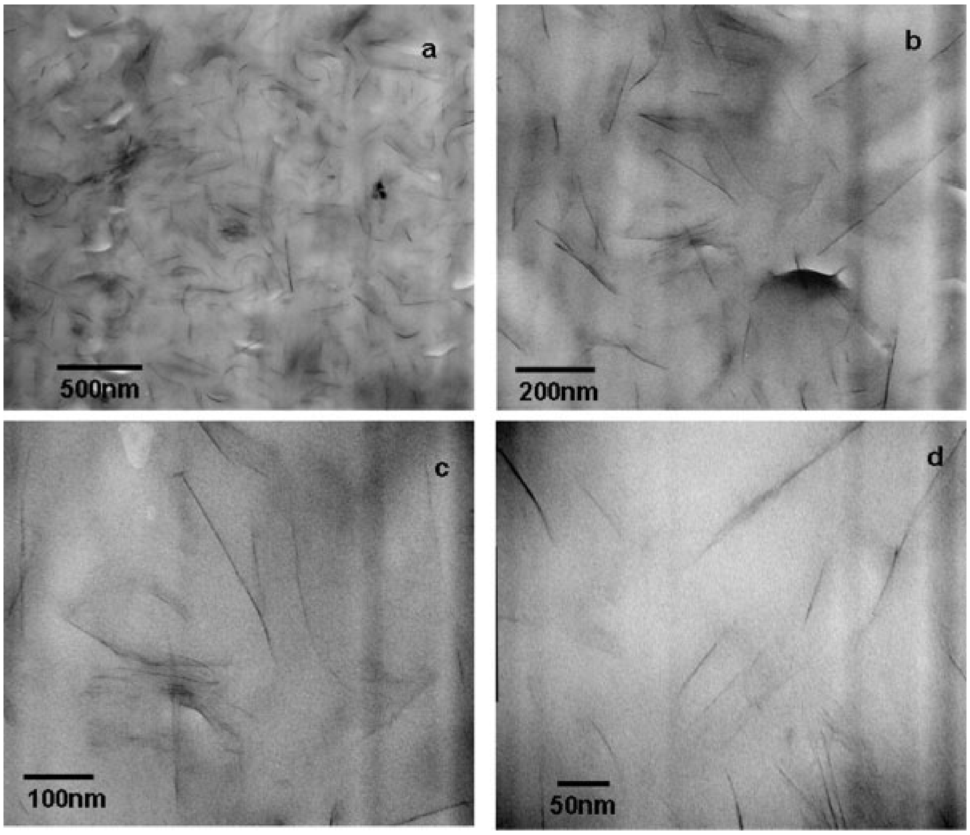

Figure 8 TEM images of the cured 3 wt % SC18/Epon 862/W nanocomposite through high-shear and US processes.

average number of nanolayers in one aggregate could be 30–200 with the assumption that the silicate nanolayer was 1 nm thick and 100 to 500 nm in lateral size. Interestingly, a morphology that has a large aggregation of several stacked nanolayers may be considered more homogeneous than an intercalated nanocomposite with small aggregates. This could be a very interesting morphology to investigate.

As discussed previously, the processing procedure for the creation of an exfoliated epoxy/layered-silicate nanocomposite was the use of high-shear mixing of the SC18/Epon 862 mixture in the presence of a large amount of acetone to break the large aggregates into relatively small ones, followed by US of the SC18/Epon 862/W mixture. One alternative is to just use the US procedure because US can also break the large aggregates into smaller ones; thus a so-called one-pot procedure was tried. In this one-pot procedure, all of the materials, including SC18, Epon 862, curing agent W, and a relatively small amount of acetone were mixed with a stirring bar and US. The presence of acetone could reduce the viscosity and make the processing window much larger. Although the container was mostly covered with aluminum foil, the acetone still gradually evaporated. After several hours, most of the acetone evaporated. Also, US caused partial curing. The sample was then degassed and cured. The SAXS of the cured 3 wt % SC18/Epon 862/W through the one-pot processing of simultaneous US and stirring-bar mixing in the presence of acetone is shown in Figure 6. There was no peak in the very low q region of the SAXS of this nanocomposite, which indicated that there was no ordered structure in this nanocomposite. The TEM images of this nanocomposite from low to high magnification are shown in Figure 9. As clearly shown by the low-magnification images, the silicates were very well dispersed throughout the entire epoxy matrix. From the high-magnification images, most of the silicate nanolayers were not individual nanolayers. The aggregation of most layered silicates was composed of several silicate nanolayers (2–8 layers), and only a few of the layered silicates were composed of approximately 10 nanolayers. In addition, all of these very small aggregations were randomly and homogeneously dispersed in the whole epoxy matrix. Randomly distributed aggregates with such short-range order are difficult to detect with SAXS. However, this one-pot procedure not only can provide a convenient alternative to make a well-dispersed epoxy/layered-silicate nanocomposite but also provides a new morphology that is different from the general intercalated and exfoliated nanostructure. Although the silicate nanolayers were still aggregated, the aggregation consisted of several silicate nanolayers that were randomly and homogeneously dispersed

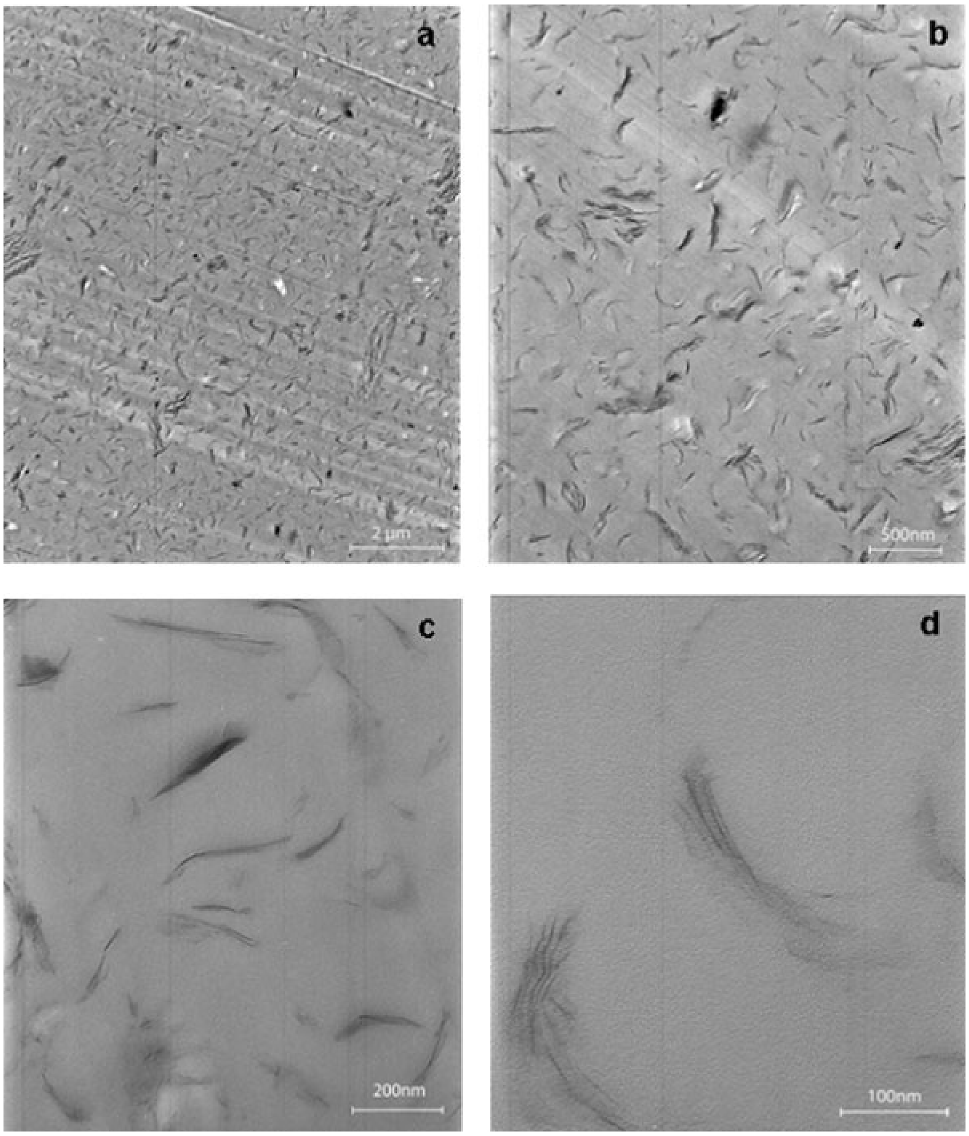

Figure 9 TEM images of the cured 3 wt % SC18/Epon 862/W nanocomposite through one-pot processing with simultaneous US and stirring-bar mixing with acetone.

throughout the epoxy matrix. This morphology was close to the exfoliated nanostructure. It will be very interesting to investigate the effect of the morphology of these dispersed aggregates of several nanolayers on a nanocomposite's mechanical properties, such as toughness.

A summary of the different morphologies through different processing procedures is shown in Table I. A complete property measurement, including mechanical and barrier properties, is needed in the future to determine the morphology–property relationship for this type of epoxy/layered-silicate nanocomposite.

CONCLUSIONS

The behavior of the OLS in all stages of processing—in the solvent, Epon 862, and in the epoxy through cure—was investigated. When the organoclay was mixed with acetone, the large particles of the clay were broken into much smaller ones under high-shear mixing, whereas there were few changes in the particle size under low-shear mixing. The particle size in low-shear mixing was from several micrometers to about 40 μm, whereas the particle size was a few micrometers after high-shear mixing. Epon 862 penetrated into the gallery of the organoclay during the processing procedure of the epoxy nanocomposite at both low-shear and high-shear mixing, and the silicate nanolayers were still stacked together. The nanoscale morphology development of the mixture of 3.7 wt % SC18/Epon 862 with high-shear mixing with acetone and curing agent W was essentially the same as that observed for low-shear mixing with just curing agent W. Thus, the high-shear mixing affected the morphology on the micrometer level and had little effect on the nanoscale level.

On the basis of the understanding of the morphology development of the epoxy/layered-silicate nanocomposite during processing, the morphology of the epoxy/layered-silicate nanocomposite can be regulated through carefully designed processing procedures. The consistent characterization from both SAXS and TEM of these nanocomposites verified the different morphologies of the epoxy/layered-silicate nanocomposites. The different morphologies of the epoxy/layered-silicate nanocomposite included (a) an intercalated nanocomposite with the silicate with 150-Å interplanar spacing and relatively large-size aggregates (> 10 μm), (b) an intercalated nanocomposite with 150-Å interplanar spacing and relatively small-size aggregates (0.5–3 μm, ~30 to 200 nanolayers per aggregation), (c) a well-dispersed nanocomposite with random and homogeneous dispersion of several-nanolayered aggregation (~2 to 8 nanolayers), and (d) an exfoliated nanocomposite, in which the individual silicate nanolayers were randomly and continuously dispersed in the polymer matrix.

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