Silica Nanoparticles Modified with Vinyltriethoxysilane and Their Copolymerization with *N,N′*-Bismaleimide-4,4′-Diphenylmethane

Heng-Lei Su,[1] Jung-Mu Hsu,[2] Jing-Pin Pan,[2] Chorng-Shyan Chern[1]

[1] *Department of Chemical Engineering, National Taiwan University of Science and Technology, Taipei 106, Taiwan*
[2] *Material and Chemical Research Laboratories, Industrial Technology Research Institute, Chutung, Hsinchu 310, Taiwan*

Received 7 March 2006; accepted 4 August 2006
DOI 10.1002/app.25313

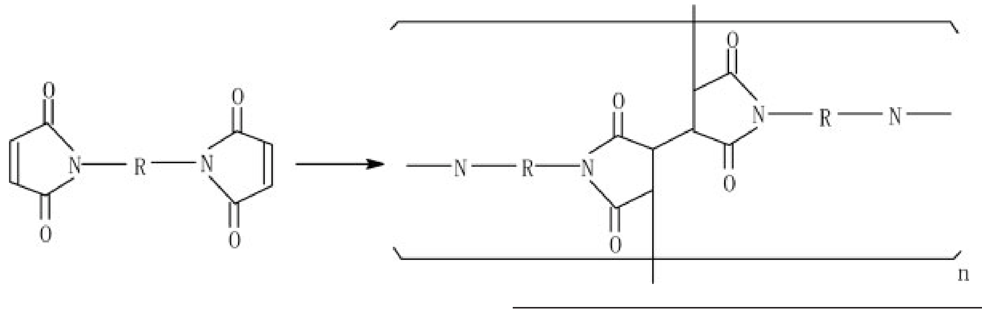
ABSTRACT: The synthesis and characterization of the vinyltriethoxysilane-modified silica nanoparticles were investigated. It was shown that the vinyltriethoxysilane molecules had been successfully grafted onto the silica nanoparticles. The native and silane-modified silica dispersions in *N*-methyl-2-pyrrolidone with the total solids contents within the range 1–6 wt % exhibited dramatically different flow behaviors. The polymerization of *N,N′*-bismaleimide-4,4′-diphenylmethane (BMI) initiated by barbituric acid in the presence of the native or vinyltriethoxysilane-modified silica nanoparticles were then carried out in γ-butyrolactone (total solids content = 20%). The higher the level of silica, the better the thermal stability of the BMI/silane/silica composite particles. The silane-modified silica particles significantly improved their dispersion capability within the continuous BMI oligomer matrix. Furthermore, the degree of dispersion of the vinyltriethoxysilane-modified silica particles in the BMI oligomer matrix decreased with the weight percentage of silica based on total solids increased from 20 to 40 wt %. © 2006 Wiley Periodicals, Inc. J Appl Polym Sci 103: 3600–3608, 2007

Key words: nanocomposites; silicas; radical polymerization; TGA; rheology

INTRODUCTION

Alkoxysilanes have been widely used to modify the silica particle surfaces.[1–7] The reaction mechanism generally involves the hydrolysis of the alkoxy groups of silane to form silanol groups, followed by the condensation reaction between the silanol groups of silane molecules and silica particles. Goerl et al.[2] studied the surface reaction of bis(3-triethoxysilylpropyl)tetrasulfane with siliica. These investigators proposed a horizontal reaction model in which a single siloxane bond was first formed with the silica particle surface. This was followed by condensation reactions between the silanol groups of the silane molecules bound to the silica particle surface. Both reactions could be accelerated in acidic or alkaline pH. When used as fillers (or additives) for various polymeric materials, silica modified by the coupling agent organosilanes offers advantageous performance properties such as the improved compatibility between the organic and inorganic phases in addition to the excellent scratch and abrasion resistance, hardness, thermal stability, rheological property, and gas solubility associated with silica.[8–13] Furthermore, alkoxysilanes containing various functional groups such as amino, epoxy and vinyl groups find a wide range of industrial applications.[1,3,4]

Pan et al.[14] successfully used barbituric acid (BTA) to initiate free radical polymerization of *N,N′*-bismaleimide-4,4′-diphenylmethane (BMI) with two C=C:

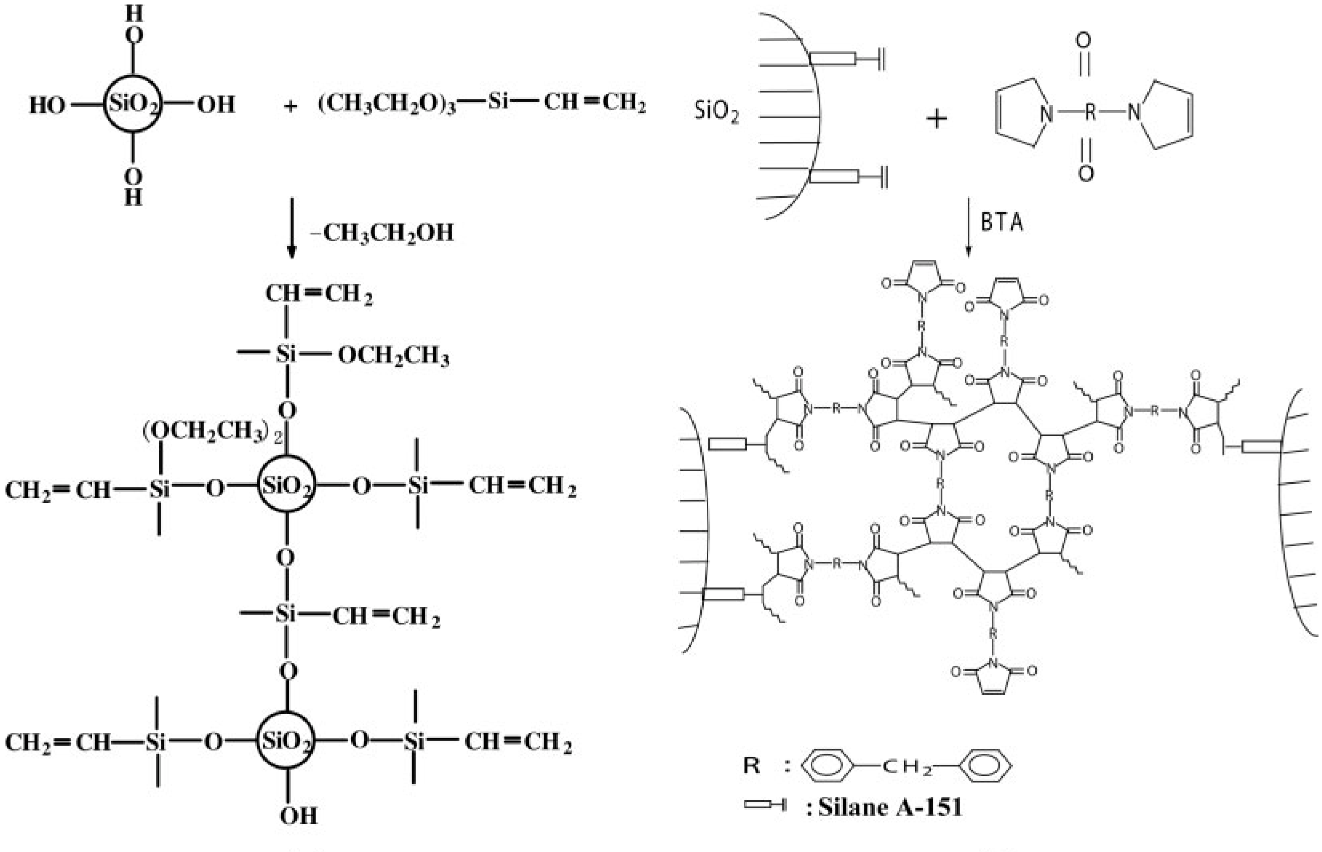


It was shown that the polymerization was primarily controlled by the level of BTA, and the resultant hyper-branched oligomers (molecular weight [MW] = ∼3000) were soluble in organic solvents. Thus, such

---

Correspondence to: C.-S. Chern (cschern@mail.ntust.edu.tw).

Journal of Applied Polymer Science, Vol. 103, 3600–3608 (2007)



(a) (b)

Scheme 1 (a) Modification of silica nanoparticles with vinyltriethoxysilane. (b) Polymerization of BMI initiated by BTA in the presence of the vinyltriethoxysilane-modified silica nanoparticles.

a unique BMI oligomer with excellent mechanical, thermal, and processing properties represents a molecular-level reinforcing agent. In this study, we propose to modify silica nanoparticles by vinyltriethoxysilane. The vinyl groups of silane molecules grafted onto the silica nanoparticle surface can participate in the subsequent free radical polymerization of BMI molecules initiated by BTA (Scheme 1). The objective of this work is to prepare and characterize such organic-inorganic hybrid colloidal particles, which can impart even better performance properties to the formulated composite materials.

EXPERIMENTAL

Materials

The hydrophilic fumed silica Aerosil 200 (Degussa, Frankfurt am Main, Germany) used in this work has a silica content of ≥99.8 wt %, a moisture content of ≤1.5 wt %, an average primary particle diameter of 12 nm and a specific surface area of 200 ± 25 $m^2\ g^{-1}$. The coupling agent vinyltriethoxysilane ($CH_2{=}CH{-}Si(OC_2H_5)_3$, Silquest A-151) was obtained from OSi Specialties (Crompton, Middlebury, CT). Other chemicals used in this study include *N,N*′-bismaleimide-4,4′-diphenylmethane (Sigma-Aldrich, St. Louis, MO), barbituric acid (Merck, Darmstadt, Germany), γ-butyrolactone (TEDIA, Fairfield, OH), *N*-methyl-2-pyrrolidone (Asia Union Electronic Chemical, Taipei, Taiwan), and *n*-hexane (TEDIA, Fairfield, OH). All chemicals were used as received.

Experimental methods

The vinyltriethoxysilane-modified silica nanoparticles were prepared by adding Aerosil 200 and Silquest A-151 [1/2 (w/w)] into a 250-ml three-neck flask equipped with a magnetic stirrer, thermometer, and a condenser. The solvent-free hydrolysis and condensation reactions were carried out at 190°C for 1 h. The resultant crude product was washed with an excess of *n*-hexane, followed by filtration. This procedure was repeated three times in order to remove the unreacted silane molecules (if present). The purified product was then dried at 165°C for less than 30 min. The free radical polymerization of BMI initiated by BTA in the presence of the silane-modified silica was initiated in a 250-ml three-neck flask at 70°C for 10 min and then continued at 130°C for 4 h. γ-butyrolactone was used as the major

TABLE I
Recipes for Polymerization of BMI Initiated by BTA in the Presence of Native or Vinyltriethoxysilane-Modified Silica Nanoparticles

BMI (g)	Native silica (g)	Silane-modified silica (g)	Silica (wt %)[a]	Appearance	Flow
0.386	0.0444	—	10	Transparent[b]	Excellent
0.386	0.100	—	20	Transparent[b]	Excellent
0.386	0.171	—	30	Not dispersive	No fluidity
0.386	0.267	—	40	Not dispersive	No fluidity
1.689	—	0.194	10	Transparent[c]	Excellent
1.689	—	0.437	20	Transparent[c]	Excellent
1.689	—	0.750	30	Transparent[c]	Excellent
1.689	—	1.166	40	Transparent[c]	Excellent

[a] Weight percentage of silica based on total solids.
[b] Brown.
[c] Dark brown.

component of the continuous phase and its amount was adjusted to achieve the target total solids content of 20 wt % in the formula. The polymerization was then terminated by cooling the reaction system down to ambient temperature. The recipes used in this work are summarized in Table I. The molar ratio of BMI to BTA was kept constant at 10 : 1 and the amount of the native or silane-modified silica was adjusted accordingly to give a prescribed weight percentage of silica based on total solids (10, 20, 30, or 40 wt %) throughout this work.

The BMI/silane/silica hybrid materials were characterized by Fourier transform infrared (FTIR) in the transmission mode at 400–4000 cm^{-1} using a Spectrum GX spectrometer (Perkin-Elmer, Shelton, CT). Thermal loss curves were obtained from the thermal gravimetric analysis (TGA; Perkin-Elmer TGA7, Shelton, CT). Measurements were performed from ambient temperature to 800°C at a heating rate of 10°C min^{-1} in N_2 atmosphere. The morphologies of the colloidal particles were observed by field emission scanning electron microscopy (FESEM; JEOL JFM-6500F, Tokyo, Japan) equipped with energy-dispersive X-ray (EDX) spectroscopy. Before the TGA and FESEM experiments, the samples were prepared by coating the colloidal dispersions on glass substrates, followed by baking the composite films at 205°C for 30 min and then at 250°C for another 30 min. The rheological properties of the dispersions at 25°C were measured by a strain-controlled rheometer with a parallel plate geometry (diameter = 50 mm, gap = 0.4 mm) (TA Instruments AREL-LS1, New Castle, DE). The native silica and vinyltriethoxysilane-modified silica particles were dispersed in *N*-methyl-2-pyrrolidone with the total solids content within the range of 1–6%. Two modes used in this study were the steady-rate sweep test (viscosity as a function of the shear rate within the range of 1–10,000 s^{-1}) and the transient thixotropic loop test (i.e., measured the shear stress in response to changes in the shear rate, which was continuously increased from 0 to 4000 s^{-1} and then decreased from 4000 to 0.1 s^{-1}).

RESULTS AND DISCUSSION

FTIR analysis

The FTIR spectra for the vinyltriethoxysilane and the native and silane-modified silica particles are shown in Figure 1. The absorption peaks at 2975, 1390, and 544 and 470 cm^{-1} are attributed to the $=CH_2$ symmetric stretching vibration and $C—CH_3$ asymmetric deformation vibration, $C—CH_3$ and C—H asymmetric deformation vibrations, and $CH_3—CH_2$ deformation vibration, respectively. The native silica exhibits the Si—OH and O—H stretching at 3400 and 1630 cm^{-1} and the Si—O—Si network vibration at 1089 cm^{-1}. The observed characteristic absorption peaks at 2975 and 1089 cm^{-1} and the disappearance of the absorption peaks at 3400, 1630, and 544 cm^{-1} confirm the successful grafting of vinyltriethoxysilane onto the silica particles.

TGA analysis

Figure 2(a) shows the TGA curves of the native silica particles and vinyltriethoxysilane-modified silica particles [silica/silane = 1/2 (w/w)]. It should be noted

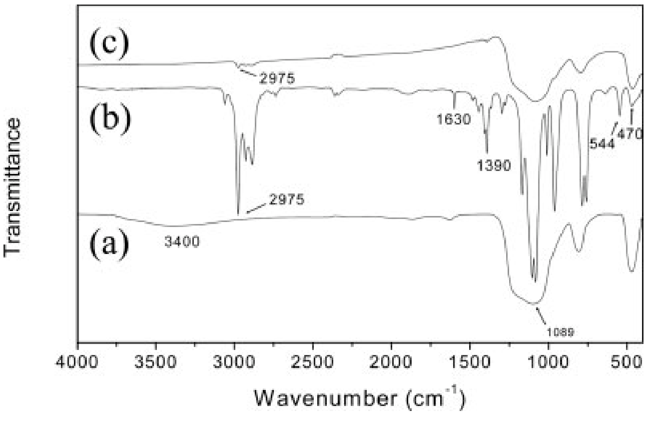


Figure 1 FTIR spectra for (a) native silica, (b) vinyltriethoxysilane and (c) silane-modified silica.

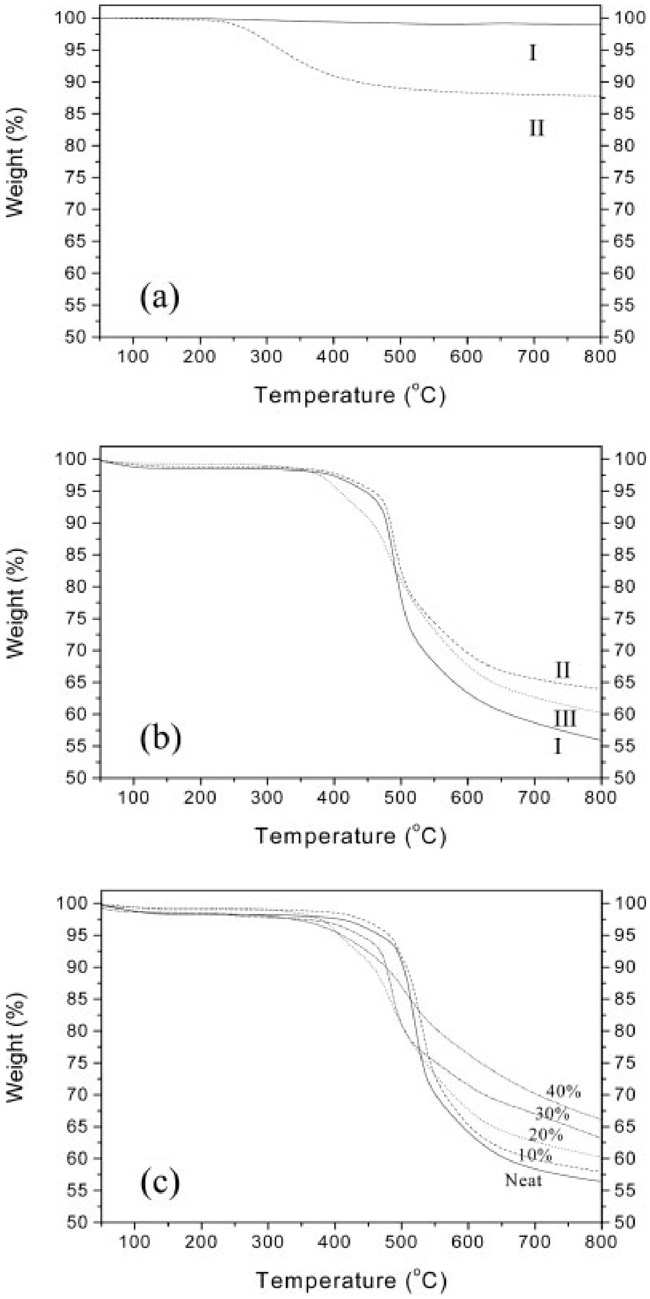


Figure 2 TGA curves of (a) native silica particles (I) and vinyltriethoxysilane-modified silica particles [silica/silane = 1/2 (w/w)] (II), (b) neat BMI oligomer (I) and BMI/silica (II) and BMI/silane/silica composite particles (20 wt % silica based on total solids) (III), and (c) neat BMI oligomer and BMI/silane/silica composite particles (10–40 wt % silica based on total solids).

that both the native silica and vinyltriethoxysilane-modified silica samples were not thermally treated at 205 and then 250°C for 30 min each stage. The weight loss before 200°C is presumably attributed to the content of water adsorbed on the silica particle surfaces (~0.05% and 0.29% for the native silica particles and vinyltriethoxysilane-modified silica particles, respectively. The vinyltriethoxysilane-modified silica sample displays a lower decomposition temperature than that of the native silica sample. The difference in the weight loss between the native silica particles and vinyltriethoxysilane-modified silica particles is taken as the level of vinyltriethoxysilane that has been incorporated into the silica nanoparticles (~11%). Assuming that all the triethoxy groups participate in the hydrolysis and condensation reactions, the vinyl ($-CH{=}CH_2$) content that will be thermally decomposed at 800°C is estimated to be about $[2 \times (27/190)/(2 + 1)] \times 100\% = 9.5\%$, which is quite close to the TGA data. In the above calculation, the values of 2 and 1 represent the weights of vinyltriethoxysilane and silica used in the recipe, respectively, and those of 190 and 27 represent the molecular weights of vinyltriethoxysilane and $-CH{=}CH_2$, respectively.

Figure 2(b) shows that the weight loss of the neat BMI oligomer is 44.0% when the temperature increases from ambient temperature to 800°C. Exactly the same procedure described in the section of Experimental Methods was used to prepare the neat BMI oligomer except that no native silica particles or vinyltriethoxysilane-modified silica particles were present in the polymerization system. It is noteworthy that there might be some residual BMI monomer and BTA present in the neat BMI oligomer sample,[14] thereby contributing to the reduced thermal stability as well. The TGA data demonstrate the very good thermal stability of the neat BMI oligomer. The ultimate weight losses of the BMI/silica and BMI/silane/silica composite particles with 20 wt % silica based on total solids can be further reduced to 36.0% and 39.7%, respectively. This is simply because the level of the organic components in decreasing order is the neat BMI oligomer > BMI/silane/silica > BMI/silica. Similarly, the ultimate weight losses of the neat BMI oligomer and BMI/silane/silica composite particles with 10–40 wt % silica based on total solids in decreasing order are the neat BMI oligomer (44.0%)> BMI/silane/silica with 10% silica based on total solids (42.0%) > BMI/silane/silica with 20% silica based on total solids (39.7%) > BMI/silane/silica with 30% silica based on total solids (36.8%) > BMI/silane/silica with 40% silica based on total solids (33.8%). The numeric values shown in parentheses represent the corresponding ultimate weight losses of the samples. As expected, the higher the level of silica, the better the thermal stability of the BMI/silane/silica composite particles.

Morphological studies

The appearance of both the BMI/silica and BMI/silane/silica composite particles dispersed in γ-butyrolactone (total solids content = 20 wt %) are transparent (brown or dark brown) (Table I). It should be noted that the BMI/silica particles with the weight percentage of silica based on total solids equal to 30 or 40 wt % completely failed to disperse in γ-butyrolactone, most likely due to the exceedingly solvent-absorbing capability of the native porous silica. However, the BMI/silane/silica colloidal systems with the weight percentage of silica based on total solids greater than 20 wt % still show excellent fluidity as compared with the BMI/silica counterparts. Furthermore, the hydrodynamic average particle sizes of both types of the nonaqueous dispersions could not be determined by the dynamic light-scattering technique (DLS, Otsuka, Photal LPA-3000/3100) because of the very weak light scattering intensity. These observations indicate that the colloidal particles are not large enough to effectively scatter the incident laser light used in this study.

Figure 3 shows FESEMs of (a) the native silica particles and (b) the vinyltriethoxysilane-modified silica particles [silica/silane = 1/2 (w/w)] dispersed in γ-butyrolactone (total solids content = 20 wt %). The distinct primary silica nanoparticles within the secondary aggregated structure are observed. The average primary particle size is estimated to be 30 and 40 nm in diameter for the native and silane-modified silica particles, respectively. The slightly larger primary particle size for the silane-modified silica particles is most likely due to the grafted silane molecules around the silica particles. Figure 4 shows FESEMs of (a) the BMI/silica particles (20 wt % silica based on total solids) and (b–d) the BMI/silane/silica particles with the weight percentage of silica based on total solids equal to 20, 30, and 40 wt %, respectively. The primary silica nanoparticles are still distinguishable for the extremely fine BMI/silane/silica composite particles with the weight percentage of silica based on total solids equal to 20 wt %, in contrast to the counterpart in the absence of the coupling agent A-151 characterized by a rather coarse grain-like structure [Fig. 4(a,b)]. This result illustrates the important role of vinyltriethoxysilane in preparing such MBI/silane/silica hybrid materials; the vinyltriethoxysilane-modified silica nanoparticles significantly improve their dispersion capability within the continuous BMI oligomer matrix. Furthermore, the degree of dispersion of the vinyltriethoxysilane-modified silica particles in the BMI oligomer matrix decreases with the weight percentage of silica based on total solids being increased from 20 to 40 wt % [Fig. 4(b)–(d)].

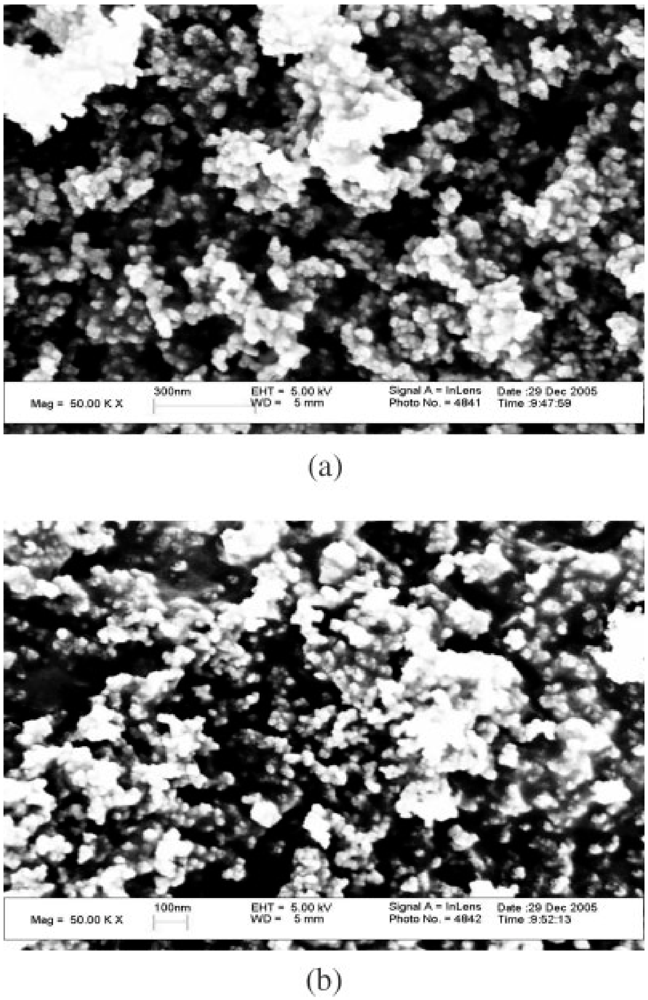


Figure 3 SEMs of (a) native silica particles and (b) vinyltriethoxysilane-modified silica particles [silica/silane = 1/2 (w/w)] dispersed in γ-butyrolactone (total solids content = 20 wt %).

The BMI/silane/silica composite particles with the weight percentage of silica based on total solids equal to 20% and 40% were further examined qualitatively by the EDX spectroscopy. The results thus obtained are summarized in Table II. The decreased ratio of N/Si with the weight percentage of silica based on total solids is consistent with the formulas employed. The decreased ratio of C/Si can be attributed to the scenario that the competitive effect of the concomitantly increased C content originating from vinytriethoxy silane is weaker than that of the increased Si content when the weight percentage of silica based on total solids increases from 20% to 40%. Similarly, the decreased N/C ratio is caused by the dilution effect provided by the concomitant increase of the C content associated with vinytriethoxy silane when the weight percentage of silica based on total solids increases from 20% to 40%.

Rheological studies

Figure 5 shows the viscosity of the native silica particles and vinyltriethoxysilane-modified silica particles dispersed in *N*-methyl-2-pyrrolidone as a function of shear rate. The total solids content is varied from 1 to 6 wt %. Such levels of the silane-modified silica nanoparticles are typical in industrial formulas. It is of vital importance to gain a better understanding of the role of the silane-modified silica nanoparticles in the rheological behaviors in order to optimize the application properties of the related organic-

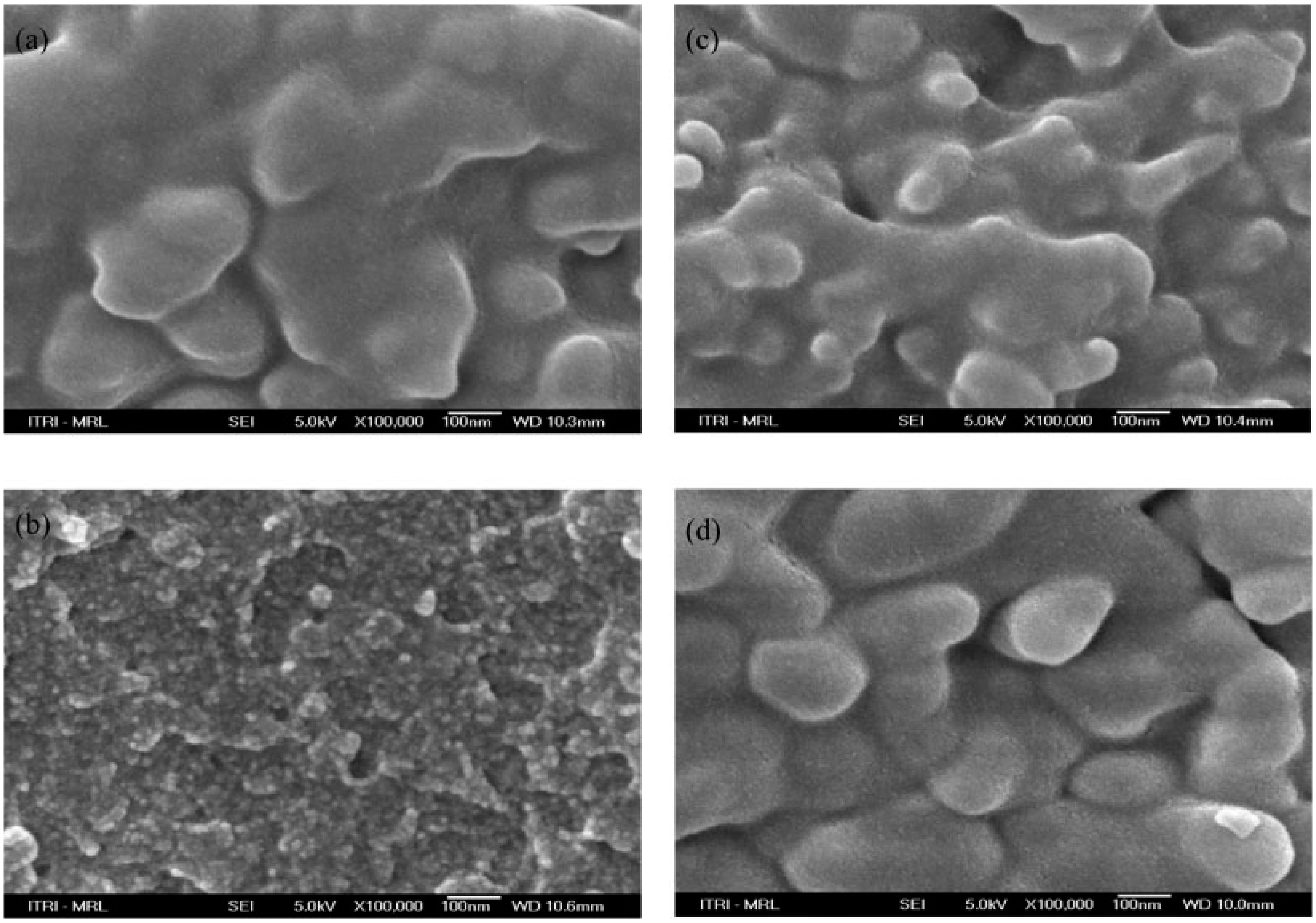


Figure 4 SEMs of (a) BMI/silica composite particles (20 wt % silica based on total solids), (b) BMI/silane/silica composite particles (20 wt % silica based on total solids) (c) BMI/silane/silica composite particles (30 wt % silica based on total solids) and (d) BMI/silane/silica composite particles (40 wt % silica based on total solids) dispersed in γ-butyrolactone (total solids content = 20 wt %).

inorganic hybrid products. Both the native silica and silane-modified silica dispersions show a typical shear-thinning behavior; the viscosity first decreases and then levels off with increasing shear rate. At constant shear rate, the viscosity of the colloidal dispersion increases with increasing total solids content. However, such a crowding effect becomes diminished as the shear rate approaches 10^4 s^{-1} and predominates in the flow behavior of the colloidal system. More importantly, at constant total solids content, the viscosities of the silane-modified silica dispersions are much lower than those of the native silica counterparts. This trend is attributed to the enhanced compatibility between the silane-modified silica nanoparticles and the continuous nonaqueous phase. The solubility parameters of the vinyl group of vinyltriethoxysilane [10.1 $(\text{cal cm}^{-3})^{1/2}$] and *N*-methyl-2-pyrrolidone [11.3 $(\text{cal cm}^{-3})^{1/2}$] are quite close to each other. The numeric values in the parentheses were estimated according to the group contribution method or taken from the literature.[15,16] The closer the two δ values, the stronger the interaction between the pair of molecules. The significantly enhanced compatibility may prevent the silane-modified silica particles from forming a

TABLE II
EDX Results Obtained from BMI/Silane/Silica Particles with Weight Percentage of Silica Based on Total Solids Equal to 20 and 40 wt %

Silica (wt %)[a]	C (%)	N (%)	O (%)	Si (%)	C/Si[b]	N/Si[b]	N/C[b]
20	45.6	5.9	33.1	15.4	2.97	0.39	0.13
40	41.1	3.1	34.3	21.6	1.90	0.14	0.075

[a] Weight percentage of silica based on total solids.
[b] Weight ratio.

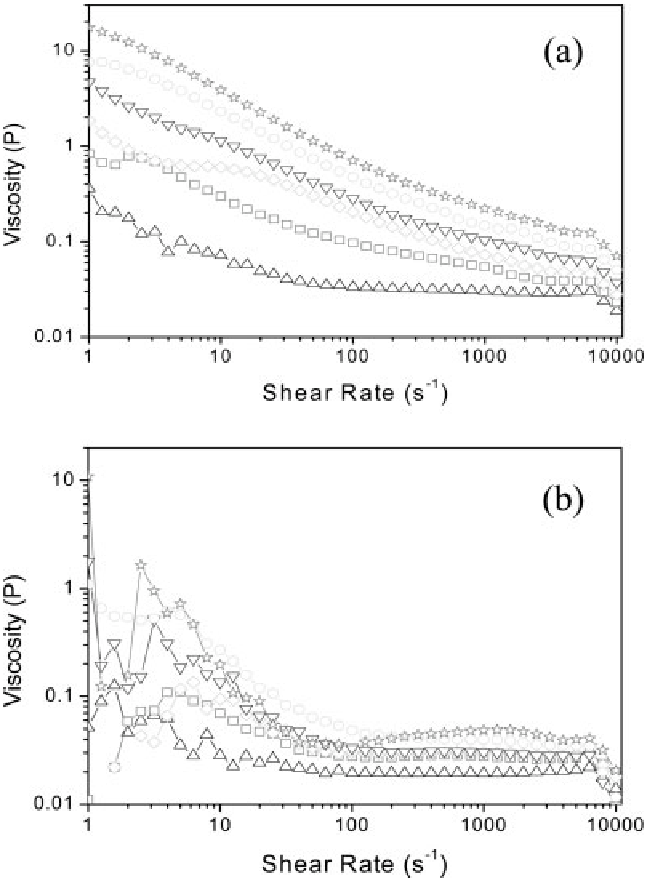


Figure 5 Viscosity of (a) native silica particles and (b) vinyltriethoxysilane-modified silica particles dispersed in *N*-methyl-2-pyrrolidone as a function of shear rate. Total solids content (wt %): (△) 1, (□) 2, (◇) 3, (▽) 4, (○) 5, (☆) 6.

strongly aggregated structure, leading to the much better dispersion fluidity. In addition, the filling of silica particle pores by the chemically grafted vinyltriethoxysilane molecules that greatly retards the solvent absorption process is also responsible for the improved flow properties.

Figure 6 shows the shear stress of the native silica particles and vinyltriethoxysilane-modified silica particles dispersed in *N*-methyl-2-pyrrolidone as a function of shear rate when the rheological measurements were operated in the transient thixotropic loop test mode. This cyclic mode was used to study the dynamic changes in the structures of these nonaqueous silica dispersions. For all the silane-modified silica dispersions with different total solids contents (1–6 wt %), the differences in the shear stress evolution in up-and-down changes of shear rate are much smaller than those of the native silica counterparts. This result suggests that the rates of restoration of the structures of the silane-modified silica colloids during the thixotropic loop experiments are much faster than those of the native silica counterparts. One possible explanation for this observation is that the silica nanoparticles coated with a layer of organosilane species are very compatible with the continuous nonaqueous phase. As a consequence, the structures of the silane-modified silica dispersions are more labile to destruction (i.e., less viscous) in comparison with the native silica counterparts when subjected to the applied shear stress. Furthermore, at constant shear rate, it would take a shorter period of time for the silane-modified silica dispersions to restore the original shear stress than the native silica counterparts. The destruction and restoration mechanisms involved in the structural transformation among the native or silane-modified silica particles are not clear at this point of time. Nevertheless, the rheological data obtained from this work clearly demonstrate the fact that the native and silane-modified silica dispersions exhibit dramatically different flow behaviors. These rheological measurements are in qualitative consistence with the observations described in Table I.

The BMI/silane/silica composite nanoparticles prepared in this work can be added to polymeric materials such as polyimide to improve the performance properties (e.g., mechanical and thermal properties). These unique organic–inorganic hybrid materials may find a variety of industrial applications. This subject will appear in a forthcoming paper.

CONCLUSIONS

The synthesis and characterization of the vinyltriethoxysilane-modified silica nanoparticles [silica/silane = 1/2 (w/w)] were investigated. The FTIR and TGA data indicated that the vinyltriethoxysilane species had been successfully grafted onto the silica nanoparticles. The native and silane-modified silica dispersions with *N*-methyl-2-pyrrolidone as the continuous nonaqueous phase and the total solids contents within the range of 1–6 wt % showed dramatically different rheological behaviors. At constant total solids content, the viscosities of the silane-modified silica dispersions are much lower than those of the native silica counterparts. Furthermore, for all the silane-modified silica dispersions with different total solids contents, the differences in the shear stress evolution in up-and-down changes of shear rate are much smaller than those of the native silica counterparts. It was postulated that the silica nanoparticles coated with a layer of organosilane species were very compatible with the continuous nonaqueous phase. Thus, the structures of the silane-modified silica dispersions are more labile to destruction in comparison with the native silica counterparts when subjected to the applied shear stress. At constant shear rate, it would take a shorter period of time for the silane-modified silica dispersions to restore the original shear stress than the native silica counterparts.

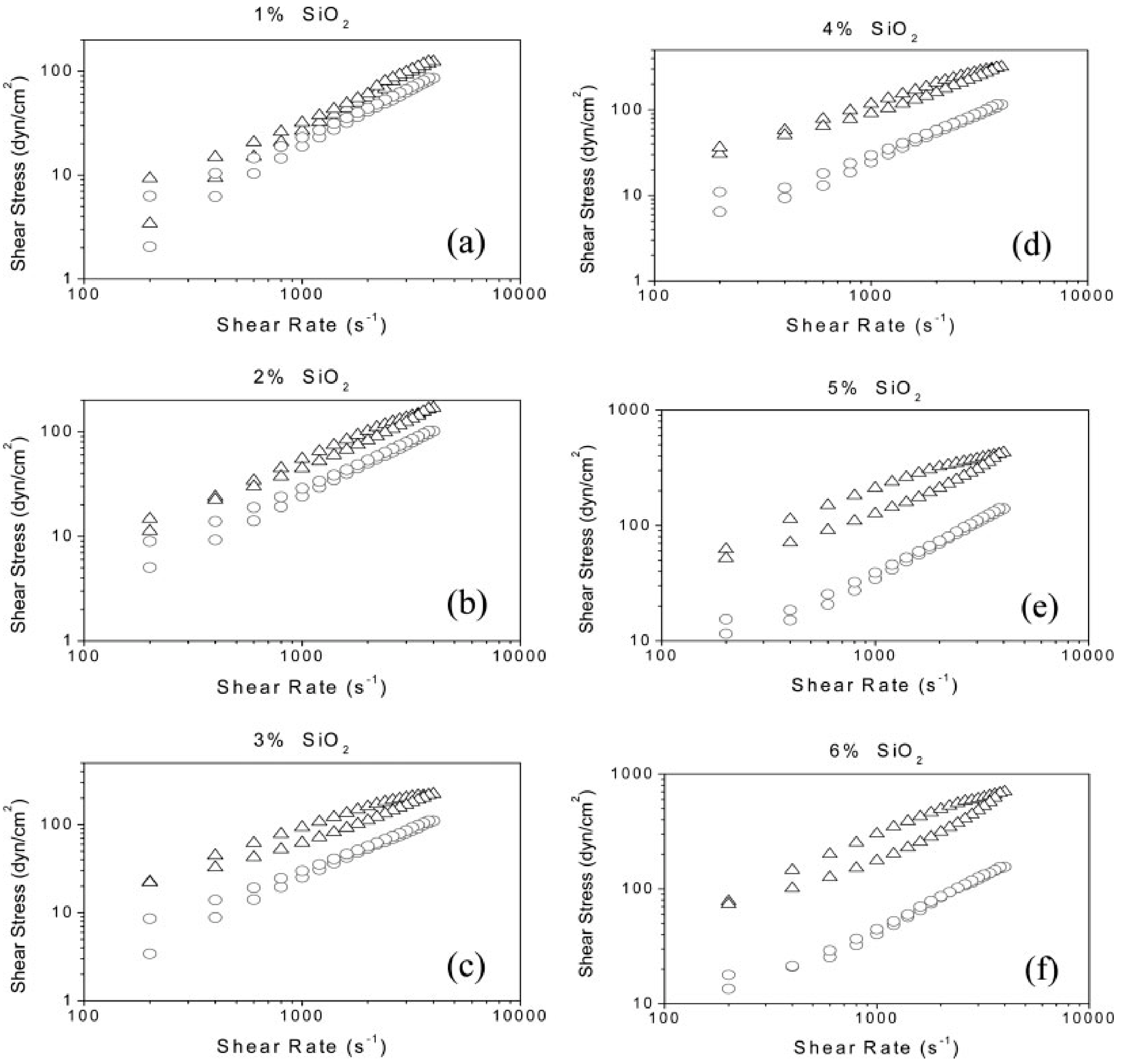


Figure 6 Shear stress of (△) native silica particles and (○) vinyltriethoxysilane-modified silica particles dispersed in *N*-methyl-2-pyrrolidone as a function of shear rate when the rheological measurements were operated in the transient thixotropic loop test mode. Total solids content (wt %): (a) 1, (b) 2, (c) 3, (d) 4, (e) 5, (f) 6.

The polymerization of *N*,*N*′-bismaleimide-4,4′-diphenylmethane (BMI) initiated by barbituric acid in the presence of the native or vinyltriethoxysilane-modified silica nanoparticles were carried out in γ-butyrolactone (total solids content = 20%). The higher the level of silica, the better the thermal stability of the BMI/silane/silica composite particles. The BMI/silica particles with the weight percentage of silica based on total solids equal to 30 or 40 wt % completely failed to disperse in γ-butyrolactone. In contrast, the BMI/silane/silica colloidal systems with the weight percentage of silica based on total solids greater than 20 wt % still showed excellent fluidity. The vinyltriethoxysilane-modified silica particles significantly improved their dispersion capability within the continuous BMI oligomer matrix. Moreover, the degree of dispersion of the vinyltriethoxysilane-modified silica particles in the BMI oligomer matrix decreased with the weight percentage of silica based on total solids being increased from 20 to 40 wt %.

The authors thank V. Chou (Asia Electronic Material Co., Ltd.), J. Yu (Career Technology (MFG.) Co., Ltd.), F. J. Kao (Greencure Technology Co. Ltd.), and E. Shao (Chenchem Co., Ltd.) for their constructive comments on and discussion of the experimental data.

References

1. Vrancken, K. C.; Possemiers, K.; Van Der Voort, P.; Vansant, E. F. Colloids Surf A: Physicochem Eng Aspects 1995, 98, 235.
2. Goerl, U.; Hunsche, A.; Mueller, A.; Koban, H. G. Rubber Chem Technol 1997, 70, 608.
3. Daniels, M. W.; Francis, L. F. J Colloid Interface Sci 1998, 205, 191.
4. Li, Y.; Yu, J.; Guo, Z. X. J Appl Polym Sci 2002, 84, 827.
5. Byers, J. T. Rubber Chem Technol 2002, 75, 527.
6. Chen, H.; Zhou, S.; Gu, G.; Wu, L. J Dispers Sci Technol 2005, 26, 27.
7. Pere, E.; Cardy, H.; Latour, V.; Lacombe, S. J Colloid Interface Sci 2005, 281, 410.
8. Tamaki, R.; Chujo, Y. Chem Mater 1999, 11, 1719.
9. Hsiue, G. H.; Chen, J. K.; Liu, Y. L. J Appl Polym Sci 2000, 76, 1609.
10. Sereda, L.; Gonzalez, M. M. L.; Visconte, L. L. Y.; Nunes, R. C. R.; Furtado, C. R. G.; Riande, E. Polymer 2003, 44, 3085.
11. Liu, Y. L.; Hsu, C. Y.; Wei, W. L.; Jeng, R. J. Polymer 2003, 44, 5159.
12. Yu, Y. Y.; Chen, C. Y.; Chen, W. C. Polymer 2004, 44, 593.
13. Chen, B. K.; Chiu, T. M.; Tsay, S. Y. J Appl Polym Sci 2004, 94, 382.
14. Pan, J. P.; Shiau, G. Y.; Lin, S. S.; Chen, K. M. J Appl Polym Sci 1992, 45, 103.
15. Barton, A. F. M. CRC Handbook of Solubility Parameter and Other Cohesion Parameters; CRC Press: Boca Raton, FL, 1983.
16. Brandrup, J.; Immergut, E. H., Eds. Polymer Handbook, 3rd ed.; Wiley: New York, 1989.