www.amlett.com, DOI: 10.5185/amlett.2010.4119

Published online by the VBRI press in 2010

Effect of ultrasonic on the properties of silicone/ montmorillonite nanocomposites by *in-situ* intercalative polymerization

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Received: 26 April 2010, Revised: 4 May 2010 and Accepted: 8 May 2010

ABSTRACT

Using ultrasonic technology, phenylmethylsilicone/organic montmorillonite (OMMT) nanocomposites were prepared by in-situ intercalative polymerization. The internal structure and morphology of nanocomposites were characterized by X-ray diffraction (XRD), scanning electron microscope (SEM) and transmission electron microscope (TEM) analyses. The basal spacing of OMMT increases with the increasing of ultrasonic time. Intercalated and exfoliated phenylmethylsilicone/OMMT nanocomposites were successfully prepared by convenient process of changing ultrasonic time for 15 min and 30 min, respectively. The barrier property for water was obviously improved by adding OMMT into silicone. The water absorption rate of intercalated nanocomposite containing 4wt% OMMT is 0.7687%, which is 10 times less than that of pure silicone. However, the water absorption rate of exfoliated nanocomposite is 2.802%, which is 3 times less than that of pure silicone. The nanocomposite containing 6wt% OMMT has the best mechanical performance. The impact strength of intercalated nanocomposite is 1.593 2 kJ/m², which is enhanced 7.45% more than that of pure silicone, and the impact strength of exfoliated nanocomposite is improved 14.4% to 1.695 7 kJ/m². The tensile strength of intercalated nanocomposite was markedly enhanced from 1.247 MPa of pure silicone to 7.021 MPa and to 6.264 MPa for exfoliated nanocomposite. When nanocomposite contains 4wt% OMMT, the compressive strength of pure silicone is also evidently improved from 2.834 MPa to 10.232 MPa of intercalated nanocomposite and to 11.503 MPa of exfoliated nanocomposite, respectively. The hardness of exfoliated and intercalated PLS nanocomposites are both firstly increased with the increasing of OMMT content, and then decreased, and finally increased again. Copyright © 2010 VBRI press.

Keywords: Phenylmethylsilicone; montmorillonite; in-situ intercalative polymerization; ultrasonic; intercalated nanocomposite; exfoliated nanocomposite



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Introduction

During the last 10 years, preparation and characterization of organic-inorganic nanocomposites have attracted a great deal of attention in material science [1-5]. Nanocomposites offer the potential for the diversification of applications for polymers due to their excellent properties, such as high temperature resistance, dimensional stability, improved retardancy and enhanced barrier property, flame **[4–8**]. thermomechanical property Polymer/MMT nanocomposites have been successfully prepared by intercalating polymers into MMT. According to the starting materials and process techniques, the preparation methods of polymer/MMT nanocomposites are divided into three main groups: solution intercalation of polymer or prepolymer, in-situ intercalative polymerization and melt intercalation [9]. Depending on the degree of penetration of the polymer chains into the interlayer space, intercalated and exfoliated nanocomposites are distinguished [10].

By hydrothermal reaction, Wang prepared exfoliated nylon-6/montmorillonite nanocomposites through meltcompounding [11]. Atomic force microscopy (AFM) shows that the OMMT nanoparticles are dispersed in nanoscale and arranged along the injection direction. Mechanical tests show that, upon incorporation of only 2 wt% OMMT, the impact strength of nylon-6 is greatly improved whereas other properties are only slightly improved [11]. The exfoliation of the organoclay is achieved by adding a small amount of maleic anhydride grafted polyolefin, which is but with poor solvent resistance and mechanical properties. Intercalating silicone into MMT to obtain polymer/clay nanocomposites can improve these performances. Several studies have reported about silicone rubber/MMT nanocomposites [13-15]. However, there are few studies about silicone/clay nanocomposite and few literatures about the effect of ultrasonic on the performance of nanocomposites.

In the present research, ultrasonic technology was used to prepare phenylmethylsilicone/OMMT nanocomposites by in-situ intercalative polymerization. The internal structure and morphology of the nanocomposites were characterized by XRD, SEM and TEM analyses. The effect of time for ultrasonic processing on the dispersing of MMT lamellar was also studied. It is interesting that intercalated and exfoliated phenylmethylsilicone/OMMT nanocomposites were prepared by simple process of changing ultrasonic time of 15 min and 30 min, respectively. The barrier performance for water and mechanical properties of intercalated and exfoliated nanocomposites were also studied.

Experimental

Materials

Phenylmethylsilicone (UN6302) was purchased from Shanghai United Chemicals & New Materials Co. Ltd.



Scheme 1. Schematic diagram of curing reaction for phenylmethylsilicone in the layer of OMMT.

miscible with the polyolefin [12]. Silicone has many excellent properties, such as high temperature resistance, weatherability, hydrophobic property, electrical insulation,

(Shanghai, China). Sodium montmorillonite (Na-MMT) was provided by Zhejiang Fenghong Clay Chemicals Co. Ltd. (Zhejiang, China). Triethanolamine, methyl

methacrylate (MMA), benzoperoxide (BPO) and toluene were purchased from Sinopharm Chemical Reagent Co. Ltd. (Beijing, China) and were used without further purification. The organic treatment agent of cetyltrimethylammonium bromide (CTAB) was obtained from Jining Chemical Institute (Jining, China).

Preparation of silicone/OMMT nanocomposites

The organic MMT was prepared with organic treatment agent of CTAB according to previous literatures [1, 9]. In a two-neck (250 mL) flask equipped with a stirrer, nanocomposites were prepared by swelling desired concentration of OMMT, MMA with toluene and prepolymer of phenylmethylsilicone, and then with mechanical stirring for 0.5 h at 75 °C through water bath heating. In order to obtain the effect of time for ultrasonic processing on the dispersing of MMT lamellar, ultrasonic time of zero min (only with mechanical stirring), 10 min, 20 min and 30 min were selected. The reaction system was taken into water bath again and stirred for 1.5 h at 75 $^\circ C$ after suitable amount of BPO was added. BPO can induce MMA polymerization. After drying in a vacuum oven at 60 $^{\circ}$ C for 1.5 h, the mixture was heated to 95 $^{\circ}$ C with continuous stirring. Triethanolamine, the curing agent, was added and the reaction system was stirred for an additional 5 min. The curing mechanism was explained in Scheme 1. The mixture was poured into a home-made open round mold, followed by curing process in a vacuum oven at 90 °C for 36 h. Finally, the polymer/OMMT (polymer/layered silicate, PLS) nanocomposites were obtained.

Measurements and characterizations

Morphological analysis: Small-angle X-ray diffraction (SXRD) measurement was carried out using D/max 2500VB3+/PC (Rigaku, Japan) equipped with a Nifiltered monochromatic Cu-Ka radiation ($\lambda = 1.541$ 8Å) at a scanning rate of 1°/min and divergence slit 1°. Measurements were made to examine the interlayer activity in the composite as prepared. The plates of silicone/OMMT nanocomposites which were produced during the molding process have smooth surface. Therefore, a rectangle shaped specimen (10 mm \times 5 mm \times 1 mm) was cut from these plates and directly analyzed by X-ray analysis. Bragg's Law $(n\lambda = 2d \sin\theta)$ was used to calculate the crystallographic spacing. A JEOL JSM-6480 (Japan) scanning electron microscope was used to observe the particle size of MMT and particle-matrix adhesion in the cured composite on the fracture surface. The specimens were coated by gold. TEM analysis was used to confirm the morphological information obtained from the XRD data on the platelet dispersion and distribution. The morphology can not be fully characterized only by XRD analysis. The absence of scattered intensity peaks in the XRD diagrams can not always surely demonstrate achievement of the disordered intercalated or the exfoliated structures. The ultra-thin TEM specimens from the powder of specimen were immersed in the alcohol and dispersed by ultrasonic. The solution was dripped on copper grids and was viewed with a JEOL JEM-2100 electron microscope at an accelerating voltage of 200 kV.

Measurements of barrier property for water: To determine barrier property of water, rectangle shaped specimens (20 mm \times 8 mm \times 2 mm) of silicone/OMMT nanocomposites were prepared. The specimens were heated in deionized water at 80 °C for 0.5 h. Water adsorption rate (*a*) was used to denote barrier property of water and defined as follows:

$$a = \frac{m_2 - m_1}{m_1} \times 100\%$$

where m_1 (g) and m_2 (g) represent the weight of specimen before and after boiled, respectively.

Mechanical properties: The tensile test was conducted on WDS electronic tensile testing machine (Chengde Precision Testing Machine Co. Ltd., China) at room temperature. The test specimens were cut in the shape $100 \text{ mm} \times 10 \text{ mm} \times 4$ mm. The tensile test speed was set for 5 mm/min. The compression test was taken on WDW-11 computercontrolled electronic universal testing machine (Jinan Shidai Shijin Instrument Co. Ltd., China). All compression specimens were shaped for 30 mm \times 10 mm \times 4 mm and the loading rate is 2 mm/min. Unnotched impact strength test was performed on XJ-50Z Combined Impact testing machine (Chengde Precision Testing Machine Co. Ltd., China) and the impact pendulum was selected for 2 J. To reduce experimental error, every tensile specimen, compression specimen and impact specimen of nanocomposites were all tested for five samples and the average value was taken as tensile, compressive and impact strength. Rockwell hardness was performed on XHR-15 Rockwell Hardometer (Shanghai Material Testing Machine Factory, China). The diameter of spherical sclerometer indentor was selected for 6.35 mm and the total loading was 588.4 N. To obtain the hardness value of nanocomposites, at least 7 points were selected for every sample.

Results and discussion

XRD characterization of silicone/OMMT nanocomposites

The basal spacing of OMMT and PLS nanocomposites with different ultrasonic time were showed in **Fig. 1**. The calculated crystallographic spacings according to Bragg's Law ($n\lambda = 2d \sin\theta$) were listed in **Table 1**. The diffraction angle (θ) decreases with the increasing of ultrasonic time, resulting in the increasing of crystallographic spacing (d_{001}) from 3.6 nm to 8 nm. This phenomenon indicates that ultrasonic can help monomer or prepolymer effectively disperse into montmorillonite layers. In aqueous solution, through ultrasonic technology, the micro-mixing at molecular level can be obtained and micron or even nanometer scale particles can be formed.

Compared to mechanical stirring, ultrasonic technology is more effective to produce nanometer scale particles during swelling procedure. Due to the strong shock caused by ultrasound wave at solid-liquid interface, transient cavity and asymmetry collapse are formed to produce micro-jet which impacts solid surface to accelerate mass transfer and to make the solid surface active, leading to quick reaction [16]. The mass transfer and diffusion are accelerated because of the intensive mass motion caused by

ultrasonic, which makes the mass stripped from the surface and the surface is updated [17]. Hence, the basal spacing of MMT can be expanded even in short time. During a certain sound radiation period, the whole sonochemical yield increases linearly with the irradiation time [18], therefore, the basal spacing of MMT increases with the ultrasonic. It can be concluded that the ultrasonic time can markedly affect the increasing extent of basal spacing of MMT.



Fig. 1. XRD spectrum of OMMT and PLSN with different ultrasonic time A: OMMT; B: Mechanical stir; C: Mechanical stir +10 min ultrasonic; D: Mechanical stir +20 min ultrasonic; E: Mechanical stir +30 min ultrasonic (All samples OMMT 6 wt%).

Adv. Mat. Lett. 2010, 1(2), 135-142

Table	1.	Basal	spacing	of	OMMT	and	PLSN	with	different	ultrasonic
time.										

Intercalation method	$2\theta/^{\circ}$	<i>d</i> ₀₀₁ /nm
OMMT	3.60	2.4
Mechanical stir for 30 min	2.44	3.6
Mechanical stir +10 min	2.40	3.7
ultrasonic		
Mechanical stir +20 min	2.30	3.8
ultrasonic		
Mechanical stir +30 min	No obvious	>8
ultrasonic	peak	

SEM and TEM spectra of intercalated and exfoliated PLS nanocomposites

PLS To prepare intercalated and exfoliated nanocomposites, 15 min and 30 min ultrasonic time were selected, respectively. According to Bragg's Law, the basal spacing for 15 min ultrasonic process sample was calculated about 3.74 nm since 2θ equals to 2.38° in SXRD spectrum. This indicates that the lamellar of MMT has not been exfoliated, forming intercalated nanocomposite. There is no diffraction peak at low angle region of SXRD spectrum for 30 min ultrasonic process sample, which means that the *d*-spacing of OMMT within this type of nanocomposite is at least over 8 nm. This illuminates that molecules of silicone chains have intercalated into the layers of MMT, and the interlayer distance of MMT is expanded effectively after polymerization of MMA monomer and phenylmethylsilicone prepolymer, even forming exfoliated nanocomposite. To distinguish the inner intercalated exfoliated structure of and PLS nanocomposites, SEM and TEM spectra were given in Fig. 2 and Fig. 3. The fracture surfaces of silicone/OMMT (6 wt%) nanocomposites were observed by SEM spectrum. MMT was characterized by morphological hierarchy comprising lamellar assembly into primary particles which amass into aggregates. Since nanostructure can not be simply deduced from change of d-spacing measurement which is used to detect lamellar activity, the fracture surfaces of silicone/OMMT (6 wt%) nanocomposites were also observed by SEM spectrum. Searching for mineral domains at the fracture surface of nanocomposite (Fig. 2) reveals the absence of large mineral aggregates. This suggests that the mineral domains are submicron in size. It can be concluded that OMMT and silicone are compatible very well, which also means that each lamella of MMT has been dispersed well in polymer matrix. However, some pores with about 10 µm diameter can be seen in SEM image. These pores may be due to the solvent evaporation and will affect mechanical performance of nanocomposites.

The most powerful and direct evidence to describe whether the nanocomposite is exfoliated or intercalated is by TEM. TEM spectra of thin films of exfoliated and Advanced Materials *Letters*

intercalated OMMT/silicone nanocomposites were shown in **Fig. 3**. The overall picture shows that MMT layers have not occupied the full volume and large regions of pure silicone are visible. The molecules of silicone chains have inserted into the interlayer of MMT, forming light stripes alternated with dark stripes. TEM spectrum of a thin film of OMMT/silicone nanocomposite which stirred for another 15 min under ultrasonic condition is shown in **Fig. 3A** and **B** (A: 6 wt% OMMT; B: 8 wt% OMMT). The layers are entirely orderly intercalative structure in the whole region with a short-range ordered range. This structure is similar to polymer brushes, and the nano-space between the lamellar of MMT has a restrictive effect for the movement of polymer chains, forming intercalated nanocomposite.



Fig. 2. SEM spectra of PLS nanocomposite with different magnification, A,B: mechanical stirring +15 min ultrasonic; C, D: mechanical stirring +30 min ultrasonic % wt% OMMT).

TEM spectrum of a thin film of OMMT/silicone nanocomposite which stirred for another 30 min under ultrasonic condition is shown in Fig. 3C and D (C: 6wt% OMMT; D: 8wt % OMMT). In Fig. 3D, a small amount of very dark area can be seen clearly. The reason may be that MMT was added too much, leading MMT to disperse unevenly and agglomerate to some large particles. The data illuminates that the absence of 001 reflection may be caused by a wider distribution of layer spacing. The layers are not entirely orderly intercalative structure in the whole region, but basically orderly and partially exfoliated exists morphology. Most organo-MMT of in nanocomposite with bending status, which illuminates that montmorillonite expresses somewhat flexible under external force and can bear some deformation.



Fig. 3. TEM spectra of PLSN with different ultrasonic time, A, B: mechanical stirring + 15 min ultrasonic; C, D: mechanical stirring + for 30 min ultrasonic; A, C: 6 wt% OMMT; B, D: 8 wt% OMMT.

Barrier property for water

Compared with pure polymer or mechanical blending composites, PLS nanocomposite has better barrier properties for gas and liquid. This may be due to that the silicate layers have a large aspect ratio. When these layers of MMT disperse well in polymer matrix, the diffusion movement of gas or liquid molecules has to bypass these silicate layers. Therefore, the increasing of efficient paths improves the barrier properties for gas and liquid. The permeability coefficient can be simply calculated as follows [19].

where, P_{PLSN} is permeability coefficient of nanocomposite, P_{matrix} is permeability coefficient of polymer; α is OMMT content, Φ is aspect ratio of MMT. The water absorption rate basically decreases with the increasing of P_{PLSN} .

The relationship curve between water adsorption rate and OMMT content is described in Fig. 4. For both intercalated and exfoliated nanocomposites, the water absorption rate basically decreases with the increasing of OMMT content, which indicates that the barrier property for water of silicone/OMMT nanocomposite is increased. This conclusion is coincident with Formula (1). The silicone/ OMMT nanocomposites in the study were prepared by in-situ solution intercalative polymerization. Therefore, solvent evaporation may leave some pores in nanocomposites which were presented in SEM images of Fig. 2. These pores will obviously affect the barrier property of nanocomposites, so that the curve of the water absorption rate vs. OMMT content is not linear which should be linear according to Formula (1). When the silicone/OMMT nanocomposite contains over 4 wt% OMMT, the barrier property for water almost does not change anymore. This may be due to that only intercalation or exfoliated nano-film layer can play a role to barrier

property for water, and micron or submicron particles have no barrier efficiency.

The water absorption rate of pure silicone is 8.403%, and that of intercalated nanocomposite containing 4wt% OMMT is 0.7687%, which is 10 times less than that of pure silicone. However, the water absorption rate of exfoliated nanocomposite is 2.802%, which is 3 times less than that of pure silicone. This indicates that adding of OMMT into polymer can markedly improve the barrier performance of silicone. It is obviously that the barrier property of intercalated PLS nanocomposite is much better than that of exfoliated PLS nanocomposite, since the water absorption of intercalated PLS nanocomposite is only about one third of exfoliated PLS nanocomposite's for the same OMMT content. Known from the TEM spectra in Fig. 3, the MMT layers exist in the form of short-range order in intercalated nanocomposite, while in exfoliated nanocomposite the layers disperse disorderly in silicone matrix. Hence, the barrier performance of intercalated PLS is better than that of exfoliated PLS nanocomposite.



Fig. 4. Effect of OMMT content on the barrier properties for water.



Fig. 5. Effect of OMMT content on impact strength of PLS nanocomposite.

Mechanical properties

Impact property: The impact strength of intercalated and exfoliated PLS nanocomposites are showed in Fig. 5. The impact tests were carried out to examine the influence of organo-clay inclusion and the degree of organo-clay distribution on mechanical properties of nanocomposites. The impact strength increases with the increasing of OMMT content, and then decreases with the further increasing of OMMT content for both intercalated and exfoliated PLS nanocomposites. The impact strength of exfoliated PLS nanocomposite is higher than that of intercalated PLS nanocomposite. The intercalated and exfoliated nanocomposites which contain 6 wt% OMMT have highest impact strength of 1.5932 kJ/m² and 1.6957 kJ/m², respectively. Compared to the impact strength of pure silicon of 1.4827 kJ/m², the impact strength of intercalated and exfoliated PLS nanocomposites are increased 7.45% and 14.4%, respectively. The structure of MMT is destroyed and dispersed uniformly in silicone matrix. The silicone chains which insert into MMT are strongly restricted by the nano-space among the layers of layered silicate. The layer silicates play a role of physical association and make silicone chains connect with each other, resulting in the increasing of crosslinking point number and density of PLS nanocomposite. Hence, the impact strength of nanocomposite is improved by adding OMMT. However, the impact strength begins to decrease when the OMMT content is over 6wt%. This may be due to the number enhancement of primary particles and the inhomogeneous dispersion of OMMT as OMMT content reaches a certain value, leading to the increasing of defects number. The stress concentration can be easily caused by these defects when PLS nanocomposites endure external force, leading to the decreasing of impact strength. On the other hand, single layer of MMT is easier to be dispersed in polymer matrix than multi-layers of MMT aggregate. Hence, the reinforced effectivity of OMMT on exfoliated PLS nanocomposite is better than that on intercalated PLS nanocomposite.



Fig. 6. Effect of OMMT content on Rockwell hardness of PLS nanocomposites.

Hardness performance: The effect of OMMT content on the hardness of PLS nanocomposites is showed in Fig. 6.

The hardness of both exfoliated and intercalated PLS nanocomposites firstly increases with the increasing of OMMT content, then decreases, and finally increases again. A small amount of MMT particles (<2 wt%) can be dispersed well in silicone matrix. The ability of resisting external mechanical stress is improved since MMT particles play a role of physical association and make silicone macromolecular chains wind with each other. With the further increasing of OMMT content, the particles of MMT play a role of improving ductility, the hardness decreases a little. The hardness of both exfoliated and intercalated PLS nanocomposites slowly increases again when the OMMT content is increased continuously. The silicone chains which insert into MMT are strongly restricted by nano-space between the layers of layered silicate, resulting in the enhancement of winding density of silicone macromolecular chains. Hence, the free movement and secondary relaxation of silicon main chains and branched chains are inhibited, leading to the hardness increasing. When the nanocomposite contains 10 wt% OMMT, the hardness of intercalated and exfoliated nanocomposites are 95.7 and 92.4, respectively, which are 1.5 times more than that of pure silicone.

It seems that there is little difference between the hardness of intercalated and exfoliated PLS nanocomposites. The hardness of exfoliated PLS is higher than that of intercalated PLS nanocomposite when OMMT content is less than 5 wt%. However, when OMMT content is over 5 wt%, the situation is the opposite. A small amount of MMT addition plays a role of physical association. In this situation, most MMT layers exist in the form of single layer in exfoliated nanocomposite, while the layers are entirely orderly intercalative structure in the whole region and with a short-range order in intercalated nanocomposite. So the number of particles in exfoliated PLS is more than that in intercalated PLS, and the hardness of exfoliated PLS is higher than that of intercalated PLS when OMMT content is less than 5 wt%. While OMMT content increases over 5 wt%, the silicone chains which insert into MMT are strongly restricted by nano-space between layers of layered silicate, which is contributed to the hardness. The density of nano-space layers of intercalated PLS is much higher than that of exfoliated PLS, resulting in higher hardness of intercalated PLS when OMMT content is over 5 wt%.

Tensile and compressive properties: The effects of OMMT content on tensile strength and compressive strength of nanocomposites are described in Table 2. The presence of organo-clay observably improves the tensile strength and compressive strength for all PLS nanocomposite samples even with a small amount of clay addition. This improvement is directly attributed to the reinforcement provided by the dispersed silicate nanolayers. The exfoliated and intercalated nanocomposites, containing 6 wt% OMMT, have the highest tensile strength of 6.264 and 7.021 MPa, respectively. Both of them are 5 times higher than that of pure silicone. When the nanocomposite contains 4 wt% or 6 wt% OMMT, the maximal compressive strength of exfoliated and intercalated PLS nanocomposites are 11.503 and 10.232 MPa, respectively. The maximal compressive strength of the exfoliated and the intercalated PLS nanocomposites are both 4 times bigger

than that of pure silicone. The tensile and the compressive strength decrease with the increasing of OMMT content when OMMT content reaches 6 wt%. In silicone matrix, OMMT primary particles and intercalated particles exist at the same time. The silicone chains which intercalated into OMMT layers are limited by dipole interaction and nanospace restriction of OMMT layers, which results in the enhancement of tensile and compressive strength, and the decreasing of breaking elongation. The sheet OMMT particles may slip by external force along the force-oriented direction, which can play a similar role of plasticizer to improve mechanical properties of nanocomposites. The decreasing of mechanical properties may be due to the number enhancement of primary particles and the inhomogeneous dispersion of OMMT as OMMT content increases, and leads to the increasing of defects number. These defects are prone to result in stress concentration and lead to the decreasing of mechanical properties.

Table 2. Mechanical properties of PLS nanocompositeswith different OMMT content.

OMMT/%wt			2	4	6	8	10
	Flexural Strength/MPa	1.247	4.62 8	5.216	7.021	3.91 5	2.63 2
Intercalated	Elongation ratio /%	2.65	1.95	1.89	1.71	1.62	1.25
	Compressive strength /MPa	2.834	5.23 5	10.23 2	8.423	8.06 8	7.23 7
	Flexural Strength /MPa	1.247	1.84 6	5.446	6.264	3.75 1	3.17 1
Exfoliated	Elongation ratio /%	2.65	2.24	2.10	1.85	1.72	1.51
	Compressive strength	2.834	3.78	11.50	10.76	9.36	5.65
	/MPa	2.854	6	3	7	2	2

Conclusion

Phenylmethylsilicone/OMMT nanocomposites were prepared by in-situ intercalative polymerization. The effect of ultrasonic time during dispersion prepolymer or monomer into the layers of OMMT on the properties of PLS nanocomposites was investigated. Morphological structure of PLS nanocomposites was characterized by XRD, SEM and TEM analyses. When the ultrasonic time is less than 20 minutes, the OMMT lamellars exist orderly in PLS nanocomposite, resulting intercalated in nanocomposite. However, when the ultrasonic time is over 30 minutes, the OMMT lamellars exist disorderly in nanocomposite and most of layers were dispersed in nanosize, leading to the formation of PLS exfoliated nanocomposite. It is very convenient to prepare intercalated phenylmethylsilicone/OMMT and exfoliated nanocomposites just by changing ultrasonic time for 15 min and 30 min, respectively. The mechanical properties of intercalated and exfoliated PLS nanocomposites were compared by impact strength, hardness, tensile and compressive strength. Compared with pure silicone, the tensile and compressive strength of intercalated and exfoliated nanocomposites are markedly improved when PLS nanocomposites contain 6wt% OMMT. In conclusion, the mechanical properties of exfoliated PLS nanocomposite are better than those of intercalated PLS nanocomposites.

However, the situation is the opposite for barrier performance of PLS nanocomposite.

Acknowledgements

The work is supported by National Natural Science Foundation of China (20207003, 20704019), Major Program from the Science & Technology Council of Zhejiang Province and Jiangsu University Foundation for Excellent Young Teacher. The authors wish to express their appreciation to the Analytical Center at Jiangsu University for the analyses of SEM, XRD and TEM.

References

- Akat, H.; Tasdelen, M.A.; Prez, F.D.; Yagci, Y. European Polymer Journal 2008, 44, 1949.
- Kohno, Y.; Kinoshita, R.; Ikoma, S.; Yoda, K. Applied Clay Science 2009, 42, 519.
- Africa, Y.R.; Pedro, A.L.; Carolina, C.; Alejandro, B.R.N. Applied Clay Science 2009, 43, 91.
- 4. Salahuddin, N.A. Polym. Adv. Technol. 2004, 15, 251.
- 5. Ray, S.S.; Yamada, K.; Okamato, M.; Ueda, K. Polymer 2003, 44, 857.
- Fornes, T.D.; Yoon, P.J.; Hunter, D.L.; Keskkula, H.; Paul, D.R. Polymer 2002, 43, 5915.
- 7. Fornes, T.D.; Paul, D.R. Polymer 2003, 44, 4993.
- 8. Alexandre, M.; Dubois, P. Mater. Sci. Eng. 2000, 28, 1.
- 9. Morgan, A.B.; Harris. J.D. Polymer 2004, 45, 8695.
- Homminga, D.; Goderis, B.; Dolbnya, I.; Groeninckx, G. Polymer 2006, 47, 1620.
- 11. Wang, Z.B.; Wang, X.; Li, G.C.; Zhang, ZK. Applied Clay Science 2008, 42, 146.
- 12. Yamamoto, K.; Tanaka, H.; Sakaguchi, M.; Shimada, S. Polymer 2003, 44, 7661.
- 13. Dong, W.F.; Zhang, X.H.; Liu, Y.Q.; Gui, H.; Wang, Q.G.; Gao, J.M. European Polymer Journal **2006**, *42*, 2515.
- Kaneko, M.L.Q.A.; Yoshida, I.V.P. J. Appl. Polym. Sci. 2008, 108, 2587.
- Kim, E.S.; Kim, E.J.; Shim, J.H.; Yoon, J.S. J. Appl. Polym. Sci. 2008, 110, 1263.
- 16. Xia, H.S.; Wang, Q. Polymer Materials Science & Engineering 2001, 17, 1.
- 17. Torres-Sanchez, C.; Corney, Ultrasonics Sonochemistry 2008, 15, 408.
- Van-Iersel, M.M.; Benes, N.E.; Keurentjes, J.T.F. Ultrasonics Sonochemistry 2008, 15, 294.
- Pinnavaia, T.J.; Beall, G.W. In Polymer-clay nanocomposites, John Willy & Sons, Ltd, Chichester, England, 2000.