

# Morphology Prediction of Ternary Polypropylene Composites Containing Elastomer and Calcium Carbonate Nanoparticles Filler

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**ABSTRACT:** In this work, the morphology was studied in ternary composites of polypropylene (PP) with nano-sized calcium carbonate (nano-CaCO<sub>3</sub>) fillers and elastomer inclusions and the thermodynamic consideration was used to analyze the formation of phase structure of the composites. The wetting coefficient ( $\omega_a$ ), interfacial tension ( $\gamma_{AB}$ ), and work of adhesion ( $W_{AB}$ ) were calculated to predict dispersion state of nano-CaCO<sub>3</sub> fillers. A comparison of the prediction and SEM analysis was given. The results show that three types of phase structures were formed: an encapsulation of the filler by elastomer, a separate dispersion of the filler and elastomer, and a particular structure

of the filler at the PP/elastomer interface. The predictions by  $\omega_a$  were all successfully supported-up by SEM analysis and the predictions by  $W_{AB}$  were however trustless. Both  $\gamma_{AB}$  and  $W_{AB}$  can predict a separate dispersion or an encapsulation phase structure, but they were not available for the particular structure of the filler at the PP/elastomer interface.  $\omega_a$  was competent and favored for the prediction of all three types of morphology among the three parameters. © 2006 Wiley Periodicals, Inc. *J Appl Polym Sci* 103: 1578–1584, 2007

**Key words:** polypropylene; ternary composite; prediction; morphology; thermodynamic consideration

## INTRODUCTION

It is well known that polypropylene (PP) is a kind of semicrystallitic polymer material.<sup>1–3</sup> PP becomes a kind of important and general resin that is applied extensively, because of its advantages, such as excellent comprehensive property, cheapness, etc. On the other hand, because the PP has high crystallinity, its toughness is bad and changes worsely at low temperature, and undoubtedly, this restricts the application of PP in many places. Therefore, in past several decades, the toughening of PP always was a popular research topic in plastic modification field.<sup>4</sup> The investigatives found that compounding soft elastomer

is an efficient, simple, economic, and widely-used method to toughen PP. To this day, much perfect elastomer toughening mechanism is developed,<sup>5</sup> however, rubber toughening can not avoid decreasing rigidity and strength of the PP at the time of achieving an ideal toughening effect. To overcome the shortcoming and achieve an optimum balance of impact strength and stiffness, the ternary polymer composites of PP/elastomer/rigid filler were developed. They make use of the characteristics of elastomer-toughening and rigid-filler-reinforcing, and put both advantages together well to carry out excellent comprehensive property.

A number of investigations found that the properties of the ternary composites are determined not only by the composition and the characteristics of components, but also by the phase morphology of composites, and in particular, the dispersion state of additive components. Thus, the researchers always paid attention to the investigations on the formation of phase morphology of such composites, and expect to find a few of intrinsic rules to help precise design and control of phase structure. The investigations showed that, the formation of particular phase structure depends on several factors, such as thermodynamic consideration (the free energy change of the process, adhesion between the phases

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and so on), concentration of components, the melting viscosity of polymers, the blending sequence, processing condition, etc. However, thermodynamic consideration had been confirmed to be a determining factor in many previous studies<sup>6,7</sup> and was usually used to predict phase structure. Using thermodynamic consideration, three types of methods were reported to be used to predict the phase structure: (1) interfacial tension method<sup>6,8,9</sup>; (2) adhesive work method<sup>6,10</sup>; (3) wetting coefficient method,<sup>11</sup> but an applicability comparison and a difference of three types of methods have not been reported.

The objective of the present study was to analyze the formation of the ternary PP composites containing elastomer and calcium carbonate fillers, using thermodynamic consideration. The three types of predict methods of phase morphology were all investigated by contrasting with SEM analysis, which is expected to give an applicability comparison in such ternary composites.

## EXPERIMENTAL

### Materials

The base material for the composites was polypropylene (PP) homopolymer F401 [melt flow rate (MFR) = 3.0 g/10 min at 230°C and 2.16 kg load] supplied by Guangzhou Petrochemical Industrial, China. Two types of elastomers used were poly (butyl acrylate) (PBA) [weight-average molecular weight ( $M_w$ ) = 69,000, polydispersity index (PDI) = 4.23, measured by Waters 991 gel permeation chromatography (GPC) with tetrahydrofuran as the solvent], prepared by irradiation-induced polymerization according to the process stated in the Ref. 12, and ethylene-octene copolymer (POE) Engage 8200 [consisting of 24 wt % octene, MFR = 5.0 g/min at 190°C and 2.16 kg load, density ( $\rho$ ) = 0.87 g/cm<sup>3</sup>] supplied by DuPont Dow Elastomer (USA). The filler used was nanosized calcium carbonate (nano-CaCO<sub>3</sub>) with an average primary particle size of 44 nm and a surface area of 28 m<sup>2</sup>/g, produced by Guangdong Enping Chemical and Engineering, China. Nano-CaCO<sub>3</sub> particles used in this study were in three forms: untreated CaCO<sub>3</sub> (denoted by CaCO<sub>3</sub>), CaCO<sub>3</sub> coated by stearic acid with an activation rate of 95% (denoted by CaCO<sub>3</sub>-stearic), and CaCO<sub>3</sub> modified by grafting PBA onto surface with a percent grafting of 5.4% (denoted by CaCO<sub>3</sub>-g-PBA, prepared according to the method stated in the Ref. 12, the  $M_w$  of grafted PBA = 33,000, the PDI of grafted PBA = 2.18), respectively.

### Compounding

In this study, the ternary composites were prepared by melt-blending in the mixer of a Haake rheocord

300p torque rheometer. The mixing temperature, mixing time, and rotor speed were 190°C, 15 min, and 60 rpm, respectively. To prepare PP/PBA and PP/POE composites, as comparison samples, the PBA and POE droplets in a particular ratio, to achieve required concentration of the elastomer in the PP matrix, were first simply blended with PP droplets in a cup, then, the blends were added into the mixer of Haake together with antioxidant 1010. Subsequently, the mixer chamber was closed, and all materials were further mixed for 15 min. After mixing finished, the compound was taken off and cut into small pieces in a crusher. On the other hand, the ternary composites were prepared using two-step process, in which PBA/CaCO<sub>3</sub> and POE/CaCO<sub>3</sub> composite particles were prepared first. The former was prepared by the *in situ* irradiation-induced polymerization technique described in detail in Ref. 12; the latter was prepared by melt-blending on the open two-roll mill. The melt-blending was performed at 110°C for 10 min. The ratio of elastomer and CaCO<sub>3</sub> was controlled to achieve required concentration of the elastomer and filler in the PP matrix. Subsequently, the PBA/CaCO<sub>3</sub> and POE/CaCO<sub>3</sub> composite particles were melt-mixed with the PP in Haake mixer under the condition like preparing binary composites.

A vertical injection molding machine Y-350 was used for preparing standard bars for mechanical tests. The injection pressure was 40 bar and the temperature of barrel, nozzle, and mold was 220, 210, and 40°C, respectively. Although the mechanical properties of the composites were not investigated in present study, the samples for SEM analysis were cut from the impact bar prepared according to ISO 179-2.

### Characterization of phase structure

In the present study, the phase structure of composites was examined by scanning electron microscopy (SEM). To prepare specimens for SEM, the corresponding impact test bars were fractured using a hammer after immersing in liquid nitrogen for 5 min. The fractured surfaces were then immersed into a special solvent at 50°C for 5 min to increase the contrast between the PP and elastomers by removing the elastomers from the composites. The solvent used for etching elastomer was acetone for PBA and heptane for POE. After being dried in an oven at 60°C for 1 h, the etched surfaces were sputter-coated with platinum/palladium alloy and then examined in a JSM-6330F Field Emission Scanning Electron Microscope.

The surface tension of various CaCO<sub>3</sub> particles was determined by powder contact angle method at room temperature using a Kruss model K12 contact angle meter. In the powder contact angle method,

water and toluene were used as reference liquids. On the other hand, the surface tension of PP, PBA, and POE was examined by pendent drop method at 190°C (melt-mixing temperature), using a Dataphysics model ACA50, fully automatic video-based contact angle measuring system. The value of the surface tension of polymers and CaCO<sub>3</sub> particles was the sum of dispersion and polar components.

## RESULTS AND DISCUSSION

### SEM analysis

In ternary component composites, three types of phase structure are usually formed: (1) a separation dispersion where the elastomer and filler particles are dispersed separately in the polymer matrix<sup>6,7,10,13–15</sup>; (2) an encapsulation microstructure where the filler particles are encapsulated by the elastomer<sup>6,7,10,13–15</sup>; (3) a microstructure mixed of both (1) and (2).<sup>13,14</sup> One of the most viable methods for analyzing phase structure is by using scanning electron microscopy.

SEM photographs of etched cryogenic fracture surface of all specimens are given in Figure 1. It is well known that the solvent can remove the elastomer from the PP matrix, as a result, the dark holes reveal the elastomer droplets that were etched by selective solvent, contrarily, and the bright particles represent CaCO<sub>3</sub> fillers. Figures 1(a) and 1(c) show that two-phase microstructure is clearly viable in the binary systems. The PBA droplets are remarkably elongated and quite different in size, whereas, the POE droplets are nicely dispersed and more uniform in size. The droplet size of PBA is in the range of 0.2–1.0 μm, which is much bigger than that of POE (0.04–0.25 μm). Various phase structures, on the other hand, are formed in the ternary composites. The voids around the filler particles undoubtedly indicate that the PBA is located at that place in Figure 1(b). As a characteristic,<sup>6,7,15</sup> the void structure reveals an encapsulation microstructure in the PP/PBA/CaCO<sub>3</sub>-g-PBA composite, where CaCO<sub>3</sub> fillers are included in PBA phase, moreover, isolated PBA droplets or CaCO<sub>3</sub> particles dispersed in the PP matrix was not apparent, which indicates an almost perfect encapsulation microstructure.

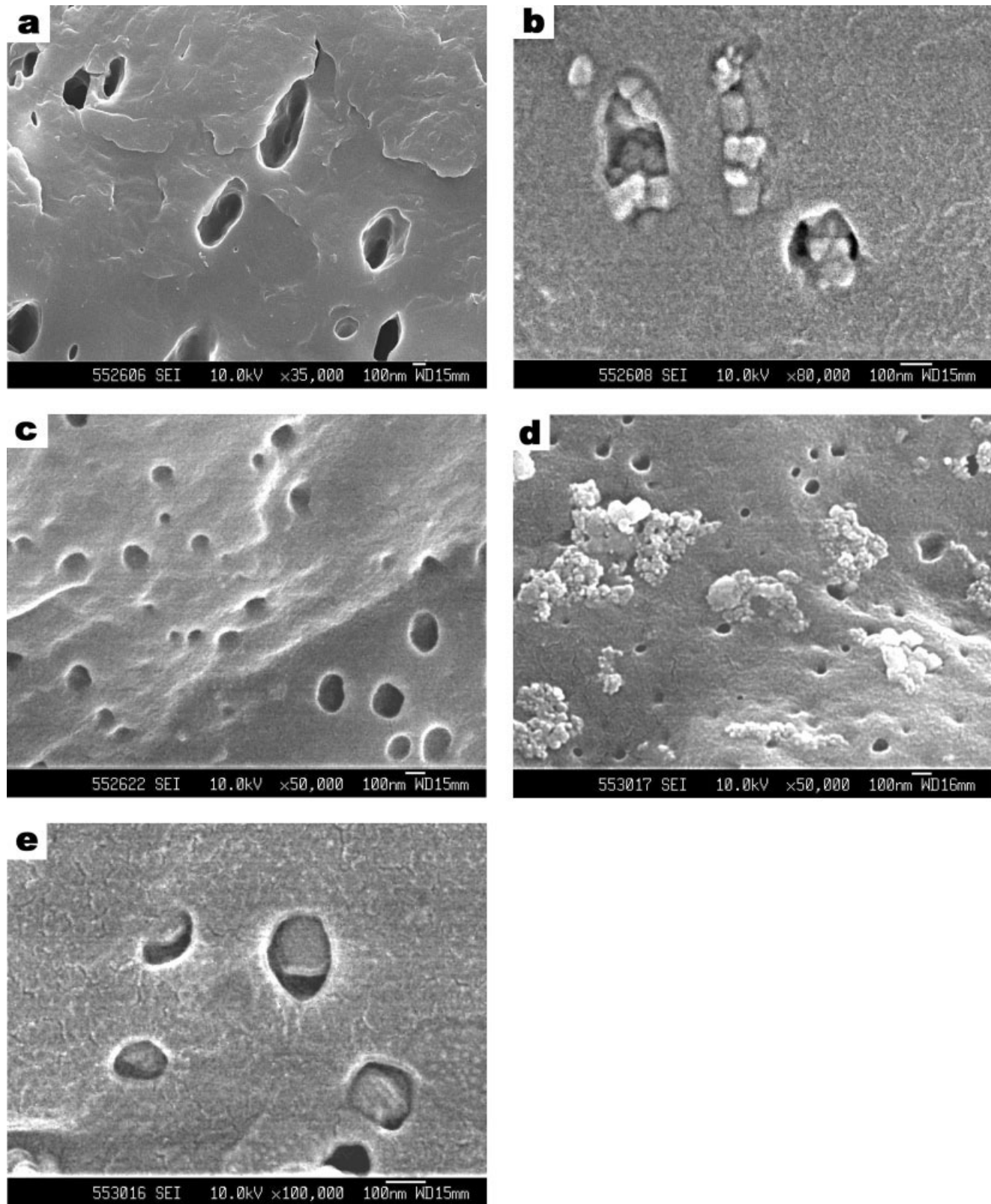
In the ternary composites containing POE, the formation of phase structure depends on whether CaCO<sub>3</sub> particles are surface-modified by stearic acid or not. Figure 1(d) shows that phase structure of PP/POE/CaCO<sub>3</sub> is a separate dispersion microstructure, where the POE and CaCO<sub>3</sub> fillers are dispersed separately in the PP matrix, and that a number of the CaCO<sub>3</sub> particles badly aggregate together because of high surface area and surface energy as nature of nanoparticles. To improve the compatibility between filler and polymer and to

decrease aggregation of filler particles in polymer matrix, the filler is usually surface-modified with organic materials, such as stearic acid, coupling agent, or grafted polymer like PBA grafted onto surface of CaCO<sub>3</sub> in the present study. A particular morphology is shown in Figure 1(e) for the PP/POE/CaCO<sub>3</sub>-stearic composite, which is not a separate dispersion shown in Figure 1(d) or an encapsulation microstructure shown in Figure 1(b). There is not an aggregate of CaCO<sub>3</sub> particles like that shown in Figure 1(b), but an isolated granule that is much larger than CaCO<sub>3</sub> particle in the voids. On the basis of the above evidences, it is concluded that the granule is a POE droplet, and the void around POE granule is the place where the CaCO<sub>3</sub> particles locate. This implies a particular phase structure of the PP/POE/CaCO<sub>3</sub>-stearic composite, where CaCO<sub>3</sub> particles locate at the interface between PP and POE. After etching, the CaCO<sub>3</sub> particles were removed together with interface because the interface can also be etched by heptane.

### Prediction of phase structure

Many investigations<sup>6,7,10,15</sup> on the formation of phase structure in the ternary composites of polymer/elastomer/filler reveal that a main determining factor for phase structure is the thermodynamic characteristic of components, such as surface tension, interfacial tension between the phases, interfacial adhesion between the phases, wetting ability, and so on. Using these thermodynamic considerations, the formation of phase structure can be successfully explained.

In the ternary composites of PP/elastomer/filler, both the phase structure of separate dispersion of components and encapsulation of filler particles by elastomer result in the formation of two new interfaces, PP/elastomer, PP/filler in the former and PP/elastomer, elastomer/filler in the latter. It is well known that, the input of some energy is required to create new surface and interface so that the composites tend to form a phase structure with a minimum total free energy to meet thermodynamic stabilization of the system.<sup>10</sup> The value of the surface free energy is proportional to the size of the new surfaces and to the value of the interfacial tension. Thus, which kind of phase structure would be formed can be concluded by a comparison of interfacial tensions of all possible new interfaces according to the law of minimum total free energy. Premphet et al.<sup>6</sup> investigated the effect of the elastomer polarity on the phase structure of ternary PP/elastomer/CaCO<sub>3</sub> filler and analyzed the formation of the phase structure using a parameter of interfacial tension between the phases. In the PP/EVA/CaCO<sub>3</sub> composite, the calculated interfacial tension of PP/CaCO<sub>3</sub> (113.07 mJ/m<sup>2</sup>) is much great than that of EVA/CaCO<sub>3</sub> (96.8 mJ/m<sup>2</sup>), thus, the formation of EVA/CaCO<sub>3</sub> makes



**Figure 1** SEM photographs of etched fracture surface generated at low temperature for PP composites: (a) PP/PBA, 95/5; (b) PP/PBA/CaCO<sub>3</sub>-g-PBA, 95/5/5; (c) PP/POE, 95/5; (d) PP/POE/CaCO<sub>3</sub>, 95/5/5; (e) PP/POE/CaCO<sub>3</sub>-g-stearic, 95/5/5. Etching details: (a, b) PBA removed by acetone; (c, d, e) POE removed by heptane.

the total free energy of the system minimum and the phase structure of the composite tends to be an encapsulation microstructure, where  $\text{CaCO}_3$  is encapsulated by EVA. The conclusion is consistent with SEM analysis. When the interfacial tensions for PP/filler and elastomer/filler are similar, which implies both PP and elastomer have similar tendency to encapsulate the filler particles, the formation of phase structure changes complicated because of effects from other factors, such as concentration of components, blending sequence and rheology of components, etc. The interfacial tension can be calculated from the surface tension of components using the geometric mean equation of Wu,<sup>16</sup> which is given as follows:

$$\gamma_{AB} = \gamma_A + \gamma_B - 4 \left( \frac{\gamma_A^d \gamma_B^d}{\gamma_A^d + \gamma_B^d} + \frac{\gamma_A^p \gamma_B^p}{\gamma_A^p + \gamma_B^p} \right) \quad (1)$$

where  $\gamma_{AB}$  is the interfacial tension between phase A and B,  $\gamma_A$  and  $\gamma_B$  are respectively, the surface tensions of phase A and B, and  $\gamma^d$  and  $\gamma^p$  are the dispersion and polar components, respectively ( $\gamma = \gamma^d + \gamma^p$ ).

Pukanszky et al.<sup>10</sup> analyzed the phase structure formation of the ternary composites of PP/EPDM/ $\text{CaCO}_3$  and found that the adhesion between phases has a crucial effect on the final phase structure of the composites. If the adhesion between the elastomer and filler particles is not strong enough to stand against the shear forces in the melt during the mixing process, although thermodynamic consideration help to form an encapsulation phase structure, the elastomer and filler particles are finally separately dispersed in the PP matrix because the shear forces tend to tear the formed encapsulation. The magnitude of the adhesion between phases in the composites can be characterized by the thermodynamic work of adhesion ( $W_{AB}$ ), which is calculated using the following formula<sup>17</sup>:

$$W_{AB} = 2(\gamma_A^d \gamma_B^d)^{1/2} + 2(\gamma_A^p \gamma_B^p)^{1/2} \quad (2)$$

where  $W_{AB}$  is the work of adhesion between phase A and B. The larger  $W_{AB}$  between EPDM and  $\text{CaCO}_3$  filler is, than that between the PP and  $\text{CaCO}_3$  filler, the stabler the encapsulation is, which helps to form an encapsulation phase structure in the final composites. Premphet et al.<sup>6</sup> also successfully analyzed the formation of phase structure of PP/EVA/ $\text{CaCO}_3$  composite using the  $W_{AB}$ .

Sumita et al.<sup>11</sup> found that, using the wetting coefficient, we can successfully predict the dispersion state of fillers in the ternary composites. The wetting coefficient can be calculated from the interfacial

tensions in the system using Young's equation as follows:

$$\omega_a = \frac{\gamma_{\text{Filler-B}} - \gamma_{\text{Filler-A}}}{\gamma_{A-B}} \quad (3)$$

where  $\omega_a$  is the wetting coefficient,  $\gamma_{\text{Filler-B}}$ ,  $\gamma_{\text{Filler-A}}$ , and  $\gamma_{A-B}$  are the interfacial tension of filler-phase B, filler-phase A, and phase A-phase B, respectively. The interfacial tension can be calculated using the eq. (1). Well then, the dispersion state of filler in the composites can be predicted according to the value of  $\omega_a$ . The criterion is that the filler is dispersed in phase A when  $\omega_a > 1$ , at the interface when  $-1 < \omega_a < 1$ , and in phase B when  $\omega_a < -1$ .

The surface tensions including its dispersion and polar components of all materials used in this study are shown in Table I. The data reveals that the surface tension of PP is similar to that of PBA; however, their dispersion and polar components are different. The polar component of PBA is the greatest among three materials, which means that the polarity of PBA is the strongest. That the polar component of PP is almost zero implies that the PP is a nonpolar polymer material. The surface tension of POE is smaller than the former two. This indicates that POE has a lower surface free energy. After surface modification, the surface tension and polar component of  $\text{CaCO}_3$  particles are markedly decreased, which means the surface free energy and polarity of  $\text{CaCO}_3$  particles are greatly decreased. This tends to decrease aggregation of  $\text{CaCO}_3$  particles and to improve dispersion of  $\text{CaCO}_3$  particles in the polymer and compatibility between  $\text{CaCO}_3$  particles and polymer.

From the data in Table I, the calculated wetting coefficient ( $\omega_a$ ) of different composites, interfacial tension ( $\gamma_{AB}$ ), and work of adhesion ( $W_{AB}$ ) for all possible polymer/elastomer, polymer/filler, and elastomer/filler pairs in the ternary composites are shown in Table II. A comparison of SEM analysis and predictions by the parameters of  $\omega_a$ ,  $\gamma_{AB}$ , and

TABLE I  
The Surface Tension and Its Dispersion, and Polar Component of the Materials

Material	Surface tension (mJ/m <sup>2</sup> )		
	Total ( $\gamma$ )	Dispersion component ( $\gamma^d$ )	Polar component ( $\gamma^p$ )
PP (190°C)	21.7	21.2	0.5
PBA (190°C)	21.8	19.7	2.1
POE (190°C)	17.5	15.6	1.9
$\text{CaCO}_3$ (25°C)	158.3	69.4	88.9
$\text{CaCO}_3$ -stearic (25°C)	42.4	26.4	16.0
$\text{CaCO}_3$ -g-PBA (25°C) <sup>a</sup>	47.1	23.4	13.7

<sup>a</sup> Percent grafting = 5.4%, see the details in Ref. 12.

**TABLE II**  
Wetting Coefficient ( $\omega_a$ ) of Different Composites, Interfacial Tension ( $\gamma_{AB}$ ), and Work of Adhesion ( $W_{AB}$ ) for All Possible Polymer/Elastomer, Polymer/Filler, and Elastomer/Filler Pairs

Composite	A phase	B phase	$\omega_a$	Possible pairs	$\gamma_{AB}$ (mJ/m <sup>2</sup> )	$W_{AB}$ (mJ/m <sup>2</sup> )
PP/PBA/CaCO <sub>3</sub> -g-PBA	PBA	PP	3.30	PP/PBA	1.11	42.9
				PP/CaCO <sub>3</sub> -g-PBA	22.4	49.7
				PBA/CaCO <sub>3</sub> -g-PBA	18.8	53.7
PP/POE/CaCO <sub>3</sub>	POE	PP	-2.54	PP/POE	1.71	38.3
				PP/CaCO <sub>3</sub>	113	89.8
				POE/CaCO <sub>3</sub>	117	91.9
PP/POE/CaCO <sub>3</sub> -stearic	POE	PP	0.76	PP/POE	1.71	38.3
				PP/CaCO <sub>3</sub> -stearic	15.2	52.9
				POE/CaCO <sub>3</sub> -stearic	13.9	51.6

$W_{AB}$  for the dispersion state of CaCO<sub>3</sub> particles in the ternary composites is given in Table III. A study of the data shows that, in the ternary composite of PP/PBA/CaCO<sub>3</sub>-g-PBA, the  $\gamma_{AB}$  for PP/CaCO<sub>3</sub>-g-PBA is greater than that for PBA/CaCO<sub>3</sub>-g-PBA. Therefore, the PBA/CaCO<sub>3</sub>-g-PBA pair tends to be formed to meet the law of minimum total free energy. This results in an encapsulation phase structure of the CaCO<sub>3</sub>-g-PBA particles by PBA. The  $W_{AB}$  of PBA/CaCO<sub>3</sub>-g-PBA is as well greater than that of PP/CaCO<sub>3</sub>-g-PBA. This reveals a strong PBA/CaCO<sub>3</sub>-g-PBA interaction which in turn improves the stability of the formed encapsulation of the CaCO<sub>3</sub>-g-PBA particles by PBA during the melt-mixing. Hence, an encapsulation structure is formed in the composite. Because  $\omega_a > 1$ , the CaCO<sub>3</sub>-g-PBA particles are dispersed in the PBA phase. This also reflected that the phase structure of the composite is an encapsulation of the CaCO<sub>3</sub>-g-PBA particles by the PBA. The comparison in Table III reveals that the predictions for the morphology of the system by three parameters are consistent with each other and successfully supported-up by SEM analysis.

In the ternary composite of PP/POE/CaCO<sub>3</sub>, on the other hand, the  $\gamma_{AB}$  of POE/CaCO<sub>3</sub> is greater than that of PP/CaCO<sub>3</sub>. Thus, the phase structure leading to the lowest free energy of the system is when CaCO<sub>3</sub> particles and POE are separately dispersed in the PP matrix. The  $W_{AB}$  results show that the adhesion of PP/CaCO<sub>3</sub> is weaker than that of POE/CaCO<sub>3</sub>. At this rate, if an encapsulation structure is favored in the composite under some condition, the encapsulated units can stand against shear

forces during mixing so that the encapsulation structure is retained in final composite.  $\omega_a < 1$  indicates that the CaCO<sub>3</sub> particles are dispersed in the PP matrix. The comparisons in Table III show that the predictions for the morphology of the system by the parameters expect  $W_{AB}$  are successfully approved by the fact observed by SEM.

In the ternary composite of PP/POE/CaCO<sub>3</sub>-stearic, the  $\gamma_{AB}$  for PP/CaCO<sub>3</sub>-stearic is greater than that of POE/CaCO<sub>3</sub>-stearic. Hence, the free energy of the system is the lowest when CaCO<sub>3</sub>-stearic particles are dispersed in the POE and an encapsulation structure is formed. The  $W_{AB}$  of PP/CaCO<sub>3</sub>-stearic is greater than that of POE/CaCO<sub>3</sub>-stearic, which implies a stronger PP/CaCO<sub>3</sub>-stearic interaction. This worsens the stability of the inclusions against shear forces during mixing. As a result, a separate structure is favored in this composite.  $-1 < \omega_a < 1$  indicates that the CaCO<sub>3</sub>-stearic particles are at the interface between the PP and POE. This result is clearly inconsistent with the predictions from the  $\gamma_{AB}$  and the  $W_{AB}$ , however, the SEM analysis reveals that the CaCO<sub>3</sub>-stearic particles are indeed at the interface. Undoubtedly, using the wetting coefficient method is more advisable to predict the morphology of this system than using others.

## CONCLUSIONS

In this work, the morphology of the ternary PP composites containing different elastomer and CaCO<sub>3</sub> fillers was investigated. The SEM analysis reveals

**TABLE III**  
Comparisons of SEM Analysis and Predictions by Different Parameters for Dispersion State of CaCO<sub>3</sub> Particles in the Composites

Composite	SEM	Parameter		
		$\gamma_{AB}$	$W_{AB}$	$\omega_a$
PP/PBA/CaCO <sub>3</sub> -g-PBA	In PBA	In PBA	In PBA	In PBA
PP/POE/CaCO <sub>3</sub>	In PP	In PP	In POE	In PP
PP/POE/CaCO <sub>3</sub> -stearic	In interface	In POE	In PP	In interface

that three types of phase structures were formed in the different system: an encapsulation of the filler by elastomer in the PP/PBA/CaCO<sub>3</sub>-g-PBA composite, a separate dispersion of the filler and elastomer in the PP/POE/CaCO<sub>3</sub> composite, and a particular phase structure of the filler at the PP/POE interface in the PP/POE/CaCO<sub>3</sub>-stearic composite. The calculated  $\omega_a$ ,  $\gamma_{AB}$ , and  $W_{AB}$  from surface tensions of components all can predict the phase structure. Their predictions were consistent with each other and successfully supported-up the fact observed by SEM in the PP/PBA/CaCO<sub>3</sub>-g-PBA composite, and however conflicting in the other two composites.  $\omega_a$  was the most reliable for the predictions and, by contraries,  $W_{AB}$  was the most trustless. In addition, both  $\gamma_{AB}$  and  $W_{AB}$  can only predict a separate dispersion or an encapsulation phase structure and, however, were not available to predict the particular structure of the filler at the PP/elastomer interface.  $\omega_a$ , on the other hand, was competent for the prediction of three types of morphology. In a word,  $\omega_a$  was the most advisable among the three parameters for morphology prediction. When the thermodynamic consideration is investigated to analyze the formation of phase structure in the ternary composites, using all

of three parameters is advised to achieve much forceful prediction results.

## References

1. Jacoby, P.; Bersted, B. H.; Kissel, W. J.; Smith, C. E. *J Polym Sci Part B: Polym Phys* 1986, 24, 461.
2. Varga, J. *J Mater Sci* 1992, 27, 2557.
3. Lotz, B.; Wittman, J. C.; Lovinger, A. J. *Polymer* 1996, 37, 4979.
4. Kager-Kocsis, J., Ed. *Polypropylene: Structure, Blends and Composites*; Chapman & Hall: London, 1995.
5. Liang, J. Z.; Li, R. K. Y. *J Appl Polym Sci* 2000, 77, 409.
6. Premphet, K.; Horanont, P. *Polymer* 2000, 41, 9283.
7. Premphet, K.; Horanont, P. *J Appl Polym Sci* 2000, 76, 1929.
8. Guo, H. F.; Packirisamy, S.; Gvozdic, N. V.; Meier, D. J. *Polymer* 1997, 38, 785.
9. Zhang, M. Q.; Yu, G. *Macromolecules* 1998, 31, 6742.
10. Pukanszky, B.; Tudos, F.; Kolarik, J.; Lednický, F. *Polym Compos* 1990, 11, 98.
11. Sumita, M.; Sakata, K.; Asai, S.; Miyasaka, K.; Nakagawa, H. *Polym Bull* 1991, 25, 265.
12. Ma, C. G.; Rong, M. Z.; Zhang, M. Q.; Friedrich, K. *Polym Eng Sci* 2005, 45, 529.
13. Long, Y. U.; Shanks, R. A. *J Appl Polym Sci* 1996, 61, 1877.
14. Long, Y. U.; Shanks, R. A. *J Appl Polym Sci* 1996, 62, 639.
15. Premphet, K.; Preechachon, I. *J Appl Polym Sci* 2003, 89, 3557.
16. Wu, S. *Polymer Interface and Adhesion*; Marcel Dekker: New York, 1982; p 89.
17. Wu, S. *J Macromol Sci Rev Macromol Chem Phys* 1974, 10, 1.