Polymer Blends with Modified Coupling Agent. II. Effects of LICA 44 Grafted Styrene–Maleic Anhydride Copolymer on Properties of Flame Retardant Acrylonitrile–Butadiene–Styrene Blends

WEN-YEN CHIANG, CHIA-HAO HU

Department of Chemical Engineering, Tatung Institute of Technology, 40, Chungshan N. Rd., 3rd Sec., Taipei 104, Taiwan, Republic of China

Received 27 October 1997; accepted 5 January 1998

ABSTRACT: Flame retardant acrylonitrile-butadiene-styrene (FR-ABS) blends were prepared by blending tetrabromobisphenol A (TBBA) and antimony trioxide (Sb₂O₃) into the ABS resin. LICA 44 grafted styrene-maleic anhydride (SMA-g-L44) copolymers were used as high molecular weight (MW) coupling agents to modify the properties of the FR-ABS blends, and the copolymers with different LICA 44 grafting ratios were produced via the *in vivo* and the *in situ* reactions, respectively. The LICA 44 percentage and the MW of the SMA-g-L44 copolymers are important factors influencing the effects of the high MW coupling agent. The impact strength and the tensile yield stress of SMA-g-L44 modified FR-ABS blends increased obviously. The elongation at break and the limiting oxygen index of which also showed an increasing trend after the modification. The coupling effect of SMA-g-L44 became weaker at a higher grafting ratio. SEM observation showed that the interfacial boundary in the FR-ABS became fuzzy after using the SMA-g-L44 copolymers. © 1999 John Wiley & Sons, Inc. J Appl Polym Sci 71: 865-874, 1999

Key words: flame retardant acrylonitrile–butadiene–styrene; high molecular weight coupling agent; grafted styrene–maleic anhydride; LICA 44

INTRODUCTION

Current polymer materials (polymer alloys, blends, and composites) are designed for "on demand properties" and performance by mixing a polymer with other polymers, additives, or fillers. For example, flame retardant (FR) polymer materials are designed to reduce the possible danger caused by the flammability of polymers in any fire hazard. The employment of FR additives in polymer matrices is still the most common method for preparing these materials due to the low cost and convenience. Because the polymer materials are usually mixtures with a multiphase structure, the miscibility on the interphase of the polymer alloys and the adhesion on the interface of the polymer composites acts as key characteristics for the physical properties and performance of these materials.^{1,2}

Many effective FR additives are able to successfully reduce the flammability of polymer materials. Among these, the halogen-antimony synergism FRs are the most effective. Antimony trioxide, Sb_2O_3 , is known as an important assistant

Correspondence to: W.-Y. Chiang.

Contract grant sponsor: National Science Council; contract grant number: NSC86-2216-E-036-001.

Journal of Applied Polymer Science, Vol. 71, 865–874 (1999) © 1999 John Wiley & Sons, Inc. CCC 0021-8995/99/060865-10

for halogen-contained FR polymer materials. The effect of Sb_2O_3 on the flammability and thermal stability of polymers was studied by Carty and White.³ Camino and coworkers reported on the possible mechanism of the halogen-Sb synergism.^{4,5} Although the performance of these FR additives in flame retardancy is outstanding, these additives also cause a serious loss of mechanical properties, which include tensile strength, impact strength, and the elongation of polymers.⁶ The melting point and the molecular weight (MW) of the halogencontaining FR is usually much lower than that of the matrix. This makes the FR difficult to disperse well in the matrix. Moreover, the plasticizer-like characteristics of the halogen-containing FR also decrease the mechanical performance of polymers.⁷ The reason for the loss of mechanical properties is the poor compatibility between the polymer matrix and the Sb_2O_3 particle. As a result, the rigid Sb_2O_3 particle cannot be well dispersed and bonded in the matrix and finally conduces a stress concentration.

The function of coupling agents is to improve the adhesion condition of the matrix and inorganic fillers at the interface. In Ghosh and Maiti's report⁸ an organic titanate (LICA 38) was employed in Ag powder filled polypropylene to increase the mechanical properties of the material. In other cases,^{9,10} a LICA 44 titanate was utilized on fiber filled plastics. These coupling agents are able to offer better wetting on the filler surface and reduce the agglomeration of filler. However, these coupling agents cannot form a strong interaction between the matrix and the filler because the MWs of the coupling agents are not large enough to produce entanglements with the polymer matrix.

In this study, the LICA 44 grafted SMA (SMAg-L44) copolymers were employed as a new high MW coupling agent to modify the interfacial adhesion between the matrix and the Sb_2O_3 particle. The effect of the SMA-g-L44 copolymers, which were produced by *in vivo* and *in situ* methods, on the properties of acrylonitrile–butadiene– styrene resin (FR-ABS) was discussed according to the amine/anhydride ratio (AAR) and the addition content.

EXPERIMENTAL

Materials

The ABS used in this study was purchased from Taita Chemicals Co. Ltd., and was designated as

Table I	AAR Values	and LICA 44	Percentages
of In Viv	o SMA-g-L44	Copolymers	Used

In Vivo SMA-g-L44	LICA 44 (wt %)	LICA 44 (mol %)	AAR (mol)
A^{a}	0	0	0
VB	16.5	56.4	0.48
VD	25.8	70.1	0.92
VH	38.7	87.7	2.69

^a AAR, the amine/anhydride ratio. SMA, the raw material.

TA3000H for high impact purposes. The composition of TA3000H is 27% acrylonitrile, 24% butadiene, and 49% styrene. The melt flow index (MFI) is about 0.5–0.8 at 200°C/5 kg. Tetrabromobisphenol A (TBBA, Bromine Compounds Ltd.) was selected as an FR in the following experiments. Antimony trioxide powder was used as an FR assistant and was extra pure grade (Hayashi Pure Chemical Industries Ltd.). The antioxidant employed in the ABS blending process was Irganox 1076, and the content was 0.5 phr for each blend. The full name of Irganox 1076 is octadecyl 3-(3',5'-di-*tert*-butyl-4'-hydroxyphenyl) propionate.

FR-ABS Blends Using In Vivo Produced SMA-g-L44

The FR-ABS blends were prepared using a Brabender plasticorder at 200°C. The highest rolling speed of the plasticorder during the blending process was 70 rpm. ABS resin was dried at 90°C in an oven for 4 h before blending. The in vivo produced SMA-g-L44 copolymers were synthesized from SMA oligomer and LICA 44 previously in a reactor. The AAR and the LICA 44 percentage of the SMA-g-L44 copolymers are presented in Table I; they were calculated by multiplying the reaction feed composition of LICA 44 with the reaction ratio. The detail synthesis procedure and calculations were clearly stated in part I of this work.¹¹ In the blending process the thoroughly mixed SMA-g-L44 and Sb₂O₃ powder was first loaded into the ABS matrix and blended for 2 min. When the above process was completed, the mixture of FR, antioxidant, and thermal stabilizer was then added into the matrix. The contents of TBBA and Sb₂O₃ were fixed at 25 and 7 phr, respectively. The testing specimens for the FR-ABS blends were prepared via hot compression molding at 190°C and a hydraulic pressure of 150 kgf/cm^2 .

	In Vivo	In Situ			
		S	SMA		LICA 44
FR-ABS	SMA-g-L44 (phr)	phf	phr	phf	phr
A-20	_	_	2.0	_	0
VB-05	0.5	_	_	_	_
VB-10	1.0	_	_	_	
VB-20	2.0	_	_	_	
VB-30	3.0	_	_	_	
VD-20	2.0	_	_	_	
VH-20	2.0	_	_	_	_
SB-05	_	5.97	0.4179	1.18	0.0826
SB-10	_	11.94	0.8358	2.36	0.1652
SB-20	_	23.88	1.6716	4.72	0.3304
SB-30	_	35.82	2.5074	7.07	0.4949
SD-20	_	21.20	1.4840	7.37	0.5159
SH-20	_	17.52	1.2264	11.06	0.7742

Table II SMA-g-L44, SMA, and LICA 44 Content for *In Vivo* and *In Situ* Methods in FR-ABS Blends

FR-ABS Blends Using In Situ Produced SMA-g-L44

The FR-ABS blending procedure and the testing specimen preparation methods were the same as the above descriptions. However, the SMA-g-L44 copolymers in the blends were produced via an in situ method. The LICA 44 was coated on the surface of Sb_2O_3 powder in advance using the solvent method in which isopropanol was employed as the solvent. In the blending process the mixture of pretreated Sb₂O₃ powder and SMA oligomer was first loaded into the ABS matrix and blended for 2 min. The reaction between the LICA 44 and SMA oligomer was then initiated using the temperature and shear stress in the Brabender. When the 2 min was over, the mixture of FR, antioxidant, and thermal stabilizer was added into the matrix. The contents of SMA and LICA 44 required for preparation of the SMA-g-L44 by the in situ method are listed in Table II.

Properties Measurements

The tensile properties of all specimens were carried out according to the test standard ASTM D638 using an Instron universal test machine. The crosshead load was 500 kgf and the tensile speed was 5 cm/min under room temperature. The dimension of the specimen was type IV of the ASTM D638 standard. The specimens for notched Izod impact strength were 3 mm in thickness. The impact strength was measured according to the test standard ASTM D256.

A SUGA ONI Meter (model ON-1) was employed to determine the limiting oxygen index (LOI) of the materials. The investigation proceeded in accordance with the test standards of ASTM D2863-70. The N₂ and O₂ gas mixture was passed through a bed of glass balls at a flow rate of 11.3 L/min and homogenized for at least 30 s before testing.

The thermal analysis including thermal gravimetric analysis (TGA) and differential scanning calorimeter (DSC) analysis were performed by using a Du Pont Thermal Analyzer TA2000, models 951 and 910. The testing condition of TGA was set at a heating rate of 20°C/min and a nitrogen flow rate of 40 mL/min. The DSC proceeded under a heating rate of 10°C/min and a nitrogen flow rate of 30 mL/min.

RESULTS AND DISCUSSION

Effect of Sb₂O₃ on Mechanical Properties of FR-ABS

It is known that the addition of FR additives is disadvantageous to the mechanical properties of polymer matrices. Moreover, in the Sb-halogen FR system, the Sb_2O_3 has a more negative influ-



Figure 1 The effect of Sb_2O_3 content on the (\bigcirc) impact strength and (\blacktriangle) yield stress of FR-ABS blends.

ence than halogen-containing FR additives do. Figure 1 shows the effect of Sb_2O_3 on the impact and tensile yield stress of FR-ABS. The loss of impact strength in FR-ABS is about 60% of the original strength with the loading of 7 phr Sb₂O₃. The Sb₂O₃ particles are too rigid and deformable to absorb any external energy, which is the reason for the slight increase in tensile yield stress and large decrease in impact strength in Figure 1. Nevertheless, the ABS resin and Sb₂O₃ particles are predictably incompatible and the wetting of the interface is poor. Consequently, voids and cracks are created when external impact energy is applied to the interface, inducing the collapsing of impact strength because of stress concentration.

Model of SMA-g-L44 Molecular Structures

The effect of the SMA-g-L44 copolymers on the properties of FR-ABS blends can be discussed from two points of view throughout this study. One is the influence of the LICA 44 percentage on the SMA-g-L44 copolymers. The other is the effect of the copolymer content in the FR-ABS blends.

The LICA 44 percentage of SMA-g-L44 can also be stated in terms of AAR, which means the mole ratio of the amine groups of LICA 44 to the anhydride groups of SMA in the feed composition of the *in vivo* reaction. The molecular shapes of the SMA-g-L44 copolymers with two different AARs are described in Figure 2. Where AAR = 1.5, the number of primary amine groups in LICA 44 is equal to the anhydride groups in SMA, so the MW of the SMA-g-L44 copolymer is increased by crosslinking. However, when AAR = 4.5, there is only one primary amine group for each LICA 44 molecule possible to react to the anhydride groups of SMA. By comparing these two examples above, we know that the MW of the SMA-g-L44 copolymers is inversely proportional to the AAR in general.

Effects of In Vivo Produced SMA-g-L44

The effects of the SMA-g-L44 copolymers on the impact strength of FR-ABS are shown in Figure 3. The impact strength of FR-ABS is decreased by the addition of the SMA (A). The polarity of SMA assists in the wetting and dispersion of Sb_2O_3 particles in FR-ABS, and that induces the slight increase in impact strength at the content of 0.5 phr. However, as the content of SMA increases over 1.0 phr, the excess SMA forms an additional separated dispersion phase in the blends; in ad-



Figure 2 The model of the molecular structures of the SMA-*g*-L44 copolymers with two critical values of AAR.



Figure 3 The effect of SMA-*g*-L44 copolymers (\bigcirc) A, (\blacktriangle) VB, (\blacksquare) VD, (\diamondsuit) VH, and (\triangle) SB on the relative impact strength of FR-ABS.

dition, the MW of the SMA is too small to form strong enough entanglements with the ABS resin. As a result, increasing the SMA content decreases the impact strength of FR-ABS.

The SMA-g-L44 copolymer VB (AAR = 0.48) exhibits an obvious improvement on the impact strength of FR-ABS. The effect becomes weaker with the increase in the AAR of the copolymers, such as copolymers VD and VH. Based on the model suggested in Figure 2, the SMA-g-L44 copolymer with the higher AAR means that more LICA 44 is grafted and the MW is lower. Although the SMA-g-L44 copolymer with the higher AAR offers the stronger adhesion force because of LICA 44, the MW of SMA-g-L44 is a more important factor on impact strength. High MW copolymers are advantageous to the formation of intermolecular entanglements and end-chain rotation, which are able to absorb external impact energy. That is the reason why the copolymer VB is more effective than VH for impact strength. The impact strength of FR-ABS increases with the increase of SMA-g-L44 content, and the optimum content of each copolymer is 2.0 phr.

The tensile yield stress of FR-ABS increases with the addition of SMA, which is due to the improved wetting on the interface (Fig. 4). The SMA, which surrounds the Sb_2O_3 particles, is able to resist the propagation of cracks on the interface when a stress is applied. The main factor influencing the tensile yield stress here is the wetting of the interface. Figure 4 also shows that the tensile yield stress of FR-ABS increases in proportion to the SMA-g-L44 content and the AAR of the copolymers. The results infer that the tensile yield stress of FR-ABS is closely related to the adhesion in the ABS/Sb₂O₃ interface, and the MW of SMA-g-L44 seems not so important to tensile yield stress as impact strength. The SMAg-L44 copolymers with higher AAR can offer stronger adhesion force on the interface. However, there is a crossover between the curves of VD and VH in Figure 4. It is noted that the improved effect caused by the copolymer VH on tensile yield stress is less than that using the copolymer VD at a content over 1.0 phr. In this case the key factor to tensile yield stress for FR-ABS seems to transfer from the LICA 44 percentage to the MW while the SMA-g-L44 content increases from 1 to 2 phr. Although the LICA 44 percentage of the copolymer VH is the highest of all, the excessive LICA 44 side chains cannot reach the surface of the Sb₂O₃ particles because of steric hindrance.

The elongation behavior of FR-ABS is more complex than the impact and tensile yield stress. There are three phenomena, which can be observed in Figure 5. First, the addition of 2.0 phr SMA causes higher elongation than all *in vivo* produced SMA-g-L44 copolymers. Second, the SMA-g-L44 copolymer VH is the most disadvantageous because it reduces the percentage of elongation at break for FR-ABS. Third, there is a transition point at 2.0 phr for the copolymers VB



Figure 4 The effect of SMA-*g*-L44 copolymers (\bigcirc) A, (\blacktriangle) VB, (\blacksquare) VD, (\diamondsuit) VH, and (\triangle) SB on the relative yield stress of FR-ABS.



Figure 5 The effect of SMA-*g*-L44 copolymers (\bigcirc) A, (\blacktriangle) VB, (\blacksquare) VD, (\blacklozenge) VH, and (\triangle) SB on the elongation at break of FR-ABS.

and VD. The SMA in the ABS matrix is partially miscible and acts as a lubricant during the elongating of FR-ABS. The SMA molecules are compatible with the ABS molecules, so the ABS molecules are able to orient easily and the appearance of cracks is delayed. The SMA content of 0.5 phr is believed to be insufficient to cause the above effect. The SMA-g-L44 copolymers do not have the lubricant characteristic of SMA because of the much higher MW and the LICA 44 side chains. The MW of the SMA-g-L44 copolymers seems to be more important than the adhesion



Figure 6 The effect of SMA-g-L44 copolymers (\bullet) VD and (\blacktriangle) VH on the limiting oxygen index of FR-ABS.

force to the elongation. Thus, although the LICA 44 percentage of the copolymer VH is the highest, the copolymer VH still causes the worst effect of all to the elongation at break. By comparing the elongation curves of copolymers VB and VD, the elongation of copolymer VD is higher than that of VB at contents under 2.0 phr; but this condition reverses at 3.0 phr. In the case of two SMA-g-L44 copolymers with near MWs, the reversion can be explained by the fact that the key factor is transferred from the MW to the LICA 44 percentage. The combined effect of intermolecular entanglement and interfacial adhesion at 1.0 phr makes

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FR-ABS	Code of SMA-g-L44 Used	SMA-g-L44 Content (phr)	$\begin{array}{c} T_{pi} \\ (°{\rm C}) \end{array}$	Char Yield ^a (wt %)	$\begin{array}{c} T_g \\ (^{\circ}\mathrm{C}) \end{array}$
N ^b	Non.	Non.	296.9	3.1	86.2
A-05	А	0.5	294.0	3.4	86.2
A-20	А	2.0	295.2	2.9	86.7
B-05	В	0.5	310.4	2.5	80.2
B-10	В	1.0	320.3	2.0	81.1
B-20	В	2.0	309.4	2.6	81.0
B-30	В	3.0	315.5	4.4	80.6
D-30	D	3.0	316.5	5.8	80.8
H-30	Н	3.0	317.0	7.6	80.6

Table III Initial Pyrolysis Temperature (T_{pi}) , Char Yield, and Glass Transition Temperature (T_g) of FR-ABS with Addition of Various SMA-g-L44 Copolymers

 $^{\rm a}$ Measured by TGA at 800°C under $\rm N_2$

^b None of the coupling agent was added in the FR-ABS blends.



Figure 7 The LICA 44 percentage in SMA-g-L44 versus the impact strength of FR-ABS when employing 2.0 phr SMA-g-L44 copolymers produced by (\bullet) *in vivo* and (\bigcirc) *in situ*, respectively.

the copolymer VD present the highest elongation of all except the SMA and the *in situ* produced SMA-g-L44.

Figure 6 shows the LOI curves of FR-ABS blends with the addition of the SMA-g-L44 copolymers VD and VH. The VD curve climbs up and goes into a plateau when over 1.0 phr. The rise of the curve means that the SMA-g-L44 co-



Figure 8 The LICA 44 percentage in SMA-g-L44 versus the tensile yield stress of FR-ABS when employing 2.0 phr copolymers produced by (\bullet) in vivo and (\bigcirc) in situ, respectively.



Figure 9 The LICA 44 percentage in SMA-g-L44 versus (\bigcirc) tensile Young's modulus and (\square) elongation at break of FR-ABS when employing 2.0 phr SMA-g-L44 copolymers produced by the *in situ* method.

polymer is advantageous to the LOI, but the plateau means that the enhancement effect is saturated when the copolymer VD is over 1.0 phr. Consequently, it is inferred that the raising of LOI is possibly due to the dispersion improvement of FR and Sb_2O_3 particles caused by SMAg-L44, which reaches the maximum degree at a content of 1.0 phr. Although the effect of the copolymer VH is lower than VD, the same trend is also observed on the VH curve.

The initial pyrolysis temperature (T_{pi}) , char yield, and glass transition temperature (T_g) of FR-ABS blends are listed in Table III. The T_{pi} of all blends is increased by SMA-g-L44, which can be explained as the result of reinforcement of the FR-ABS internal bonding. The char yield of FR-ABS at 800°C is also increased by SMA-g-L44. In our previous study we found that the SMA-g-L44 copolymers are able to promote the formation of char, and a similar trend is also observed in this study. As a result, possibly for the same reason, the imide group and LICA 44 in the SMA-g-L44 offer the seeds for char production. T_g is a temperature at which the end chains and side chains of molecules start to vibrate or rotate. Although the decrease of T_g is usually considered to be the reason for the increase of impact strength of a straight polymeric material, the relationship between T_g and impact strength of FR-ABS blends is not very obvious. The decrease of T_g for FR-ABS decreased by SMA-g-L44 is primarily due to the soft segment of LICA 44 side chains in the SMA-g-L44 copolymers.



 $\label{eq:Figure 10} \begin{array}{l} \mbox{The SEM photographs of the fracture surfaces of the FR-ABS blends: (a)} \\ \mbox{A-20, (b) VB-05, (c) VB-10, (d) VB-20, (e) VD-20, and (f) VH-20.} \end{array}$

Effects of In Situ Produced SMA-g-L44

The preparation of SMA-g-L44 using the *in vivo* method is a homogeneous reaction, so the reaction ratio is high. However, the *in situ* method is a heterogeneous reaction in which the SMA and LICA 44 are dispersed in the molten ABS resin; the probability for them to react to each other is less than the *in vivo* method. Hence, in the *in situ* method the reaction efficiency is anticipated to be quite low. Only a small portion of SMA and LICA 44 are able to produce the SMA-g-L44, but the greater portion of the SMA and LICA 44 exists in FR-ABS as oligomer and low MW coupling agent.

By comparing the curves for VB and SB in Figure 3, it is clear that the *in situ* method is unable to produce enough SMA-g-L44 copolymer to increase the impact strength of FR-ABS. The same result can be observed in Figure 4. On the other hand, the curve for SB is similar to curve A in Figures 3 and 4. Furthermore, the FR-ABS employing SMA-g-L44 SB seems softer than that using SMA. The curve for SB is higher than curve A for impact strength and elongation but lower on tensile yield stress. The result may be due to the unreacted LICA 44 molecule in the FR-ABS. The unreacted LICA 44 molecule acts as an intermolecular lubricant in the blends. Furthermore, the lubricant effect of LICA 44 is better than that of SMA, so the curve SB for elongation in Figure 5 has a maximum at 0.5 phr.

Figures 7–9 show the influence of the LICA 44 percentage on the effect of *in situ* produced SMAg-L44. When using the *in vivo* method, the LICA 44 percentage is advantageous to both the impact and tensile yield stress. Conversely, when the *in situ* method is employed, the increase of unreacted LICA 44 only induces an adverse effect on the mechanical properties, including Young's modulus and elongation at break. So the SMA-g-L44 molecule can be proved effective to modify the mechanical strength of FR-ABS blends.

SEM Morphological Observation

The photo in Figure 10(a) shows the fracture surface of FR-ABS blends A. The adhesion between the matrix and the additives is very poor. The voids surround the additives like bubbles because of the surface tension of the matrix, which becomes the weak point and crack initiator of the material. By employing the SMA-g-L44 copolymer, the interface surrounding the additives in photo (b) becomes unclear. Even though the fracture surface of the modified FR-ABS in photo (c) is so craggy, several Sb_2O_3 particles are still firmly bonded with the matrix, which shows a good adhesion on the interface. Photo (d) also shows that some Sb_2O_3 particles are bonded well to each other and the matrix. An Sb₂O₃ particle is present at the center of photo (e), however, some other particles still "sink" under the surface of the matrix instead of "floating up." This means that the interface between the matrix and additives is no longer the weak point and the crack initiator of the material. Some droplets of SMA-g-L44 copolymer, which adhere to the surface of an Sb_2O_3 particle, are observed in photo (f); this result proves that the employed SMA-g-L44 does exist and make effective on the ABS/Sb₂O₃ interface in the FR-ABS blends.

CONCLUSION

The SMA-g-L44 copolymers proved to be good high MW coupling agents. However, if the SMA oligomer and the LICA 44 molecule in FR-ABS blends do not react to produce enough SMA-g-L44, it will not cause a modification of the mechanical properties of FR-ABS. Therefore, the *in vivo* method is a suitable approach to produce SMA-g-L44 instead of the *in situ* method.

Both the MW and LICA 44 percentage of SMAg-L44 are able to be controlled by AAR. The MW determines the degree of intermolecular entanglement of SMA-g-L44 and matrix, and the LICA 44 percentage is proportional to the interfacial adhesion strength. It is concluded that MW is an important factor in improving the impact strength and elongation, and the LICA 44 percentage is the determinant for the tensile yield stress. As a conclusion, the SMA-g-L44 copolymers increase the impact strength by intermolecular entanglement and increase the tensile yield stress through interfacial adhesion enhancement. The behavior of the elongation is affected by the cooperation of both the intermolecular entanglement and the interfacial adhesion. The LOI of FR-ABS is increased because of the dispersion improvement caused by SMA-g-L44, and the optimum content of SMA-g-L44 for the FR-ABS blends is 2.0 phr. The modification effect of SMA-g-L44 on the interfacial adhesion between the ABS matrix and Sb₂O₃ particles can be easily and clearly noticed by the SEM morphological observation.

The authors wish to express their appreciation to Dr. T. S. Lin, president of Tatung Institute of Technology, for his encouragement and support.

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