

Quantitative Analysis of Interfacial Tension Effect on the Impact Strength of Organic Flame Retardants and Acrylonitrile-Butadiene-Styrene Blends

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ABSTRACT: For a polymeric blend containing powder additive, quantitative analysis of its mechanical performances with interfacial tension methodology is generally a difficult work because it needs at least two groups of consistent and comparable surface tension data, either for the polymer matrix or for the powder additive, to calculate their interfacial tension. In this article, to quantitatively analyze the effect of interfacial tension on the impact strength of acrylonitrile-butadiene-styrene/flame retardants (ABS/FR) blends, the surface tension components (STC) of three organic flame retardant powders, FR-245 (2,4,6-tris(2,4,6-tribromophenoxy)-1,3,5 triazine), decabromodiphenyl oxide, and charringfoaming agent were measured through the combination of contact angle (CA)

and inverse gas chromatography technique. The relationship of the STC measured from CA between ABS resin and its homopolymer components (polyacrylonitrile, polybutadiene, and polystyrene) was also researched. Then, the interfacial tension between the three FR and ABS was calculated. It was found that the impact strength of the ABS/FR blends decreased almost linearly with the increase of the interfacial tension, and the linear correlation degree reached a very high value, 0.9969. © 2011 Wiley Periodicals, Inc. *J Appl Polym Sci* 124: 1815–1823, 2012

Key words: ABS; inverse gas chromatography; contact angle; interfacial tension; impact strength

INTRODUCTION

Acrylonitrile-butadiene-styrene (ABS) is one of commodity engineering thermoplastics. ABS is consisted of three monomers: acrylonitrile, butadiene, and styrene. The overall properties of ABS integrate the merits of the unique characteristics of each homopolymer. The component of acrylonitrile endows ABS with chemical resistance and heat and aging stability. The component of butadiene makes ABS to possess good impact resistance. The component of styrene contributes rigidity, glossy appearance, and ease of processability to ABS resin. In addition, the overall properties of ABS can be regulated through

the ratio and distribution of the three monomers. The methods of incorporation of the three monomers are also of concern.^{1,2}

One of the common ABS production techniques is: first of all, styrene-acrylonitrile copolymer (SAN) is synthesized through bulk polymerization and polybutadiene latex is prepared through emulsion polymerization. Then, styrene and acrylonitrile are grafted onto the polybutadiene chains to make the "rubber powder" through graft polymerization. Finally, SAN pellet and the rubber powder are blended to form ABS resin. Consequently, the microstructure of ABS resin consists of two phases: a rubbery phase dispersed in a continuous glassy phase of SAN.^{1–3} The molecular weight of SAN, the concentration, size, and distribution morphology of the rubber phase influence the toughness, rigidity, and impact strength of ABS. A family of ABS resins with a broad range of properties can be produced through variation of the parameters of controlling the two phases. An outstanding merit of ABS is its good ability to be alloyed with many other thermoplastics.^{1–6}

A drawback restricting ABS extensive applications is its inherent flammability. Consequently, flame

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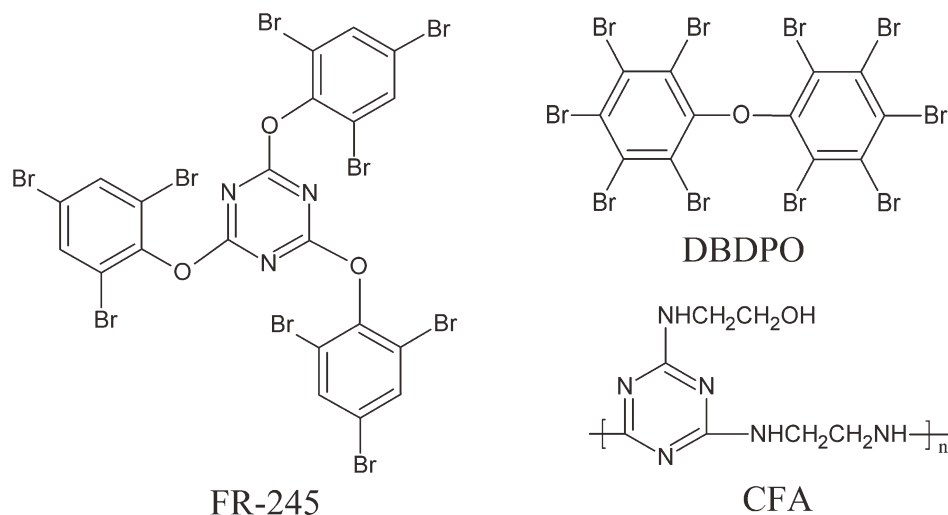


Figure 1 Chemical structure of FR.

retardants (FR) need to be added to prepare flame-retardant ABS. However, when the FR are added, the mechanical performances of ABS clearly decrease in general, especially the impact strength.^{7,8} The mechanism of how the FR influence the mechanical performance is unclear.

Generally, inorganic or organic FR are blended with ABS in powder form. Obvious interface usually exists between the ABS matrix and the powder additives. It is well known that the mechanical properties of a blend material are significantly influenced by the interfacial tension between its two composite phases. Furthermore, the interfacial tension directly relates to the surface tension of each component.⁹ Therefore, the mechanical performances of ABS/FR system must be decided by the surface tensions of ABS and the corresponding FR.

However, to our best knowledge, there are no reports on explaining the mechanical performances of polymer-powder system through quantitative researching the surface tension and interfacial tension of the components. The reason is that this procedure needs at least two groups of consistent and comparable surface tension data, either for the polymer matrix or for the powder additive, to calculate their interfacial tension. For a polymer matrix, because it generally can be easily prepared as a film with smooth surface, its surface tension or surface tension components (STC) can be obtained by using contact angle (CA) technique. However, it is very difficult to measure the surface tension of powder materials with CA technique because most powder materials cannot be prepared as the samples with the same smooth surface as the polymer matrix. Although the surface tension or STC of a powder can be measured through capillary rise or thin layer wicking experiments, direct comparison of the sur-

face tension data from two different characterization techniques is usually not accepted.^{10,11}

To quantitatively analyze the interfacial tension or surface tension effects on the impact strength of ABS/FR blends, we chose three organic FR, FR-245 (2,4,6-tris(2,4,6-tribromophenoxy)-1,3,5 triazine), decabromodiphenyl oxide (DBDPO), and charring-foaming agent (CFA). Figure 1 shows the chemical structures of them. They could be prepared as smooth samples through hot-pressing method in this experiment. Although it seemed that their surface tensions could be easily measured through CA technique when they were prepared as smooth samples, in fact, the key liquid probes (methylene iodide and bromonaphthalene) for measuring the dispersive surface tension could not be successfully used to test the FR because the liquids quickly swelled the samples.

In this article, first, the STC of a kind of ABS resin, its component of SAN resin, and its three homopolymer components (polyacrylonitrile [PAN], polybutadiene [PB], and polystyrene [PS]) were characterized by CA technique. The STC of the three FR were characterized through the combination of CA and inverse gas chromatography (IGC) technique. Then, the interfacial tensions between ABS and the FR were calculated from the surface tension values, and the effect of the interfacial tension on the impact strength was discussed. We think that this report will be beneficial for the quantitative explanation on the mechanical performance of "polymer-powder additive" system with the surface tension and interfacial tension theory. Some basic concepts of STC theory, IGC technique, and the reason for combining CA and IGC to characterize FR will be introduced in the following section.

THEORY AND METHODOLOGY

STC theory

Surface tension comes from the molecular forces in the surface. STC theory is a semiempirical theory describing the molecular forces in the surface of condensed materials. It was proposed by Fowkes and developed by Van Oss, Chaudhury, and Good in the 1980s. In this theory, three parameters are used to characterize the solid surface tension γ_S : the dispersive component γ_S^d , the acidic component γ_S^+ (or electron acceptor component), and the basic component γ_S^- (or electron donor component), such that:^{12,13}

$$\gamma_S = \gamma_S^d + 2\sqrt{\gamma_S^+ \gamma_S^-} \quad (1)$$

Using three liquids with known STC, the STC of the solid can be determined from a linear set of three equations with the following form through CA measurement techniques:

$$\gamma_{L,i}(1 + \cos \theta_i) = 2\left(\sqrt{\gamma_{L,i}^d \gamma_S^d} + \sqrt{\gamma_{L,i}^+ \gamma_S^-} + \sqrt{\gamma_{L,i}^- \gamma_S^+}\right) \quad (2)$$

$i = 1, 2, 3$

where θ_i is the equilibrium CA of liquid i with the solid surface. The interfacial tension between two solid phases (1 and 2) can be calculated using the following equation:

$$\gamma_{1,2} = \left(\sqrt{\gamma_1^d} - \sqrt{\gamma_2^d}\right)^2 + 2\left(\sqrt{\gamma_1^+ \gamma_2^-} + \sqrt{\gamma_1^- \gamma_2^+} - \sqrt{\gamma_1^+ \gamma_2^-} - \sqrt{\gamma_1^- \gamma_2^+}\right) \quad (3)$$

For this method, methylene iodide or bromonaphthalene must be used as test liquid to measure the dispersive surface tension because they contain only dispersive force.

Combination of CA and IGC technique

For measuring the STC with STC theory and CA technique, methylene iodide or bromonaphthalene is the necessary probe liquid to decide the dispersive surface tension γ_S^d .¹⁴ However, when a sample can be swollen by the two liquids or the liquids can penetrate in the sample, the STC of the sample cannot be decided directly with only CA technique. In this experiment, the chosen three FR were rapidly swollen by these two liquids.

To overcome the difficulty, IGC technique was introduced in this experiment. IGC is an important technique for the characterization of surface properties of solid materials, especially powder materi-

als.^{15,16} When a solid powder is taken as stationary phase, the dispersive surface tension can be determined by using a series of linear alkane liquids (n -alkanes) as probes. When Schultz method is adopted, the basic principle is:¹⁵ first, the dispersive adsorption free energy ΔG^{ads} of a probe with the carbon number n is calculated by the following equation:

$$\Delta G^{\text{ads}} = -RT \cdot \ln(V_{N,n}) + C \quad (4)$$

where R is gas constant, T is operation temperature, V_N is net retention volume, and C is a constant. According to the relation for work of adhesion (W_a) by dispersive force between two phases, the work of adhesion of the probe is:

$$W_a = 2\sqrt{\gamma_S^d \cdot \gamma_l^d} \quad (5)$$

where γ_S^d is the dispersive surface tension of the solid stationary phase, and γ_l^d is the dispersive surface tension of the probe. According to the following equation:

$$-\Delta G^{\text{ads}} = N \cdot a \cdot W_a \quad (6)$$

where a is the cross section area of the probe. After combining eqs. (4) and (5) with eq. (6), the following equation can be obtained:

$$RT \cdot \ln(V_{N,n}) = 2N \cdot a \cdot (\gamma_l^d)^{0.5} \cdot (\gamma_S^d)^{0.5} + C \quad (7)$$

When plotting $RT \cdot \ln(V_{N,n})$ versus $a \cdot (\gamma_l^d)^{0.5}$ for the series of liquid n -alkanes, a line can be obtained. Then, the dispersive surface tension of the solid stationary phase can be calculated from the slop of the line.

The unit of dispersive surface tension measured by IGC and CA has the same dimension, mJ/m^2 . Some researchers have found that for some polymers, IGC and CA could give similar dispersive surface tension results as listed in Table I. For some pharmaceutical powders, similar results were also found.²¹⁻²³ Consequently, when we first used IGC to decide the dispersive surface tension of the three FR, then used CA technique to decide the acidic and basic surface tensions with other two liquids (such as water and formamide), the STC of the three FR could be decided by the combination method.

EXPERIMENTAL

Materials

Five polymers were measured in this experiment. They were PAN, PB, PS, a copolymer of SAN, and

TABLE I
Dispersive Surface Tension of Some Polymers
Measured with CA and IGC

Names	γ_s^d (mJ/m ²) measured with CA	γ_s^d (mJ/m ²) measured with IGC
Polyethylene	32.0 ²	28.1 (20°C) ¹⁷
Polybutylene terephthalate	45.8 ¹⁸	42.4 (22°C) ¹⁹
PPFPMA	14.53 ²⁰	14.8 (35–45°C) ²⁰

ABS, PS was purchased from Yanshan Petrochemical Co., China. PAN, PB, SAN, and ABS were provided by Jinlin Petrochemical Co., China. The mass ratio of styrene to acrylonitrile in the SAN was 4 : 1, which was analyzed with a VarioEL Elemental Analyzer (Elementaranalysensysteme, Germany). The mass ratio of acrylonitrile to butadiene to styrene of the ABS was 1 : 2 : 4.

Three FR were chosen, which were FR-245, DBDPO, and CFA. FR-245 (T_m 230°C) and DBDPO (T_m 304°C) were purchased from Shandong Shouguang Guangda Engineering Trade Co., China. CFA (softening temperature 180°C) was provided by Jiamusi Waldo Cable Co., China. Figure 2 shows their scanning electron micrographs (SEM) at 5000 \times . They were milled as powders by a milling machine before using. The size of the particles was decided with a laser particle sizer instrument (OMEC LS 800, Zhuhai, China). For FR-245, DBDPO, and CFA, the average diameter of the particles with the content of 60% in the total particles was about 4.0, 3.6, and 4.3 μ m, respectively. The apparent density of FR-245, DBDPO, and CFA powders was 1.75, 2.43, and 0.91 g/cm³, respectively.

Measurement of STC of the polymers by CA technique

PS, PAN, SAN, and ABS smooth films were prepared with hot-pressing method by using a hot-

press instrument. For the preparation of PB film, a solvent evaporation method was used. The polymer solution prepared from PB and THF was first cast on a flat smooth plastic plate. Then the solvent was slowly vapored in an oven at 40°C for 4 h.

The polymer films with the size of 20 mm \times 10 mm were measured on a self-made dynamic cycling CA equipment using axisymmetric drop shape analysis approach. For PAN, PS, SAN, and ABS films, methylene iodide, formamide, and water were used as the test liquids. For PB film, methylene iodide, ethylene glycol, and ethylacetate were used as test liquids. The advancing CA θ_a of each drop profile was taken as the equilibrium CA.²⁴ During the calculation of STC, the used scales for the test liquids were proposed by van Oss, Chaudury, and Good.²⁵ For each polymer, three measurements were performed on three new films for each liquid.

Measurement of STC of the FR by IGC and CA techniques

As aforementioned, because methylene iodide and bromonaphtalene could swell the FR, the dispersive surface tension of the three FR had to be measured by IGC technique. For the IGC analysis, the *n*-alkanes probes were *n*-hexane, *n*-heptane, *n*-octane, and *n*-nonane. They were analytical grade solvents and purchased from Tianjin Kermel Chemical Reagents Development Centre, China. Methane was used as the noninteracting probe, which was prepared in our laboratory through heating sodium hydroxide and sodium acetate. The instrument was a GC-900A gas chromatograph (Shanghai TianPu Analytical Instrument, China), equipped with a flame ionization detector (FID). Nitrogen was used as the carrier gas. The flow rate was 15.0 mL/min. The injector and FID were heated to 180°C. The probe solvents were injected manually by using a 1.0 μ L Hamilton syringe. The injection volumes were 0.1 μ L. The columns were stainless steel tubes (0.5 m length,

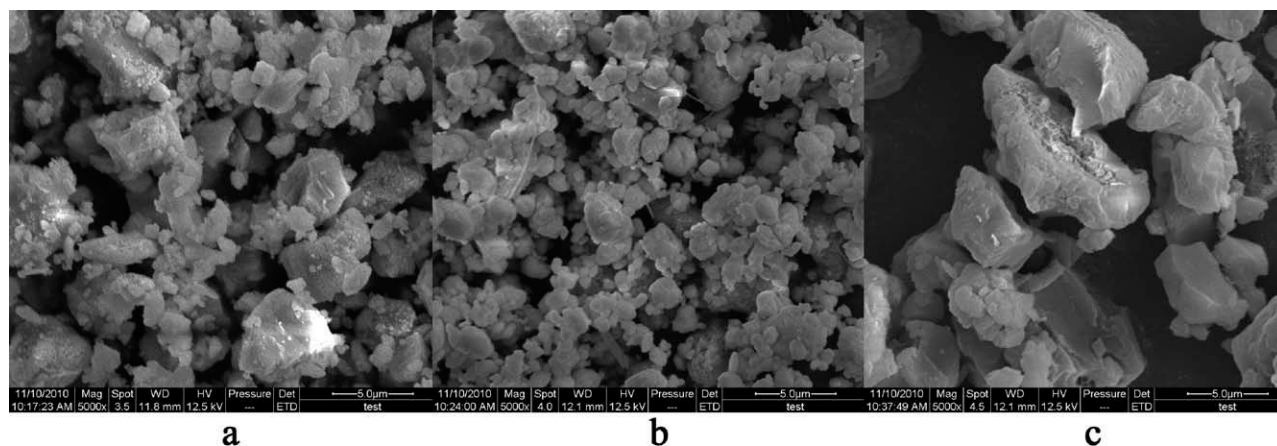


Figure 2 SEM of FR: (a) FR-245, (b) CFA, and (c) DBDPO.

TABLE II
Advancing Contact Angle (°) of Each Test Liquid on the Polymers

Names	Water	Formamide	Methylene iodide	Ethylene glycol	Ethylacetate
ABS	78.0 ± 0.5	66.1 ± 0.5	24.5 ± 0.5	–	–
SAN	75.8 ± 0.5	60.3 ± 0.5	27.4 ± 0.5	–	–
PAN	63.5 ± 0.5	62.2 ± 0.5	44.5 ± 0.5	–	–
PS	83.9 ± 0.5	54.0 ± 0.5	35.1 ± 0.5	–	–
PB	–	–	42.5 ± 0.5	83.0 ± 0.5	23.0 ± 0.5

2.56 mm i.d.). They were washed with acetone before use. The stationary phase of the FR was prepared by the method as introduced in the Ref. 26. Three columns containing DBDPO, FR-245, and CFA were prepared and were conditioned at 90°C with fast carrier gas flow rate (20 mL/min) for 12 h before measuring. The IGC experiments were performed at 30°C, 40°C, 50°C, and 60°C.

After measuring the dispersive surface tension, the CAs of formamide and water with the FR were measured by CA technique, because these FR could be prepared as sheets with smooth surface by hot-pressing method. For the sample preparation, the flame retardant sheets (3 mm thickness) were prepared with a self-made stainless steel mould and glass plates. The powders were packed in the mould and were pressed on the glass plates at 0.4 MPa and 150°C for about 5 min to obtain a smooth surface. After cooling, the sheets were taken out from the mould and tested on the CA equipment.

Measurement of impact strength of ABS/FR blends

For each flame retardant, four groups of ABS/FR blended pellets with different flame retardant mass content (5%, 10%, 15%, and 20%) were prepared from the ABS resin and the dried FR on a 35-mm twin-screw extruder with the screw's L/D value was 40 (SHJ-20, Nanjing Jieya Extruder Equipment Co., China). The design type of the twin screws was the intermeshing co-rotating type. First, the ABS resin and the flame retardant powders were mixed with a mixing machine, and then they were added in the hopper. The mixtures were fed into the extruder under gravity action. The temperatures at the front end of the barrel and the ram head were set at 190 and 220°C, respectively. The mass temperature at the die was about 180°C.

TABLE III
Average Surface Tension Components of the Polymers

Names	γ_S^d (mJ/m ²)	γ_S^+ (mJ/m ²)	γ_S^- (mJ/m ²)
ABS	46.33	1.28	12.55
SAN	45.26	0.34	11.18
PAN	37.28	0.35	28.83
PS	41.98	0.29	2.08
PB	38.33	2.79	13.19

The pellets were injected to form the standard sample bars for the impact test by using an injection molding machine (HTF86X1-A, HaiTian Groups, China) at the injection temperature of 215–225°C. The impact tests were performed on a Notched Izod impact instrument (XJC-5, Chengde Jingmi Testing Machine Co., China) at room temperature according to the GB/T 1843-2008 standards, corresponding to ASTM D 256. The cross-section morphologies of the broken bars were characterized with a SEM instrument.

RESULTS AND DISCUSSION

STC of the polymers

Table II lists the advancing CAs of each test liquid. Table III lists the average STC of the polymers, which were calculated from the data in Table II with eq. (2). For the measured dispersive surface tension value of polystyrene, 41.98 (mJ/m²), it is very close to a published value 42.0 mJ/m².²⁷ Consequently, the measured STC listed in Table III are correct.

For the copolymer SAN and the terpolymer ABS, the dispersive surface tension values were both larger than the values of their homopolymer components, PAN, PS, and PB. For the acidic and basic surface tensions, an interesting phenomenon was found, which was the measured surface tension values of SAN and ABS approximately equaled to the values calculated from the measured data of their corresponding homopolymer components, as proved in Table IV. When we adopted the molar ratio to characterize the contents of the homopolymer components in SAN or ABS, the acidic surface tension value of SAN could be calculated from the acidic surface tension values of PAN and PS through a simple molar proportional contribution method as listed in Table IV. The same conclusion was also fit for the basic surface tension values of SAN and ABS.

It is obvious that when homopolymers form copolymers or terpolymers, the dispersive and acidic/basic surface tensions obey different laws. We thought the reason was that the formation mechanisms of the two kinds of interaction forces were different. According to the molecular forces theory,¹³

TABLE IV
Comparison of Surface Tension Components of SAN and ABS between Experimental and Calculated Values

Name	SAN	ABS
Mass ratio and molar ratio of homopolymer components	Mass ratio of PS: PAN = 4 : 1 Molar ratio of PS: PAN = 2 : 1	Mass ratio of PAN: PB: PS = 1 : 2 : 4 Molar ratio of PAN: PB: PS = 1 : 2 : 2
Calculated γ_S^+ from homopolymer components	$(2 \times 0.29 + 0.35)/3 = 0.31$	$(0.35 + 2 \times 2.79 + 2 \times 0.29)/5 = 1.30$
Experimental γ_S^+	0.34 (RE _{cal} = 9.7 %)	1.28 (RE _{cal} = 1.5 %)
Calculated γ_S^- from homopolymer components	$(2 \times 2.08 + 28.83)/3 = 10.99$	$(28.83 + 2 \times 13.19 + 2 \times 2.08)/5 = 11.87$
Experimental γ_S^-	11.18 (RE _{cal} = 1.7 %)	12.55 (RE _{cal} = 5.7 %)

Note : RE_{cal} is the relative error of the calculated value.

for neutral molecules, the main component of molecular forces was the dispersive force. The origin of dispersive force was a quantum mechanism: the movement of electrons gave an instantaneous dipole to a nonpolar molecule.²⁸ However, according to the molecular orbital theory, for the acidic/basic forces, they came from the linear combination of the atomic orbitals.²⁹ Therefore, the acidic and basic surface tension in a copolymer could possibly be added from its homopolymers through a linear combination way as listed in Table IV.

STC of the FR

Figure 3 shows the plots of dispersive adsorption free energy of *n*-alkanes on DBDPO versus $a \cdot (\gamma_S^d)^{0.5}$ at the four measuring temperatures. From the slopes of the lines, the dispersive surface tension of DBDPO at each temperature was calculated. Then, the dispersive surface tension at 20°C was extrapolated from the measured values as shown in Figure 4. Table V lists the CA data of water and formamide on the sheets of the FR and the final results of STC of the FR. It can be found that the basic surface tension value of CFA is the largest. According to the chemi-

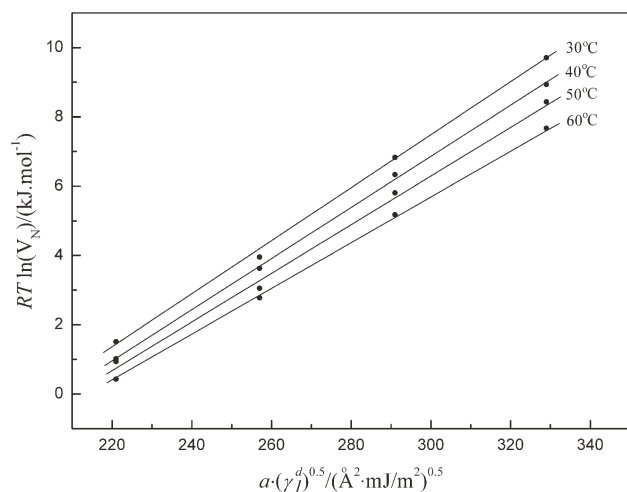


Figure 3 Dispersive adsorption free energy of *n*-alkanes on DBDPO versus $a \cdot (\gamma_S^d)^{0.5}$.

cal structures of the three FR, hydroxyl, and imido groups are contained in CFA, which present strong electron donor property (basicity). Because there are no strong polar groups in the chemical structure of DBDPO, the acidic and basic surface tensions of DBDPO are both weak.

Effect of interfacial tension on the impact strength

Figure 5 presents the plots of the notched impact strength versus the content of the FR. To find the relation between the impact strength and the interfacial tension, we needed to choose a group of impact strength data by such as a rule, which was the total interfacial area of each ABS/FR blend must be equal. We thought that only under this circumstance, the impact strength of different samples could be compared with each other. As described in the experimental part of this article, the apparent density of FR-245, DBDPO, and CFA powders was 1.75, 2.43, and 0.91 g/cm³, respectively. After calculation, we found that when the content of FR-245 was 10%, DBDPO was 15%, and CFA was 5%; these powders had the same total surface area. If they remained the same shape in their corresponding blended materials

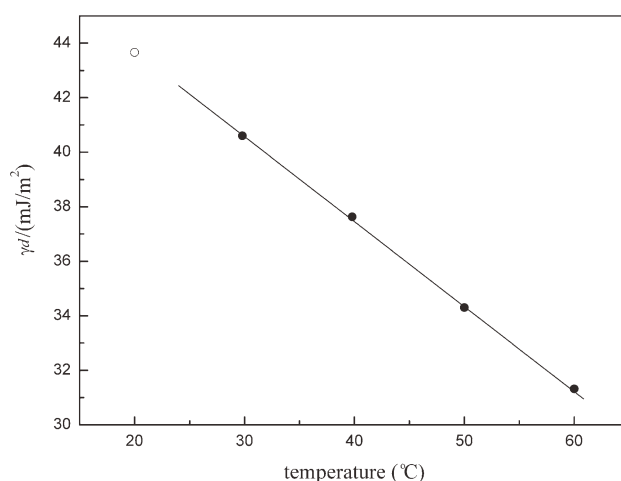


Figure 4 Dispersive surface tension of DBDPO versus temperature.

TABLE V
Surface Tension Components of the Flame Retardants

Names	Contact angle of water (°)	Contact angle of formamide (°)	Predicted γ_S^d (mJ/m ²) at 20°C by IGC	γ_S^+ (mJ/m ²)	γ_S^- (mJ/m ²)	γ_S^{total} (mJ/m ²)
FR-245	84.5 ± 0.5	77.8 ± 0.5	41.52	2.83	13.13	53.71
DBDPO	89.8 ± 0.5	51.6 ± 0.5	42.12	0.93	0.07	42.63
CFA	48.1 ± 0.5	28.5 ± 0.5	36.37	2.20	24.64	51.10

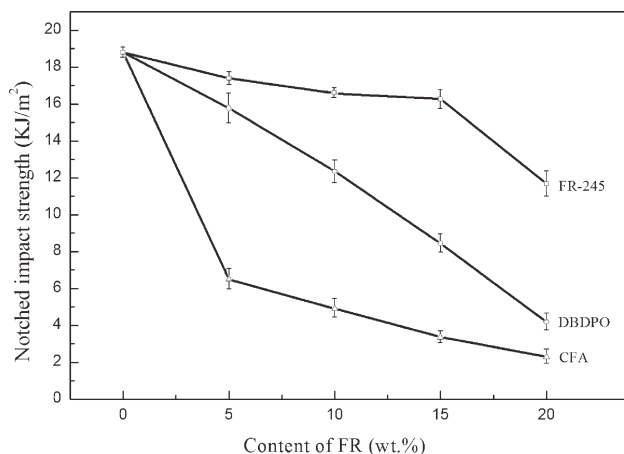


Figure 5 Notched impact strength versus content of FR.

as their initial shape, the total interfacial area would be equal.

The SEM of the cross-section morphology of the ABS/FR blends show that except DBDPO, the shape of the added FR-245 and CFA both changed, as shown in Figure 6. It can be observed that no obvious particles exist in the ABS/FR-245 sample, which means that FR-245 had melted with ABS matrix and the compatibility was good. The CFA particles formed smooth particles because CFA was intenerated at the processing temperature. The interphase region between the CFA particles and ABS

matrix was clear and the gaps in the interphase region were obvious, which indicated that poor compatibility existed between them. We can demonstrate that the interfacial area will not change although the added particles change their shape. The detailed demonstration is attached in the Appendix.

Table VI lists the chosen impact strength values and the calculated interfacial tensions with eq. (3). A large interfacial tension means that the interface is unstable, and the compatibility is not good. In this circumstance, the interface is easily broken. The impact strength of the corresponding blended material is low, such as the ABS/CFA system. Figure 7 shows the plot of the impact strength versus the interfacial tension. It presents that the impact strength of ABS/FR blends decreases almost linearly with the increase of the interfacial tension. The linear correlation degree reaches a very high value, 0.9969. Consequently, it means that for different ABS/FR blends, when their interfacial area has the same value, their impact strength is decided by their interfacial tension.

CONCLUSIONS

In this article, the STC of five polymers (ABS, SAN, PAN, PB, and PS) were measured with CA technique at room temperature. The STC of three FR (FR-245, CFA, and DBDPO) were measured with the

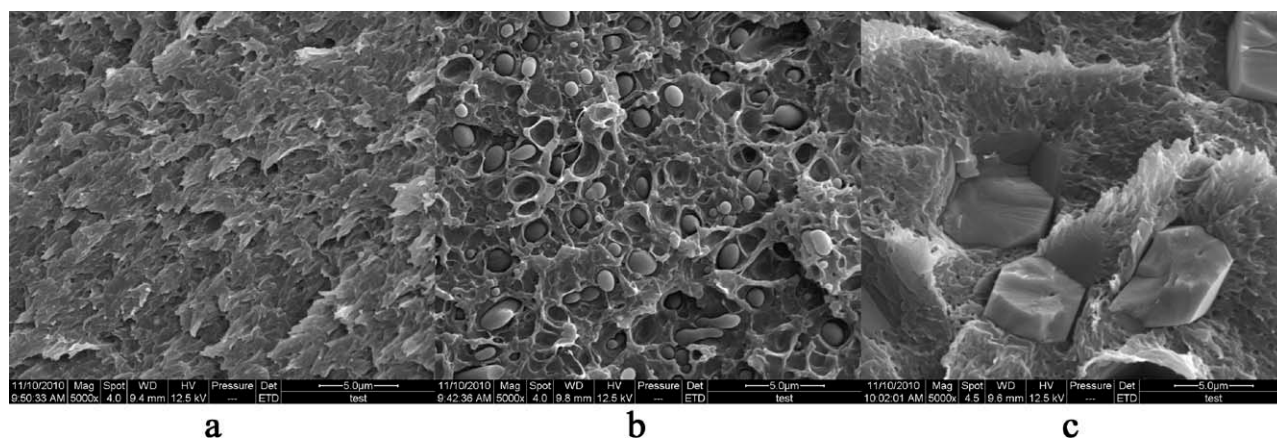


Figure 6 SEM of cross-section morphology of ABS/FR blends: (a) ABS/FR-245 (15%), (b) ABS/CFA (15%), and (c) ABS/DBDPO (15%).

TABLE VI
Impact Strength and Interfacial Tension
of ABS/FR Blends

Name	Impact strength (MPa)	Interfacial tension $\gamma_{1,2}$ (mJ/m ²)
ABS resin	18.8 ± 0.2	0.0
ABS/FR-245 (10%)	16.58 ± 0.1	0.2
ABS/DBDPO (15%)	8.43 ± 0.3	1.2
ABS/CFA (5%)	6.5 ± 0.3	1.6

combination method of IGC and CA technique. We found that for the acidic and basic STC, the measured values of the multipolymers (ABS and SAN) approximately equaled to the values calculated by a simple molar proportional contribution method from the measured data of their corresponding homopolymer components. However, the dispersive STC did not obey the law. The dispersive surface tension values of the multipolymers were both larger than the values of their homopolymer components.

The measured impact strength of ABS/FR-245, ABS/DBDPO, and ABS/CFA blends having the same interfacial area was 16.6, 8.4, and 6.5 kJ/m², respectively. The corresponding interfacial tension calculated from the surface tension values was 0.2, 1.2, and 1.6 mJ/m², respectively. It was found that the impact strength decreased almost linearly with the increase of the interfacial tension, and the linear correlation degree reached a very high value, 0.9969. It meant that the impact strength was decided by the interfacial tension, and the combination method of using IGC and CA to determine the STC of powder materials was reliable.

APPENDIX

The total interfacial area of an amount of flame retardant powder in the cross-section of a broken ABS/FR sample bar will not change with the shape of the powder. In other words, whatever if the larger flame retardant particles melt and form smaller particles or penetrate in the ABS matrix, the total interfacial area between the flame retardant and ABS matrix will not change. This is demonstrated by the following demonstration.

For an amount of flame retardant particles with an average radius of r_1 and a total number of n_1 , the total volume V of the flame retardant powder equals:

$$V = \frac{4}{3}\pi r_1^3 \cdot n_1 \quad (\text{A1})$$

when a large particle changes to several small particles with an average radius of r_2 ($r_2 < r_1$, as illustrated in Fig. A1) and the total number of small particles is n_2 , the total volume V equals:

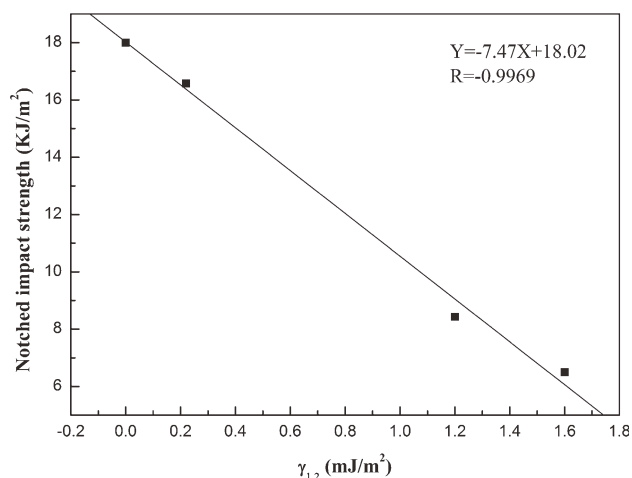


Figure 7 Plot of impact strength versus interfacial tension.

$$V = \frac{4}{3}\pi r_2^3 \cdot n_2 = \frac{4}{3}\pi r_1^3 \cdot n_1 \quad (\text{A2})$$

Then, eq. (A2) can be rearranged as:

$$\frac{n_1}{n_2} = \frac{r_2^3}{r_1^3} \quad (\text{A3})$$

When we suppose that the particle number of the flame retardant powder in the cross section of a

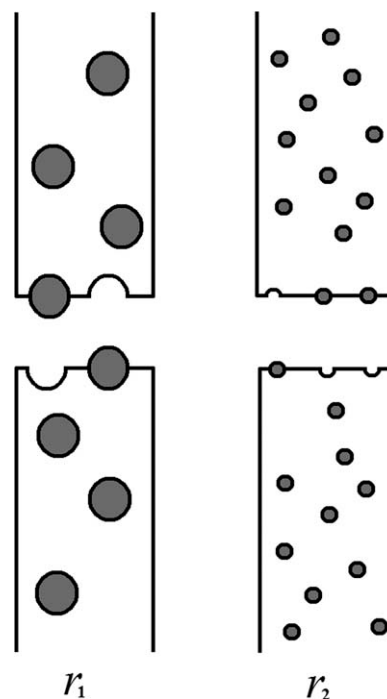


Figure A1 Schematic drawing of shape change of flame retardant powder.

broken ABS/FR bar with a unit area is n'_1 before changing its shape, and n'_2 after changing to the small particle with the average radius of r_2 , there are following relations:

$$n'_1 = n_{1, \text{ph}}^2 \quad (\text{A4})$$

$$n'_2 = n_{2, \text{ph}}^2 \quad (\text{A5})$$

For one flame retardant particle with the average radius of r_1 in the ABS/FR bar, when the sample bar is impacted to two pieces, if the particle is not broken up and remains in one piece of the ABS matrix, the interfacial area S between the particle and another piece of ABS matrix will approximately equal:

$$S = \pi r_1^2 \quad (\text{A6})$$

The relation is reasonable because the radii of all flame retardant particles in this experiment are very small. Then, the total interfacial area S_1 formed by the particles with average radius of r_1 is:

$$S_1 = n'_1 \cdot \pi r_1^2 = n_{1, \text{ph}}^2 \cdot \pi r_1^2 \quad (\text{A7})$$

And the total interfacial area S_2 formed by the particles with the average radius of r_2 is:

$$S_2 = n'_2 \cdot \pi r_2^2 = n_{2, \text{ph}}^2 \cdot \pi r_2^2 \quad (\text{A8})$$

When combining eq. (A7) with eq. (A8), the following ratio is obtained:

$$\frac{S_1}{S_2} = \frac{n_{1, \text{ph}}^2 \cdot \pi r_1^2}{n_{2, \text{ph}}^2 \cdot \pi r_2^2} = \left(\frac{n_1}{n_2}\right)^{\frac{2}{3}} \cdot \frac{r_1^2}{r_2^2} = \left(\frac{r_2}{r_1}\right)^{\frac{3}{3}} \cdot \frac{r_1^2}{r_2^2} = 1 \quad (\text{A9})$$

It means that the total interfacial area does not change with the shape of the particles.

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