

# A New Composition–Processing–Property Relationship for Studying the Tensile Modulus-Phase Plastic Blends

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**ABSTRACT:** Based on thermodynamic principles, a composition–processing–property relationship for predicting the modulus properties of multiphase plastic blends has been developed. This relationship describes the relative modulus of the blend in terms of the volume fraction and the index for the degree of mixing of an inclusion-polymer in the matrix-polymer. The relative modulus is defined as the ratio between the modulus of the blend and that of the matrix polymer. These blends include a nylon 6,6/polymethyl methacrylate (PMMA) system mixed using an injection molding process and a nylon 6/ethylene-vinyl acetate copolymer system mixed using a corotational extrusion process. Based on the values deter-

mined for the mixing index of the nylon 6,6/PMMA blends, a relationship between the mixing index and the fill time used in the injection molding has been developed. The results also imply that the degree of mixing of the blend mixed using a correlation extrusion process is better than that of the blend processed using an injection molding process. Using the above results, we now can scientifically develop new plastic blends and design optimum processing conditions for various automotive applications. © 2011 Wiley Periodicals, Inc. *J Appl Polym Sci* 121: 1593–1599, 2011

**Key words:** modulus; plastic; blends; inclusion; processing

## INTRODUCTION

Among the many physical properties, the modulus property (for example, tensile modulus, shear modulus, or flexural modulus) is considered as one of the most important requirements for selecting plastics in various automotive applications.<sup>1–3</sup> The modulus property of plastics can be varied by two different methods. The first method is to mix a hard second-phase (inclusion) material, such as glass fiber, mineral fillers, and hard polymer inclusions, into a polymer matrix. This method effectively raises the modulus value of the plastics.<sup>4,5</sup> The second method is to blend a soft second-phase material, such as soft polymer inclusions and plasticizers, into the polymer matrix. This method lowers the modulus value of the plastics.<sup>6,7</sup> The processing condition also greatly affects the modulus property of the multiphase plastics.<sup>8</sup> A systematic study of the effects of composition and processing on the modulus properties of the plastic blends is, therefore, of vital importance.

In previous research, based on thermodynamic principles, a composition–processing–property relationship for predicting the modulus property of filled polymers was developed.<sup>9–12</sup> Experimental

data on many filled polymer systems substantiated the new relationship. These include filler-reinforced elastomers,<sup>13,14</sup> plasticizer-filled elastomers,<sup>15</sup> elastomer blends,<sup>16</sup> filler-reinforced plastics,<sup>17</sup> and polymer foams.<sup>18</sup> Furthermore, this relationship also led to the development of new mixing techniques for enhancing the degree of mixing of the filler in the polymer matrix.<sup>19,20</sup>

The purpose of this work is to establish a composition–processing–property relationship for predicting the modulus property of multiphase plastic blends and to verify the above relationship using modulus data of plastic blends measured in our laboratory and obtained from the literature.<sup>21</sup>

## THEORY

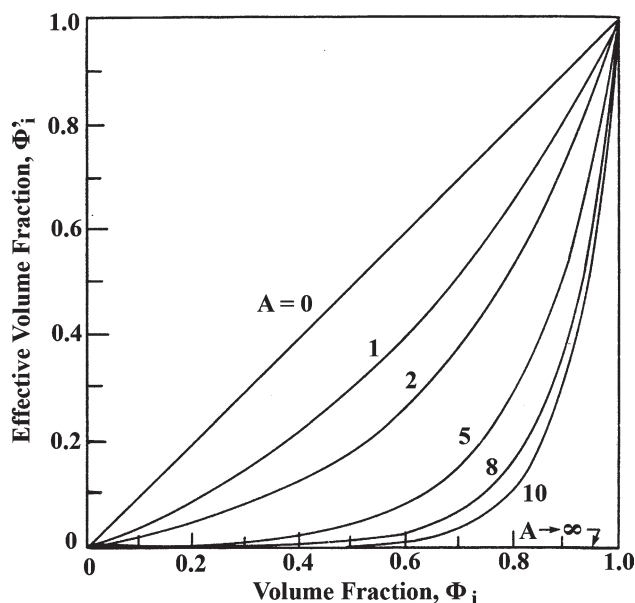
### Effective volume fraction of inclusion

Based on thermodynamic principles, a new state variable, called the effective volume fraction of the inclusion in a polymer matrix,  $\phi'$ , has been studied.<sup>22,23</sup> This new variable,  $\phi'$ , which combines the volume fraction,  $\phi$ , and the index for the degree of mixing,  $A$ , of the inclusion, has the mathematical form shown below.<sup>24</sup>

$$\phi' = \phi \exp[A(\phi - 1)] \quad (1)$$

Using eq. (1), we calculated values of  $\phi'$  with respect to various values of  $\phi$  and  $A$ . The results are shown

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**Figure 1** Relationship between the effective volume fraction ( $\phi'$ ) and the actual volume fraction ( $\phi_i$ ) of the inclusion calculated at various indexes of degree of mixing ( $A$ ) and zero strain.

in Figure 1. Physically, the results imply that as long as the degree of mixing of the inclusion deviates from its ideal mixing state, the value of  $\phi'$  is less than that of  $\phi$ . Based on the prediction shown in Figure 1, the modulus properties of a real blend system can be significantly different from those of an ideal blend system.

### Relationship for predicting tensile modulus of plastic blends

Using thermodynamic principles<sup>25,26</sup> and the concept of the effective volume fraction of the inclusion, we established a composition–processing–property relationship for the tensile modulus of polymer blends. This relationship can be described by the following:

$$\ln(E) = \ln(E_1) + \phi' \ln(K) \quad (2)$$

where

$E$  = tensile modulus of the polymer blend,

$E_1$  = tensile modulus of the matrix polymer,

$\ln(K)$  = reinforcement effectiveness, and

$\phi'$  = effective volume fraction of the inclusion polymer.

If the microvoid concentration is insignificant to affect the reinforcement effectiveness,  $\ln(K)$ , then the value of  $\ln(K)$  can be calculated using the following equation.<sup>27,28</sup>

$$\ln(K) = \ln(E_2/E_1) \quad (3)$$

In eq. (3),  $E_2$  is the effective modulus of the inclusion polymer. Its value equals the tensile modulus of the inclusion polymer, if the domain (inclusion) size is less than  $4 \mu$ .<sup>29</sup> It is noteworthy that values of the mixing index ( $A$ ) range between zero and infinity. When  $A$  equals zero, the polymer blend system is in its perfect mixing state. However, when  $A$  approaches infinity, there is no mixing between the matrix polymer and the inclusion polymer.<sup>30</sup> The value of  $\ln(K)$  in eq. (2) can be positive, zero, or negative, depending on the modulus value of the matrix polymer and that of the inclusion polymer. Equation (2) has been successfully used to predict the effects of composition and processing on the modulus properties of filler-reinforced elastomers,<sup>13,14</sup> elastomer blends,<sup>16</sup> plasticizer-filled elastomers,<sup>15</sup> filler-reinforced plastics,<sup>17</sup> and polymer foams.<sup>18</sup>

In the next section, we will first discuss the experiments that were carried out to determine values for the tensile modulus of the nylon 6,6/PMMA blend system. We will then use the modulus data to verify eq. (2).

## EXPERIMENTAL

### Material

Polymethyl methacrylate (PMMA) (Plexiglas V-052, Rohm and Haas) and polyamide (Nylon 6,6) (Zytel 101NC-10, Du Pont) were used in this investigation. The compositions of the nylon 6,6/PMMA blend system are shown in Table I.

### Mixing procedure

A dry blending procedure was used in an injection molding machine (New Britain, 75 ton) for mixing the blend compositions. The dry blending procedure is described in the following:

1. For each blend composition, a 1.5 kg of dry mixture was prepared before the injection molding process.

**TABLE I**  
Values for the Volume Fraction of the Polyamide (Nylon 6,6) Polymethylmethacrylate (PMMA) Blend

Sample designations	Volume fraction of Nylon 6,6
A1	0.00
A2	0.00
A3	0.00
B	0.20
C	0.40
D	0.60
E1	0.80
E2	0.80
E3	0.80
F1	1.00
F2	1.00
F3	1.00

2. Each of the 1.5 kg batch consisted of six premixed portions which had 0.25 kg of the mixture containing the desired blend concentration (as shown in Table I).

The injection molding temperature for the PMMA resin was determined based on a relationship between the injection molding pressure and the minimum processing temperature of amorphous polymer melts. The relationship has the form shown below.

$$T(\text{min}) = 1.212(T_g) + B(P) \quad (4)$$

where

$T(\text{min})(^\circ\text{K})$  = the minimum processing temperature of the amorphous polymer,

$T(^\circ\text{K})$  = the glass transition temperature of the amorphous polymer measured at low shear-rate and 1 atm,

$P(\text{MPa})$  = the injection molding pressure (gauge pressure), and

$B(^\circ\text{K}/\text{MPa})$  = material coefficient, being a function of the coefficients of thermal expansion and of compressibility of the polymer.

The above relationship is the controlling equation for determining the minimum processing temperature of a given amorphous polymer. The glass transition temperature of PMMA used was determined by differential scanning calorimetry and had the value of 377.0°K. The value of  $B$  shown in eq. (4) for the PMMA resin was determined to be 0.273°K/MPa,<sup>11</sup> and the injection molding pressure used was 93.1 MPa for PMMA. Using these values, in conjunction with eq. (4), we calculated the minimum processing temperature,  $T(\text{min})$ , for the PMMA resin. The value of  $T(\text{min})$  is shown below.

$$\begin{aligned} T(\text{min}) &= 1.212(377.0^\circ\text{K}) + (0.273^\circ\text{K}/\text{MPa})(93.1 \text{ MPa}) \\ &= 482.3^\circ\text{K} = 209.3^\circ\text{C} \quad (5) \end{aligned}$$

An earlier study indicated<sup>5</sup> that the optimum processing temperatures for an amorphous polymer could be determined by selecting the processing temperature at least 28.0°C higher than the minimum processing temperature of the polymer.<sup>5</sup> Using the above criterion for determining the optimum processing temperatures, we selected the injection molding temperature for PMMA to be 260°C, which was 51°C higher than its minimum processing temperature [209°C; see eq. (5)]. The melting point of nylon 6,6 was also determined by differential scanning calorimetry. The processing temperature used for nylon 6,6 was 310–315°C which was at least 45°C higher than its melting point, 265°C. The processing temperatures used for the nylon 6,6/PMMA blends were between 282 and 315°C. To investigate the effect of fill time

TABLE II  
Processing Conditions Used in the Injection Molding of the Nylon 6,6 (PMMA) Blends

Sample designations	Fill time (s)	Melt temperature (°C)	Mold temperature (°C)
A1	2.0	260	50
A2	3.5	260	50
A3	6.0	260	50
B	2.6	282	50
C	2.8	282	50
D	3.0	293	50
E1	2.0	315	50
E2	3.0	315	50
E3	6.0	315	50
F1	2.0	310	50
F2	3.0	315	50
F3	6.0	315	50

used in the injection molding process on the degree of mixing and, correspondingly, the modulus property of the blend, three fill times were used in the injection molding of the 80/20 nylon 6,6/PMMA composition. All the injection molding conditions used in this investigation are summarized in Table II.

### Sample preparation

Based on the injection molding conditions listed in Table II, all the nylon 6,6/PMMA compositions were molded into the ASTM tensile test specimens. The gauge dimensions of the tensile test specimens were 50.8 × 12.7 × 3.15 mm<sup>3</sup>. Before tensile tests, all the specimens were put in polyethylene bags containing calcium sulfate and stored in desiccators under vacuum.

### Test procedure

Tensile tests of all the dry specimens were performed at 20°C using an Irtstron tensile machine at a crosshead speed of 5.08 mm/min (0.2 in/mm) which corresponds to an initial strain rate of 1.67 × 10<sup>-3</sup> (L/sec).

## RESULTS AND DISCUSSION

### Tensile modulus of the nylon 6,6/PMMA blends

As a reminder, tensile modulus is a very important physical property that represents the resistance of a material against tensile deformation. The value of tensile modulus can be determined from the initial slope of the uniaxial tensile stress-strain curve. This method was used to determine the modulus values of all the blend compositions. Tensile-modulus data for the nylon 6,6/PMMA compositions are summarized in Table III. The modulus values of all the

**TABLE III**  
Values for the Tensile Moduli of the Nylon 6,6 (PMMA) Blends

Sample designations	Volume fraction of Nylon 6,6	Tensile modulus (Gpa)
A1	0.00	3.80
A2	0.00	3.79
A3	0.00	3.79
B	0.20	3.76
C	0.40	3.58
D	0.60	3.21
E1	0.80	3.02
E2	0.80	2.94
E3	0.80	2.86
F1	1.00	2.58
F2	1.00	2.56
F3	1.00	2.56

nylon 6,6/PMMA compositions decrease with the increase of the nylon 6,6 concentration.

In the next section, I will first use these modulus data to evaluate the validity of eq. (2). I will then determine the reinforcement effectiveness,  $\ln(K)$ , and the index for the degree of mixing,  $A$ , of each of the nylon 6,6/PMMA compositions.

#### Composition–processing–property relationship for the tensile modulus of the nylon 6,6/PMMA blends

Using eq. (2) to analyze the data shown in Table III, we obtained the relationship for the tensile modulus of the nylon 6,6/PMMA blend system as shown below.

$$\begin{aligned} \ln(E) &= \ln(E_1) + \phi' \ln(K) \\ &= \ln[3.79 \text{ (GPa)}] + \phi'[-0.39] \end{aligned} \quad (6)$$

where

$$\ln(K) = \ln(E_2/E_1) = \ln(2.57/3.79) = -0.39 \quad (7)$$

$E$  = tensile modulus of the blend,  
 $E_1$  = tensile modulus of PMMA = 3.79 GPa,  
 $E_2$  = tensile modulus of nylon 6,6 = 2.57 GPa, and  
 $\phi'$  = effective volume fraction of nylon 6,6.

It is important to note that the value of  $\ln(K)$  shown in eq. (7) for the nylon 6,6/PMMA blend system is negative. This is because the modulus value of nylon 6,6 is lower than that of the PMMA. Values for the mixing index,  $A$ , of the nylon 6,6/PMMA blend composition at various fill times were also determined using eq. (6) and are summarized in Table IV. The plots of the tensile modulus of the blend system (including the modulus values as a function of the fill time) versus the volume concentration of nylon 6,6 are shown in Figure 2.

**TABLE IV**  
Values for the Reinforcement Effectiveness,  $\ln(K)$ , and the Mixing Index,  $A$ , of the Nylon 6,6/PMMA Blends

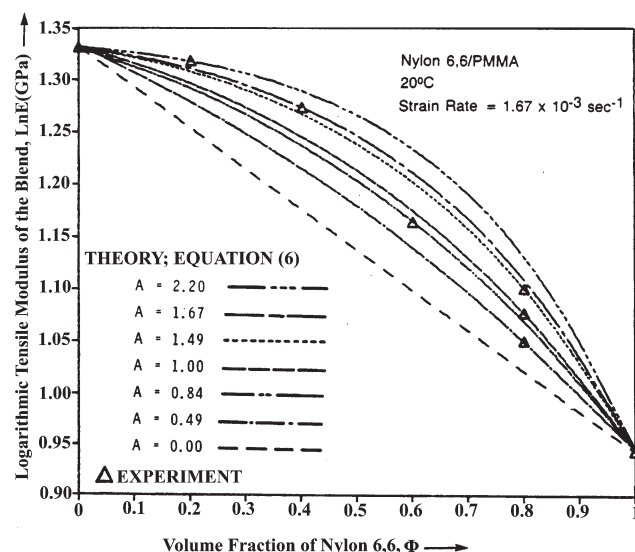
Sample designations	Reinforcement effectiveness, $\ln(K)$	Mixing index, $A$
A1, A2, A3	-0.39	0.00
B	-0.39	2.20
C	-0.39	1.67
D	-0.39	0.84
E1	-0.39	1.49
E2	-0.39	1.00
E3	-0.39	0.49
F1, F2, F3	-0.39	0.00

In the next section, I will develop a phenomenological relationship between the mixing index,  $A$ , and the fill time of the injection molding process,  $t_f$ , used in this investigation.

#### Correlation between the mixing index, $A$ , and the fill time, $t_f$ , of the 80/20 nylon 6,6/PMMA blends

Based on the results of mixing index and fill time (shown in Tables II and IV, respectively) for the 80/20 nylon 6,6/PMMA blend, we concluded that values for the mixing index,  $A$ , increase with the decrease of the fill time,  $t_f$ . The results are shown in Table V. This conclusion is significant, because, from a practical point of view, it leads to the development of optimum processing conditions for injection molding multiphase plastics.

As a reminder, the mixing index,  $A$ , describes the degree of mixing of a processed multiphase polymer (in this case, two phases), and the degree of mixing worsens as the value of mixing index increases. The fill time used to fill the mold in an injection molding



**Figure 2** Plots of the logarithmic tensile modulus,  $\ln E$ , versus the volume fraction of nylon 6,6 of the nylon 6,6/PMMA blends.

**TABLE V**  
**Values for the Fill Time,  $t_f$ , and the Mixing Index,  $A$ , of the 80/20 Nylon 6,6/PMMA Blends**

Sample designations	Fill time $t_f$ (s)	Experimental data	Equation (8) <sup>a</sup>
E1	2.0	1.49	1.49
E2	3.5	1.00	0.99
E3	6.0	0.49	0.50

<sup>a</sup> Equation (8),  $A = 2.55 \exp(-t_f/3.70)$ .

process (in this case, filling a tensile-bar mold) corresponds to an average shear-rate experienced by the polymer melt during the mold-filling process. The fill time is, therefore, an important parameter that describes changes in the degree of mixing of the blend during mold filling. Based on principles of polymer rheology and viscoelasticity, a new theory for describing the effects of rate of deformation, temperature, and pressure on the degree of mixing of multicomponent polymer liquids has been developed.<sup>27,28</sup> Using previous theories, I have derived a new relationship between the mixing index,  $A$ , and the fill time,  $t_f$ . This relationship can be expressed by the following:

$$A = A_0 \exp(-t_f/\beta) \quad (8)$$

where

$A$  = mixing index of the blend at fill time  $t_f$

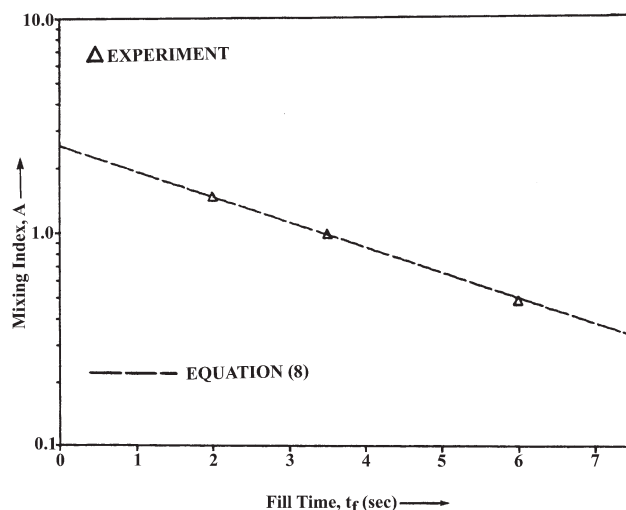
$A_0$  = initial mixing index of the blend; its value corresponding to the degree of mixing of the blend before leaving the nozzle,

$t_f$  = fill time of polymer melt in an injection molding process, and

$\beta$  = characteristic time of a given blend composition in a mold-filling process; its value also depending on the melt temperature and pressure.

Equation (8) implies that plots of the logarithmic mixing index,  $\ln(A)$ , versus the fill time,  $t_f$ , exhibits a linear relationship. The absolute value for the slope of the line defines the characteristic time,  $\beta$ . The intercept on the axis of mixing index at zero fill time defines the initial mixing index,  $A$ , of the multiphase melt before leaving the nozzle.

Using eq. (8) to analyze the data shown in Table V, I plotted the results of  $\ln(A)$  versus  $t_f$  for the 80/20 nylon 6,6/PMMA blend. The results are shown in Figure 3. The regression analysis clearly shows a linear relationship between  $\ln(A)$  and  $t_f$  with a correlation coefficient of 0.994. This linear relationship substantiates the relationship expressed in eq. (8). The values determined for the characteristic time,  $\beta$ , and the initial mixing index,  $A_0$ , of the 80/20 Nylon 6,6/PMMA blend are 3.70 sec and 2.55, respectively. These results suggest that before leaving the nozzle the 80/20 nylon 6,6/PMMA blend had a poor

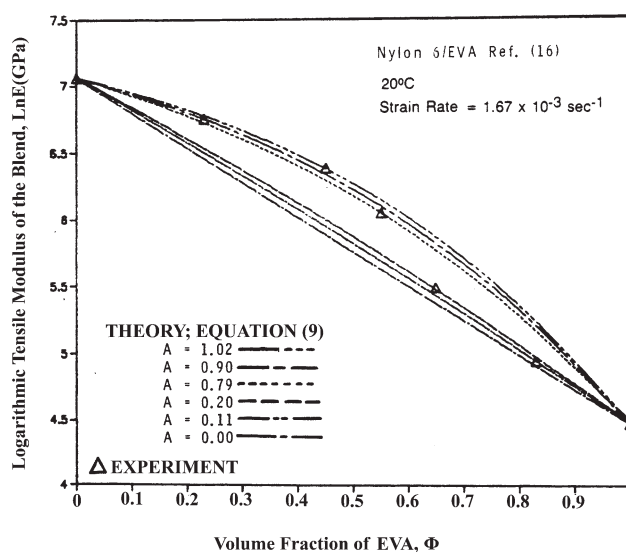


**Figure 3** Plots of the logarithmic mixing index,  $\ln A$ , versus the fill time,  $t_f$ , of the 80/20 nylon 6,6/PMMA blends.

degree of mixing (mixing index equal to 2.55), when compared with the degree of mixing after filling the mold. The results also indicate that the additional shear flow in a mold filling process enhances the degree of mixing of the 80/20 nylon 6,6/PMMA blend.

#### Application of the composition–processing–property relationship

As discussed previously, we have successfully established a composition–processing–property relationship for the modulus properties of plastic blends. This relationship has also been used to develop an analytical method for characterizing the degree of mixing of a blend mixed using a given processing



**Figure 4** Plots of the logarithmic tensile modulus,  $\ln E$ , versus the volume fraction of EVA,  $\phi$ , of the nylon 6/EVA blends.

**TABLE VI**  
**Values for the Mixing Index of the Nylon 6,6/PMMA and the Nylon 6/EVA Blends Using Different Processing Procedures**

Volume ratio of PMMA/Nylon 6,6	Nylon 6/EVA	Matrix/Domain <sup>c</sup>	Mixing index, <i>A</i>	Fill time <i>t<sub>f</sub></i> (s)
80/20 <sup>a</sup>	–	PMM/Nylon 6,6 (HM/SD)	2.20	2.6
60/40 <sup>a</sup>	–	PMM/Nylon 6,6 (HM/SD)	1.67	2.8
40/60 <sup>a</sup>	–	Nylon 6,6/PMMA (SM/HD)	0.84	3.0
20/80 <sup>a</sup>	–	Nylon 6,6/PMMA (SM/HD)	1.00	3.0
–	77/23 <sup>b</sup>	Nylon 6/EVA (HM/SD)	0.90	–
–	55/45 <sup>b</sup>	Nylon 6/EVA (HM/SD)	1.02	–
–	45/55 <sup>b</sup>	Eva/Nylon 6 (SM/HD)	0.79	–
–	35/65 <sup>b</sup>	Eva/Nylon 6 (SM/HD)	0.20	–
–	17/83 <sup>b</sup>	Eva/Nylon 6 (SM/HD)	0.11	–

<sup>a</sup> A dry blending/injection molding process was used.

<sup>b</sup> A corotational extrusion process was used.

<sup>c</sup> HM, hard matrix; SD, soft domain; SM, soft matrix; HD, hard domain.

method. In this section, I will first apply the composition–processing–property relationship to determine values for the mixing index of a nylon 6/ethylene vinylacetate(EVA) copolymer blend system studied by Han and Chuang.<sup>16</sup> I will then use these values of mixing index to determine the effect of processing procedures on the degree of mixing of the blends. The nylon 6/EVA blends were mixed using a corotational extrusion process.<sup>16</sup> The tensile modulus data were determined at room temperature using the procedure described in the ASTM D638 and a crosshead speed of 5.08 mm/min. Results for the tegressional analyses of the modulus data using eq. (2) are shown in Figure 4. All the modulus data can be described by the new composition–processing–property relationship that can be written in the form as shown below.

$$\begin{aligned} \ln(E) &= \ln(E_1) + \phi' \ln(K) \\ &= \ln(1160 \text{ MPa}) + \phi' (-2.579) \end{aligned} \quad (9)$$

where

$$\ln(K) = \ln(E_2/E_1) = \ln(88/1160) = -2.579 \quad (10)$$

*E* = tensile modulus of the blend,

*E*<sub>1</sub> = tensile modulus of nylon 6, = 1160 MPa,

*E*<sub>2</sub> = tensile modulus of the EVA, = 88 MPa, and

φ' = effective volume fraction of EVA.

Values for the mixing index of the nylon 6/EVA blends at various volume concentrations are shown in Table VI. I also list in this table values for the mixing index of the nylon 6, 6/PMMA blends.

The results in Table VI clearly show that the degree of mixing (as represented by the mixing index) of the nylon 6/EVA blend (in this case, a corotational extrusion process was used; the values of the mixing index determined range between 0.11 and 1.02) is generally better than that of the nylon 6,6/PMMA blend (in this case, a dry blending procedure

was used to mix the component polymers prior to the injection molding process); the values of the mixing index determined range between 0.84 and 2.20. In each blend system, the degree of mixing of the blend having a morphology of hard domain/soft matrix (such as the 40/60 or the 20/80 PMMA/nylon 6,6 compositions and the 35/65 or the 17/83 nylon 6/EVA compositions) is better than that of the blend having a morphology of hard matrix/soft domain (such as the 80/20 or the 60/40 PMMA/nylon 6,6 compositions and the 35/65 or the 55/45 nylon 6/EVA compositions). The above results have been predicted by using the unified theory for adhesion<sup>4</sup> in conjunction with a new rheological theory for multicomponent blends. The new rheological theory and its applications will be discussed in a future study.

## CONCLUSIONS

A new composition–processing–property relationship for predicting the tensile modulus of multiphase plastic blends has been developed. The modulus data, which were obtained from (1) the nylon 6,6/PMMA blend compositions mixed using various fill times in an injection molding process and (2) the nylon 6/EVA blend compositions mixed using a corotational extrusion process, substantiate this relationship. The experimental data on the 80/20 nylon 6,6/PMMA blend also verify a new rheological relationship between the mixing index, *A*, and the fill time used in an injection molding process, *t<sub>f</sub>*. Based on these results, we now can scientifically develop new plastic blend compositions and design optimum processing conditions for various automotive applications.

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