Original article

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The effects of morphological variation and polymer/polymer interface on the tensile modulus of binary polymer blends: a modeling approach

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Abstract: In this work, the effects of the morphological variation and the polymer/polymer interface on the tensile modulus of binary polymer blends were evaluated using a combined modeling method. The characteristics of the polymer/polymer interface region were evaluated using a neural network system and the results were used to improve the analytical model. The model accuracy was investigated by comparing its predictions with the tensile test results of some prepared iPP/PA blend samples and also some other data from literature which revealed an acceptable accuracy (error < 5%).

Keywords: analytical modeling; artificial neural network systems; binary polymer blends; Kolarik’s model; mechanical properties; tensile modulus.

1 Introduction

The manipulation of the mechanical properties of polymer materials using different processes such as the production of polymer blends/nanocomposites and blend-based nanocomposites has been always considered as a very efficient method for obtaining new structures with specifically required characteristics [1–9]. Simultaneously, it is completely accepted that mathematical modeling can definitely enhance the practicality and efficiency of the mentioned processes via providing the possibility of predicting the final properties of a sample even before performing the process itself [6, 9–15]. Tensile modulus is one of the mechanical parameters, which describe the resistance of a material (e.g., polymers, metal, ceramics, etc.) against the exerted stress before deforming and therefore it has a very crucial and important role in those structures which preferably should not be deformed under any circumstances [16–18]. So far, many analytical and numerical models have been proposed for predicting the tensile modulus of polymer blends/nanocomposite considering the effects of different parameters such as the morphological variation, polymer/polymer interface and percolation thresholds [5, 7, 15, 19–23].

In the case of binary polymer blends, the Kolarik’s cross orthogonal skeleton model (COS) is one of the most well-known models which is designed based on the interaction between the dispersed and continuous polymer phases as series and parallel sections [14, 15, 24]. However, the lack of a specific section corresponding to the droplet-matrix morphology in the structure of COS model has been always considered as a major drawback which is somewhat corrected by Wang et al. [19] They describe the morphological variation in four different intervals, as a function of the variation of the dispersed phase, and the improvement of COS model is performed by adding the Maxwell-Eucken component to the main geometrical structure. As a very good achievement, the final model (knotted and interconnected skeleton structure, KISS) is capable of altering its structure in accordance with the morphological variation of the systems.

Despite it is clear that each step of the improving process has made the Kolarik’s model more and more accurate and practical but there is still a very significant and common defect in the applied analytical modeling procedure (Figure 1). Generally, COS model is designed based on the interactions of the series and parallel parts in a geometrical structure which is also the main modeling concept in its improved forms. This means each considered parallel/series part of the dispersed phase should be evaluated based on its interaction with the corresponding parallel/series section of the continuous phase and vice versa. Though, this simple rule was not followed in KISS model because it is impossible to define the parallel section
of the continuous phase after the combination of the geometrical structures of COS and Maxwell-Eucken models. Moreover, there is no section representing the polymer/polymer interface region in the geometrical structure of the model. As an effort for solving this problem, we proposed a method by which it was possible to combine the results of COS and Maxwell-Eucken models rather than their geometrical structure in order to predict the tensile modulus of binary polymer blends [25]. This was done using a consistency parameter defined based on the fundamentals of the percolation theory [26, 27]. However, the results were acceptably accurate but the direct interaction between the droplet-matrix and cocontinuous morphologies was not considered in the modeling procedure and the polymer/polymer section was still neglected. Consequently, in another study, we evaluated the effects of the polymer/polymer interface on the prediction accuracy of the improved form of KISS model study for predicting the tensile strength of binary polymer blends [6]. The characteristics of the polymer/polymer interface region were defined using an artificial neural network which was capable of interpreting some important parameters from experimental results.

As a new approach, in this work, a new model was proposed based on a completely new geometrical structure in which the polymer/polymer interface played an important role in increasing the accuracy of the final results. The characteristics of the interface region were defined using a predesigned artificial neural network (ANN) system [6]. Moreover, the resulted framed model (FM) was capable of altering its geometrical structure in accordance with the blend morphology. Unlike KISS model, all corresponding parallel and series sections of the geometrical structure in FM model were separable and directly considered in the modeling procedure. Furthermore, the percolation theory was used in order to define the effects of the dispersed phase droplets on the final mechanical properties of the blend after percolation thresholds. As the next step, the accuracy of FM model was evaluated by comparing its predictions with the tensile test results of some prepared isotactic polypropylene (iPP)/PA blend samples and also other data from literature which showed the maximum error less than 5%. Furthermore, the performance of FM model was compared with some similar models [COS, 3 perpendicular plates (3PP), DIA and symmetrical approximation system (SAS) models] in order to prove its better performance in predicting the tensile modulus of binary polymer blends [21, 24, 25].

2 Modeling background

Figure 1 illustrates the geometrical structure of some of the well-known models capable of predicting the tensile modulus of binary polymer blends with an acceptable accuracy. In our previous work, we added an extra polymer/polymer interface region to the SAS model coupled it with an artificial neural network system (ANN) for predicting the tensile strength/modulus of binary polymer blends (Figure 1c, d). Consequently, it was proved that the application of an ANN together with an added polymer/polymer interface region can play a decisive role in increasing the accuracy of the model. Though, it seemed that the lack of some necessary parts in the geometrical structure of the model was still the main reason for its incompatibility with the actual systems [6]. As mentioned before, the Kolarik’s model is designed based on the interaction of the corresponding series and parallel parts of a considered geometrical structure which is unintentionally ignored in KISS model and its improved forms [6, 19, 21]. In fact, as it is clear in Figure 1b, the combination of COS and Maxwell-Eucken models caused some fundamental changes in the geometrical structure of both models and drastically affected their functionality [14, 28].

As it is illustrated in Figure 2, the geometrical structure of the Kolarik’s model (Figure 1a) was reformed to a simple frame which made it possible to use the intact structure of the Maxwell-Eucken model as one of the main modeling components. It should be noted that, the considered polymer/polymer interface in the modeling procedure was not illustrated in Figure 2 in order to avoid unnecessarily crowdedness. Similar to our previous studies, four different
intervals plus phase inversion point were considered for evaluating the morphological variation of the system as a function of the volume fraction of the dispersed phase:

(I) Droplet-matrix morphology in 1st and 4th intervals.
(II) Cocontinuous morphology at phase inversion point
(III) The combination of droplet-matrix and cocontinuous morphologies in 2nd and 3rd intervals.

2.1 Modeling at phase inversion point ($\phi_p$)

The model structure at the phase inversion point simply consists of a frame with unit side length made of dispersed/continuous phase.

As it is illustrated in Figure 3, FM model provides all corresponding series and parallel component of both phases. Consequently, the volume fraction of the series parts of phase 1 and 2 at each side of the model structure can be defined as follows:

\[ \phi_{1s|A} = 2a^2 \left(1 - 2a'\right) \]  
\[ \phi'_{1s|A} = 4a^2 \left(L\right) \]  
\[ \phi_{2s|A} = a \left(1 - 2a'\right)^2 \]  
\[ \phi'_{2s|A} = L \left(1 - 2a'\right)^2 \]  
\[ \phi_{in.s|A} = 2L \left(1 - 2a'\right)a \]  
\[ \phi'_{in.s|A} = 2L \left(1 - 2a'\right)a' \]

where $a$ denotes the side length of the square cross section of the series or parallel beams in the model structure and $a'$ is $a$ plus the thickness of the polymer/polymer interface ($L$).

Accordingly, the volume fraction of the parallel components of the model are:

\[ \phi_{1p} = \left(1 - 2a'\right)^2 \]  
\[ \phi_{2p} = 4a^2 \]  

The total volume fraction of the cumulative series elements acting as parallel elements with uniform parallel elements of the structure can be calculated as follows:

\[ \phi_{Ap} = 4 \left(a \left(1 - 2a'\right)\right) \]  
\[ \phi'_{Ap} = 8aL \]  
\[ \phi_{Ap} = 4L \left(1 - 2a'\right) - La' \]

A very small percentage of each polymer phase ($y_p$) is considered to participate in the polymer/polymer interface region [9, 29, 30], therefore:

\[ \phi'_{1} = \phi_{1} \times y_p \]  
\[ \phi'_{2} = \phi_{2} \times y_p \]

where $\phi'_{1}$ and $\phi'_{2}$ are the volume fractions of the participant portions of Phase (I) and Phase (II) in the interface region, respectively, $\phi_{1}$ and $\phi_{2}$ denote the actual (experimental) volume fractions of phases (I) and (II), respectively. It is now possible to calculate parameter $a$ using Eq. (14):

\[ \phi_p (1 - \left(y_p\right)) = \left(12a^2 - 16a'^2\right) \]  

Figure 2: Variation of the geometrical structure of FM model with the blend morphology. $\phi_{1cr}$ and $\phi_{2cr}$ are the percolation thresholds of the continuous and dispersed phase, respectively, and $\phi_p$ defines the phase inversion point.
where \( \varphi_p \) is the volume fraction of Phase (II) corresponding to the phase inversion point.

Considering \( a' = a + L \), the thickness of the interface \( L \) can be calculated using Eq. (16):

\[
12(a'^2 - a^2) - 16(a'^3 - a^3) = (\varphi_1' + \varphi_2')
\] (15)

It should be noted that, \( L \) is assumed to be constant and independent of the volume fraction of the polymer phases.

The next step is to define the analytical equation of the model for predicting the tensile modulus of the system at phase inversion point \( (E_t|_p) \):

\[
E_t|_p = \left( \frac{\varphi_2 E_1}{E_1} + \frac{\varphi_2 E_2}{E_2} + \frac{\varphi_3 E_3}{E_3} + \frac{\varphi_4 E_4}{E_4} + \frac{\varphi_5 E_5}{E_5} \right)^{-1}
\] (16)

The following equations can be used for calculating \( E_A, E'_A \) and \( E''_A \):

\[
E_A = \varphi_2 E_2 + \varphi_1 E_1 + \varphi_{in} E_{in}
\] (17)

\[
E'_A = \varphi_2 E_2 + \varphi_{in} E_{in}
\] (18)

\[
E''_A = \varphi_1 E_1 + \varphi_{in} E_{in}
\] (19)

### 2.2 Modeling in the 2nd and 3rd intervals

As it is shown in Figure 2, the 2nd and 3rd intervals are corresponding to the variation range of the dispersed phase content between the percolation threshold \( (\varphi_{2cr} or \varphi_{1cr}) \) and the phase inversion point \( (\varphi_p) \) in which the blend has both droplet-matrix and cocontinuous morphologies. After the percolation threshold, the interconnection of the droplets (dispersed phase) gradually leads to the formation of the frame (Figure 4) and it does not affect the Maxwell-Eucken component, as the representative of the remained dispersed droplets, at all. This completely solves the discussed problem involved with other related models.
Considering the effects of the polymer/polymer interface, the analytical equation of the model in the 2nd and 3rd intervals \((E_{i2,3})\) can be defined as follows:

\[
E_{i2,3} = \left( \frac{\varphi_s + \varphi_{cent}}{b^y E_{cent}} \right)^{-1}
\]

\[
y = \left( \frac{\varphi_{AP} \varphi_{AP}'}{E_A} + \varphi_{AP} \frac{1}{E_A} \right)^{-1}
\]

where \(b\) is a model parameter, \(\varphi_s\) and \(\varphi_{cent}\) are:

\[
\varphi_{cent} = (1 - 2\alpha)^2
\]

\[
\varphi_s = 1 - \varphi_{cent}
\]

It should be noted that the calculation of parameter \(a\) in the 2nd and 3rd intervals is somewhat different from the applied procedure at the phase inversion point. The interconnection of the dispersed droplets in the actual blend after the percolation threshold causes the formation of some small sections with cocontinuous morphology which is simultaneous with the formation of the frame in the geometrical structure. Therefore, parameter \(a\) was defined as a function of the volumetric amount of the dispersed phase that forms local cocontinuous morphology in the blend.

In our previous works, we introduced a consistency parameter \((F(D))\), based on the percolation theory, which defined a specific amount of the dispersed phase that caused the formation of local cocontinuous morphology in the blend after the percolation threshold [25, 26, 31]:

\[
F(D) = \left( \frac{\varphi_2 - (\varphi_2' + \varphi_{2cr})}{\varphi_2 (1 - (\varphi_p)) - \varphi_{2cr}} \right)^T
\]

where \(\varphi_2\) is the volume fraction of the dispersed phase that varies between percolation threshold and phase inversion point, \(T\) denotes the critical universal exponent and \(\varphi_{2cr}\) is the percolation threshold of the dispersed phase [16, 17]. However, different empirical values have been reported for \(T\) but in this study, it was preferably considered as a model parameter which could take a constant value corresponding to the blend system.

As a result, \(a\) is definable using Eq. (24):

\[
F(D)\varphi_2 = 12a^2 \quad - 16a^3
\]

The tensile modulus of the central section of the model \((E_{cent})\) can be simply calculated using the improved form of Maxwell–Eucken model considering the effects of the polymer/polymer interface as follows (Figure 4) [5]:

\[
E_{cent} = E_{ME} = \frac{E_1\varphi_c + E_2\varphi_d}{\varphi_c + \varphi_d \frac{\lambda_{EM}}{\lambda_{ME}}}
\]

where \(\varphi_c\) and \(\varphi_d\) are:

\[
\varphi_d = \left( \frac{4}{3} \pi R^3 \right)\frac{R = R' + L}{W = (3(\varphi_2 - \varphi_1) - 2a)^{2/3}} \quad \frac{R' = \left( \frac{3(\varphi_2 - \varphi_1) (1 - F(D))}{4\pi} \right)^{1/3}}
\]

As a result, considering a constant thickness of the polymer/polymer interface, \(R\) can be simply calculated.

Figure 5 represents the variations of parameters \(a\) (Equation (24)) and \(R\) (Equation (26)) with the volume fraction of the dispersed phase in the 2nd and 3rd intervals. As it is clear, \(R\) finally approaches 0 as \(\varphi_2\) approaches phase inversion point and therefore the model structure turns into a simple frame with unit side length.

Consequently, \(E'\) can be calculated using the following equation:

\[
E' = \frac{4}{3}\pi \left( E_2R' + E_1 \left( R^3 - R'^3 \right) \right)
\]

2.3 Modeling in the 1st and 4th intervals

Figure 6 illustrates the structure of FM model in the 1st and 4th intervals where there is no interconnection between the dispersed droplets and therefore there is no frame in the geometrical structure. Generally, in these intervals, the increment of the volume fraction of the dispersed phase from 0 to the percolation threshold does not affect the geometrical structure of the model while at the percolation threshold the frame starts to form and grow quickly. As a result, the characterizing equation of the model can be defined using Eq. (25) considering \(\varphi_c = 1 - \varphi_d\).

3 Characterizing the polymer/polymer interface

As it clear, there are two parameters related to the polymer/polymer interface \((\varphi_2\) and \(E_{In}\)) which were significantly important in the modeling procedure. To define these parameters, an ANN was designed using the neural network fitting tool (NF tool) in the MATLAB software whose
capability for interpreting such data from the tensile test results has been already proven \cite{6}. Figure 7 represents the schematics of the applied ANN. Considering error $< 5\%$, $|E_2 - E_1| \leq 2E_{In} \leq |E_2 + E_1|$ and $0 \leq y_p \leq 0.5$, the network trains itself in order to produce the best result with the least error which may require the network to be trained multiple times. At the finals step, the results ($y_p$ and $E_{In}$) can be tested using the model equation in the 1st or 4th intervals to define the best set with the least error. It is very important to mention that, the ANN only uses one tensile test result as the final target by which it is possible to interpret the polymer/polymer interface parameters.

4 Materials and methods

4.1 Materials

PA6 (Tecomid NB40 NL E) and the isotactic polypropylene (iPP) (MFI = 5.0 g/10 min; 216 kg and 230 °C) were purchased from Eurotec Co. and Polynar Petrochemical Co., Tabriz, Iran, respectively.

4.2 Blend preparation

All the materials were dried in a vacuum oven at 80 °C for 12 h prior to melt mixing, in order to minimize the effect of the moisture. An internal mixer (Brabender Plasticorder W50EHT) with a rotor speed of 60 rpm (at 230 °C for 8 min) was used to prepare blend samples with 10, 20, 30, 40 and 50 vol% as the total volume fraction of dispersed phase (PA). The obtained compounds were compression molded into suitable pieces for tensile tests. Molding was carried out at 230 °C followed by slow water-cooling under 10 Mpa.

4.3 Mechanical properties

Tensile tests were conducted on a Zwick/Roell tensile testing machine (Z 010) at a fixed crosshead speed of 5 mm/min at room temperature according to ISO 527. At least five specimens were tested for each composition and the resulting tensile properties were averaged.

5 Results and discussion

The presence of tow consistency parameters ($b$ and $T$, Eqs. (20) and (23), respectively) in the modeling procedure of FM model can be considered as one its significant advantages which drastically increased its compatibility with the actual blend system. It should be noted that these parameters are crucial for evaluating the share of each model section against the exerted stress. As it is shown in Figure 8, parameters $T$ and $b$ directly affect the prediction pattern of the model which should be necessarily as close as possible to the real pattern. To this end, the combination of the DIA model, as one of our previously proposed models for predicting the tensile modulus of binary polymer blends, with FM model was used for defining the best values for parameters $T$ and $b$ \cite{25}. DIA model was generally designed based on the fundamentals of the percolation theory which made it completely reliable for predicting the tensile
modulus of a polymer blend at the vicinity of the percolation thresholds [25, 26, 31]. As a result, parameter \(b\) can be defined using Eq. (28):

\[
b = \frac{\phi_s}{E_A (E_DIA - \xi)}
\]  

(28)

\[
\xi = \frac{\phi_{cent}}{E_{cent}}
\]

\[
E_{DIA} = \frac{\phi_{cos}}{E_{cos}} + \left(1 - \frac{\phi_{cos}}{E_{cos}}\right) E_{ME} 
\]

\[
E_{ME} = \frac{E_1 (1-\phi_2) + E_2 \phi_2}{(1-\phi_2) + \phi_2} \left(1 + \frac{3E_1 (\phi_1 + \phi_2)}{2E_1 + E_2}\right)
\]

\[
E_{cos} = \frac{E_1 \phi_{ip} + E_2 \phi_{ip} + (\phi_{p1} + \phi_{p2})^2}{E_1 + E_2} 
\]

where \(f\) is a model parameter [14]. It is should be also mentioned that the considered volume fraction of the dispersed phase (\(\phi_2\)) in Eq. (28) should be very close to the percolation threshold (\(\phi_{cr}\)). Consequently, parameter \(T\) can be calculated using Eqs. (20) and (23).

In order to evaluate the capability and validity of the proposed model, its predictions were compared with the tensile test results of the prepared iPP/PA samples and also some other data from literature (Figure 9) [21, 25, 28, 32]. As it is illustrated, FM model is completely capable of predicting the tensile modulus of polymer blend systems at different compositions with the least error (less than 5%). Also, the results of the applied ANN are represented in Figure 7.

At the next step, FM model was compared with COS, 3PP, DIA and SAS models (Figure 10a). The result of both approaches of Kolarik’s model (3PP and COS) was almost linear which can be attributed to the application of a simple...
geometrical structure that is not capable of reforming in accordance with the morphological variation. On the other hand, the DIA model, as the combination of COS and Maxwell-Eucken models, acted significantly better due to the involvement of the effects of the morphological variation, using a consistency parameter, in the modeling procedure. Though, it is clear that FM model is acceptably accurate and, unlike other models, is completely capable of following the variation pattern of the actual results.

It is also important to demonstrate the effects of the polymer/polymer interface on the performance of the framed model. To this end, the model equations should be rederived neglecting the parameters related to the interface region \((L = 0\) and \(a = a'\)). Consequently, modeling at the phase inversion point \((\varphi_p)\) results in the following equation:

\[
Et|_{p} = \left(\frac{\varphi_{1p}E_1 + \varphi_{2p}E_2}{\varphi_{Ap}E_A}\right)^{-1}
\]

(29)

where \(\varphi_{1p} = (1-2a)^2\), \(\varphi_{2p} = 4a^2\), \(\varphi_{Ap} = 4a-8a^2\), \(E_A = \varphi_{1p}|E_2 + \varphi_{2p}|E_1\) and \(a\) can be calculated using the following equation:

\[
\varphi_p = (12a^2 - 16a^3)
\]

The same approach can be applied in the 2nd and 3rd intervals as follows:

\[
Et|_{2,3} = \left(\frac{\varphi_{p}E_A}{b\varphi_{Ap} + \varphi_{cent}E_{cent}}\right)^{-1}
\]

(30)

where \(\varphi_{cent} = (1-2a)^2\), \(\varphi_s = 1-\varphi_{cent}\), \(a\) and \(E_{cent}\) can be calculated using Eqs. (31) and (32), respectively.

Figure 9: Comparison of the tensile test results of the different binary polymer blends with the predictions of FM model: (a) iPP/PA blend samples \((b = 4.75, T = 4)\), (b) PA/POE blend samples \((b = 9.72, T = 12)\) [25], (c) PC/ABS blend samples \((b = 4.12, T = 5)\) [28] and (d) PP/PVC blend samples \((b = 4.13, T = 3.7)\) [32].
\[
\left( \frac{\varphi_2 - \varphi_{cr}}{\varphi_p - \varphi_{cr}} \right)^T \varphi_2 = 12a^2 - 16a^3
\]

(31)

\[
E_{cent} = \frac{E_1\varphi_c + E_2\varphi_d\left( \frac{3E_1}{2E_1 + E_2} \right)}{\varphi_c + \varphi_d\left( \frac{3E_1}{2E_1 + E_2} \right)}
\]

(32)

where \(\varphi_d = 4/3\pi R^3\) and \(\varphi_c = (1-2a)^2 - \varphi_d\).

Finally, considering \(\varphi_c = 1-\varphi_d\), Eq. (32) can be used for predicting the tensile modulus of the binary polymer blends in 1st and 4th intervals (Maxwell-Eucken model). To realize the impact of the polymer/polymer interface on the accuracy of the model, a comparison between the experimental results of the prepared iPP/PA samples FM model and latest equations is represented in Figure 10b. As it is clear the, neglecting the effects of the interface region significantly increases the error to at least 5% on top of the prediction error of the FM model which can be attribute to the assumption of the presence of a very strong and perfect polymer/polymer interface. Although, the prediction pattern of the model can still work perfectly which is due to the effects of parameter \(b\).

6 Conclusion

The combination of an artificial neural network system (ANN) and a practical analytical method was used in order to propose a unique model for predicting the tensile modulus of binary polymer blends. Unlike other similar attempts, the proposed structure was capable of reforming in accordance with the morphological variation of the actual blend system and also consisted of all essential corresponding series/parallel sections of both phases as well as the polymer/polymer interface. The characteristics of the interface was defined using a practical ANN which was previously used for predicting the tensile strength of binary polymer blends. As a result, FM model could predict the tense modulus of different blend samples with less error compared to other well-known similar models. Besides, as a significant approach, the involvement of two consistency parameters in the modeling procedure (\(T\) and \(b\)) caused the final prediction pattern of the model very similar to the actual pattern.
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