



An analysis of interfacial adhesion in nanocomposites from recycled polymers



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ABSTRACT

The present paper studies the quantitative analysis of interfacial adhesion in the nanocomposites containing recycled polymers. The interfacial bonding is evaluated using different models for tensile properties of composites. A good agreement is found between the experimental results of mechanical properties and the theoretical predictions which validate the current analysis.

Many parameters such as (a) in Nicolais–Narkis, (a) in Kunori–Geil, (B) in Pukanszky model and interfacial strength (t) show the perfect interfacial adhesion. Moreover, the obtained values of (B) and (t) are compared with other studies. The current study justifies the recycling of polymers through the incorporation of nanofillers.

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1. Introduction

Nowadays, the large amounts of waste polymers threaten the people life. This problem and the other economic and petroleum considerations related to polymers have increasingly motivated the researchers to solve the problem [1,2]. Undoubtedly, the efficient treatment of waste polymeric products is recycling and reusing. However, the major problem in this field involves the degradation of polymer structure in reprocessing which cause much poorer properties [3]. In this regards, researchers have tried to introduce the best modifications that can compensate the loss of properties.

The addition of other components to the waste polymers seems to be the simplest and easiest way for reusing the recycled polymers. The nanofillers present more excellent feature for improvement of all mechanical, thermal and barrier properties [4–7]. They increase the interphase surface of the components that develop the performances through a simply processing technology with low cost. It is interesting to note that the nanofillers can enhance the melt strength of recycled polymers while the inferior melt strength causes an inconsistency of material after leaving the extruder which makes the production of sheets or profiles, impracticable [8]. Further, nanofillers increase the intrinsic viscosity of Poly(ethylene terephthalate) (PET) which is necessary for the reprocessing of PET wastes [9].

Recently, more studies have been carried out on the recycling of polymers such as PET [8,10–12], Polypropylene (PP) [13–16], high

density Polyethylene (HDPE) [17,18] and others through the addition of nanoparticles. In these works, the mechanical properties of nanocomposites were evaluated by experimental characterization. The prediction and modeling of behavior have been carried out for nanocomposites from virgin polymers, while this subject has not been studied for nanocomposites from recycled polymers.

The analysis of behavior provides more information without requiring to a large number of experiments. In other words, the models remove any need to much cost, time and also, difficulties conducted to examination of properties [19–22]. Furthermore, the models facilitate the development of most desirable products. As well known, the best properties of nanocomposites can be obtained when a perfect interfacial adhesion is provided between the matrix and nanofiller [23,24]. However, whether the nanoparticles can provide a good interfacial adhesion and so, an efficient reinforcement in waste polymers or not? On the other hand, whether the mechanical properties of nanocomposites show a strong interfacial bonding between the recycled matrix and nanofiller phases?

In this paper, much attempt is made to answer these important questions through the quantitative analysis of interfacial adhesion by modeling of mechanical properties.

2. Background

In the condition of poor adhesion, the strength of a composite is determined by the available effective region of load-bearing matrix in the absence of filler [25,26]. In this state, the interfacial layer cannot transfer stress and the tensile strength of composite

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depends on the effective load bearing cross-section area portion $(1 - \Psi)$ as:

$$\sigma_c = \sigma_m(1 - \psi) \quad (1)$$

where (σ_m) and (σ_c) are the tensile strength of the matrix and composite, respectively. If (Ψ) is assumed as a power law function of volume fraction of filler (ϕ) , Eq. (1) can be presented as:

$$\sigma_c = \sigma_m(1 - a\phi^b) \quad (2)$$

where (ϕ) is the volume fraction of the filler and (a) and (b) depend on the filler–matrix interaction and the shape and arrangement of particles.

Based on Eq. (2), Nicolais and Narkis [26,27] suggested a model for tensile strength of composites reinforced with spherical particles as:

$$\sigma_c = \sigma_m(1 - a\phi^{2/3}) \quad (3)$$

In the case of good adhesion, the interfacial layer can transfer a small portion of stress while the deformation of matrix is very small. In this case, the tensile strength includes a contribution of both matrix and filler properties. Therefore, the value of parameter (a) in Nicolais–Narkis model becomes smaller than 1.21 which shows the stronger adhesion.

Kunori and Geil [28] related the tensile strength of composites with (a) parameter, which is a stress concentration factor as:

$$\sigma_R = \exp(-a\phi) \quad (4)$$

where (σ_R) is the relative tensile strength as (σ_c/σ_m) . The higher values of (a) indicates to a greater stress concentration.

In the case of good interfacial adhesion, Piggott and Leidner [29] introduced an empirical model including a coefficient of particle–matrix adhesion (a) , as:

$$\sigma_c = K\sigma_m - a\phi \quad (5)$$

The effect of interfacial interaction and filler properties on the tensile strength of composites [30] can be presented as:

$$\sigma_R = 1 + \left(\frac{\alpha t}{\sigma_m} - 1\right)\phi \quad (6)$$

where (α) is the aspect ratio of filler and (t) is the interfacial stress transfer parameter. (α) can be calculated from the developed Halpin–Tsai model for randomly 3 dimensional (3D) platelet fillers [31,32] as:

$$E = 0.49 E_1 + 0.51 E_2 \quad (7)$$

$$E_1 = E_m \left(\frac{1 + \eta\xi\phi}{1 - \eta\phi}\right) \quad (8)$$

$$E_2 = E_m \left(\frac{1 + 2\eta\phi}{1 - \eta\phi}\right) \quad (9)$$

$$\eta = (E_f/E_m - 1)/(E_f/E_m + \xi) \quad (10)$$

$$\xi = 2\alpha \quad (11)$$

where (E_1) and (E_2) are the tensile moduli of the composite in the longitudinal and transverse directions, respectively. (E_m) and (E_f) are the tensile modulus of matrix and filler, respectively.

Pukanszky developed a model based on the spontaneous formation of interphase in composites assuming the variation of tensile strength as a function of composition [33,34]. The Pukanszky model can be presented as:

$$\sigma_c = \sigma_m \frac{1 - \phi}{1 + 2.5\phi} \exp(B\phi) \quad (12)$$

(B) parameter is related to the load carried by the dispersed phase depending on the interaction which can be applied as a quantitative measurement of filler–matrix adhesion. The (B) parameter is shown as:

$$B = (1 + A\rho l) \ln \left(\frac{\sigma_i}{\sigma_m}\right) \quad (13)$$

where (A) is the specific surface area of filler (contact surface), (ρ) is density of filler, while (l) and (σ_i) are the thickness and strength of the interphase, respectively. The Pukanszky model can be reformulated as:

$$\ln \sigma_{\text{Reduced}} = \ln \frac{\sigma_c}{\sigma_m} \frac{1 + 2.5\phi}{1 - \phi} = B\phi \quad (14)$$

According to Eq. (14), when a linear correlation is observed between the reduced tensile strength $(\ln \sigma_{\text{Reduced}})$ and the volume fraction of filler (ϕ) , the model is valid.

Sato and Furukawa [35,36] also suggested a model for tensile modulus of composites containing an adhesion parameter (ζ) as:

$$E = E_m \left[\left(1 + \frac{0.5\phi^{2/3}}{1 - \phi^{1/3}}\right)(1 - \psi\zeta) - \frac{\phi^{2/3}\psi\zeta}{(1 - \phi^{1/3})\phi} \right] \quad (15)$$

$$\psi = \left(\frac{\phi}{3}\right) \left(\frac{1 + \phi^{1/3} - \phi^{2/3}}{1 - \phi^{1/3} + \phi^{2/3}}\right) \quad (16)$$

The (ζ) parameter of 1 shows the poor adhesion, while $\zeta = 0$ indicates to the perfect adhesion.

3. Results and discussion

The tensile strength and modulus of nanocomposites containing various nanofillers such as layered silicate, CaCO₃, carbon nanotube (CNT) and different waste matrices such as PET, PP and HDPE were provided from the literature. Table 1 shows the studied nanocomposites and the attributed references in which all experimental data and other details were given. Accordingly, I refrain from the further discussion of the details.

Table 1 illustrates the calculated parameters from models for different nanocomposites. The (a) parameter from Nicolais–Narkis model is obtained from the linear plotting of experimental tensile strength against $(\phi^{2/3})$. As observed in Table 1, the value of (a) for PP/CaCO₃ nanocomposite is 0.59. It is lower than 1.21 demonstrating the good interfacial adhesion between recycled PP and CaCO₃ nanofiller.

The calculated (a) parameter from Kunori–Geil model (Eq. (4)) is also observed in Table 1. The higher values of (a) which shows the greater stress concentration are obtained in samples No. 7 and No. 8. Other (a) values are much smaller than zero presenting a perfect interfacial adhesion.

Further, the same trend of previous calculations is observed by Piggott–Leidner model (Eq. (5)), where the lowest (a) parameter, indicating to the best interfacial adhesion is obtained for samples

Table 1
The calculated interfacial parameters from different models.

No.	Sample	a (Eq. (3))	a (Eq. (4))	a (Eq. (5))	B (Eq. (12))	Refs.
1	PET/clay	–	–17.63	–23.28	21.45	[10]
2	PET/clay	–	–4.75	–6.83	8.48	[11]
3	PET/clay	–	–3.27	–3.8	6.9	[12]
4	PP/clay	–	–10.77	–15.67	15.6	[15]
5	PP/clay	–	–1.49	–1.52	4.91	[13]
6	PP/CNT	–	–4.35	–7.13	9.3	[14]
7	PP/CaCO ₃	0.59	1.52	1.48	1.74	[16]
8	HDPE/clay	–	1.61	1.61	1.76	[18]

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