

A New Model for Estimation of the Thermal Conductivity of Polymer/Clay Nanocomposites

Ahmad Mohaddespour,¹ Hossein Abolghasemi,¹ Meisam Torab Mostaedi,² Sajjad Habibzadeh¹

¹Department of Chemical Engineering, University College of Engineering, University of Tehran, Tehran, Iran, P.O. Box 11365/4563

²FC Research School, Science and Technology Research Institute, End of North Karegar Ave., Tehran, Iran, P.O. Box 1439951113

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ABSTRACT: High density polyethylene- and polypropylene-clay nanocomposites are synthesized by melt blending, in which polyethylene glycol and polypropylene glycol are used as compatibilizers to increase the space of galleries. The morphology properties of nanocomposites are explored by X-ray diffraction and transition electron microscopy. The thermal conductivity coefficient (K) of nanocomposites is also measured along with the thermal stability. A conventional model based on developed Maxwell-Garnett formula is also established to predict the thermal conductivity of polymer/clay nanocomposites with clay loading. Morphology results indicate that two intercalated and exfoliated structures are formed. The established model satisfactorily predicts the K values of

nanocomposites for low range of clay content. Thermogravimetric analysis shows remarkable thermal stability of nanocomposites with 10 wt % of clay content. The deviation of our model from experimental result for 10 wt % of clay can be attributed to the intercalated structure of layered silicates into the matrices. Although the K values do not considerably increase in 5 wt % with respect to the increase occurs for 10 wt % of clay, but it increases about 28 and 37% at 50°C for high density polyethylene- and polypropylene-clay nanocomposites, respectively. © 2010 Wiley Periodicals, Inc. *J Appl Polym Sci* 118: 1042–1050, 2010

Key words: thermal conductivity; nanocomposite; nanoclay; Maxwell-Garnett formula; modeling

INTRODUCTION

Polymer/clay nanocomposites have been of paramount importance in recent years on account of their remarkable thermal and mechanical properties.^{1–3} Synthesizing of these nanocomposites is generally based on the use of a low concentration of expandable smectite clays, such as montmorillonite (MMT), into the matrix. The surface of layered silicates is not undeniably compatible with a polymer with nonpolar chains. The modification of this blend can be achieved by treatment on layered silicates or polymer chains as well as considering both of them simultaneously. The addition of organically modified layered silicates (OMLS) into a polymer matrix results in different clay distributions that do not always provide an improvement in performance, since the modification of the layered silicate depends on the properties of modifying groups, such as length, head structures, and cation exchange capacity.⁴ Among the different structures of layered silicates into the polymer matrices are inter-

calated, partially intercalated-exfoliated, and fully exfoliated.⁵

The thermal properties of composite materials have been of considerable attention in literature, but the thermal conductivity of these materials seems to hold more increasable potential for research. The thermal conductivity behaviors of some composite materials such as carbon fiber/polymer composites⁶ and metal powder/polymer composites⁷ have been established in recent years. The thermal conductivity properties of nanostructure materials are almost unknown owing to their novel adventure in science arena. In recent years, a few researchers have investigated the thermal properties of nanostructure materials especially the thermal conductivity. The results of this attempt consider the thermal conductivity of carbon nanotube buchypapers and their composites.^{8,9} The thermal conductivity of multiwall carbon nanotube/silica-based nanocomposites,¹⁰ single wall carbon nanotubes in alumina-based nanocomposites,¹¹ nanofiber/epoxy resin nanocomposites,¹² single wall carbon nanotube/polyethylene nanocomposites,¹³ epoxy/carbon multiwalled nanotube nanocomposites,¹⁴ poly(L-lactide)/multiwalled carbon nanotube nanocomposites,¹⁵ CuO/SiO₂ and NiO/SiO₂ nanocomposites,¹⁶ and ethylene vinyl acetate copolymer/nanofiller composites¹⁷ have also been regarded recently.

Correspondence to: A. Mohaddespour (a.mohaddespour@yahoo.com).

Polymer/clay nanocomposites belong to the novel materials whose thermal conductivity has not been investigated as considerable as other nanocomposites. The thermal conductivity of N6/clay nanocomposites has been experimentally studied, and the result shows that this characterization increases markedly for intercalated structure while it decreases against the clay content for fully exfoliated structure of layered silicates into the matrix.¹⁸ The clay type used in the mentioned study was natural MMT modified by octadecyl diethanolamine. The experimental data for thermal conductivity of high density polyethylene (HDPE)- and polypropylene (PP)-clay nanocomposites with intercalated and exfoliated structures reveal a conspicuous augmentation in value by the clay content.¹⁹ Our data presented the K values with different clay contents as well as different temperatures. The results indicated a remarkable increase in thermal conductivity coefficient for both HDPE and PP nanocomposites. We satisfactorily applied polyethylene glycol (PEG) and polypropylene glycol (PPG) as compatibilizer to ameliorate the dispersion of clay layers into the HDPE and PP matrices, respectively. What also presented was the numerical solution of a Partial Differential Equation (PDE) for samples, which their dimensions were adapted to some particular applications of these two matrices. In fact, by the use of experimental data for thermal conductivity coefficient of HDPE- and PP-clay nanocomposites, we predicted the temperature distribution in synthesized samples.

In this work, we do not aim to present the experimental data for K values, but we establish a simple model to understand the thermal behavior of HDPE- and PP-clay nanocomposites based on a conventional model. Other studies predicted this behavior for carbon nanotube based composites.²⁰ Finally, the validation of the presented model is explored by the comparison of our experimental data for K values with the predicted amounts resulted from the model. This exploration can be considered as the first model, which predicts the thermal conductivity of polymer/clay nanocomposites for low range of clay content.

EXPERIMENTAL

Materials

The main matrices of synthesized nanocomposites are HDPE (5818, density 0.93 g/cm^3 , MFI 5 g/10 min) and PP (10800, density 0.91 g/cm^3 , MFI 8.5 g/10 min) purchased from Bandar Imam Petrochemical Co. (Bandare Imam, Iran). PEG (density 0.91 g/cm^3 , MFI $1\text{--}1.5 \text{ g/10 min}$ at 230°C , maleic anhydride $0.5\text{--}0.7 \text{ wt } \%$) and PPG (density 0.92 g/cm^3 , MFI 5 g/10 min at

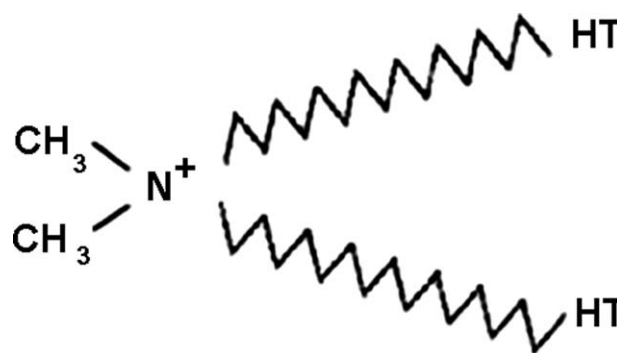


Figure 1 Structure of the quaternary ammonium salt.

230°C , maleic anhydride $0.5\text{--}0.7 \text{ wt } \%$) are provided from Kimia Javid Sepahan Co., Iran, as compatibilizer incorporated into the matrices to enhance the dispersibility of silicate layers into the matrices. MMT with cation exchange capacity about 125 meq/100g that was provided by Southern Clay Products (Gonzales, Texas, USA) is chosen as the nanoscale part of polymer nanocomposites, which is organically modified by alkylammonium salt called quaternary ammonium salt (2M2HT dimethyl, dihydrogenated tallow, quaternary ammonium where HT is hydrogenated tallow ($65\% \text{ C18}$, $30\% \text{ C16}$, $5\% \text{ C14}$, anion: chloride), Fig. 1).

Processing and compatibilizer incorporation

For synthesizing HDPE and PP nanocomposites, which clay as a layered silicate is applied, the most convenient method can be described as the melt blending, by which the polymers are heated and mechanically mixed while the layered silicate is added into the polymer blends, which have formerly been heated to the annihilation point. In this study, HDPE and PEG are fed into the chamber by the volume of 50 cm^3 in which a twin screw by the radial speed of 70 rpm for 10 min holds the responsibility of rendering the mixture homogenous. The temperature of the mixture is controlled to be fixed at 160°C . First, PEG is added into the chamber to be blended well with melted HDPE and then organically modified montmorillonite (OMMT) is gradually added into the mixture to reduce the probability of having layered silicates agglomerated. The clay percent into the polymer blend is set to be 0 , 5 , and $10 \text{ wt } \%$ while the PEG amount is fixed at $15 \text{ wt } \%$. The synthesized samples are melted and also molded again to achieve $20 \times 20 \times 1 \text{ cm}^3$ sheets to be adapted for thermal conductivity measurement. The same process is exactly employed for preparing PP/PPG/OMMT nanocomposites.

The thermal conductivity coefficient of samples can be calculated by eq. (1) with measuring the amount of slope, K , directly:

$$q_x'' = -K \frac{\partial T}{\partial x} \quad (1)$$

Where q'' is the heat flux (W/m^2), K is the thermal conductivity coefficient (W/mK), and $\partial T/\partial x$ is the temperature gradient (K/m).

For the estimation of thermal conductivity, sampling time is fixed at 60 s. This defines the period in which the data exchange with the measuring system takes place. The current control and process quantities are determined and displayed at the end of this cycle. The sample mean temperatures are the temperatures present at measuring points across the sample during a period of time. A number of 1 to 5 temperatures can be preset for automatic operation of the measurement system. These temperatures should preferably be set at $10^\circ C$ intervals. The temperature difference between the cooling and heater plates relates to the sample mean temperature and must be at least $10^\circ C$. In fact, for estimating the thermal conductivity at a certain temperature, the temperature gradient of about $10^\circ C$ is set between the cold and hot sides. The average value of those two temperatures (cold and hot) is considered as the process temperature, and the thermal conductivity is subsequently estimated at this temperature. For example, for estimating this factor at $20^\circ C$, the temperature of cold side is $15^\circ C$ and its corresponding value of hot side is $25^\circ C$. With measuring the required heat flux to steadily hold this temperature gradient between two sides of the sheet, the K value can be estimated from eq. (1). Thermal conductivity at other three temperatures ($30, 40, 50^\circ C$) is measured likewise.

Instruments

In this work, Brabender (Plasti-Corder® Lab-Station, Germany) is employed for melt blending process of polymer nanocomposites. X-ray diffraction analysis (XRD) and transition electron microscopy (TEM) are also applied for estimating the morphology properties of nanocomposites. XRD experiments are appraised at room temperature by a Philips X'Pert X-ray diffractometer (40 KV, 40 mA) with Cu ($k = 0.154$ nm). Bright field TEM images are obtained at 120 kV, at low-dose conditions, with a Phillips 400T electron microscopy. The samples are ultramicrotomed with a diamond knife on a Leica Ultracut UCT microtome at room temperature to provide 70-nm-thick section. The section is transferred from water to carbon-coated Cu grids of 200 mesh. Regarding the distinct contrast between the layered silicate and the polymer phase for imaging, no heavy metal staining of sections is required. The thermal conductivity coefficient of samples is determined by calculating the slope of K in eq. (1) with

TCA200 instrument (Tarus Co., Germany). Finally, thermogravimetric analysis (TGA) for estimating the thermal stability of all samples is performed on a STA 1500 unit, under 30–35 mL/min argon flow. The temperature is first at $25^\circ C$ and then is raised to $600^\circ C$ at a scan rate of $20^\circ C/min$.

RESULTS AND DISCUSSION

Morphology properties

As can be seen in Figure 2(a), the diffraction peak for OMMT appears at 7.19° of 2θ . The intercalation structure can be observed when the corresponding peak appears in lower angles of 2θ for synthesized nanocomposites. Figure 2(c,e) represents the 5.25° and 5.07° of 2θ for HDPE and PP nanocomposites with 10 wt % of clay content, respectively. The structure of these nanocomposites is considered as an intercalated, in which the distance between the clay layers have increased but not the same as an exfoliated structure. This structure for 10 wt % of clay content can be attributed to the agglomeration of some clay layers in comparison with the lower amounts of clay content into the polymer blends. An

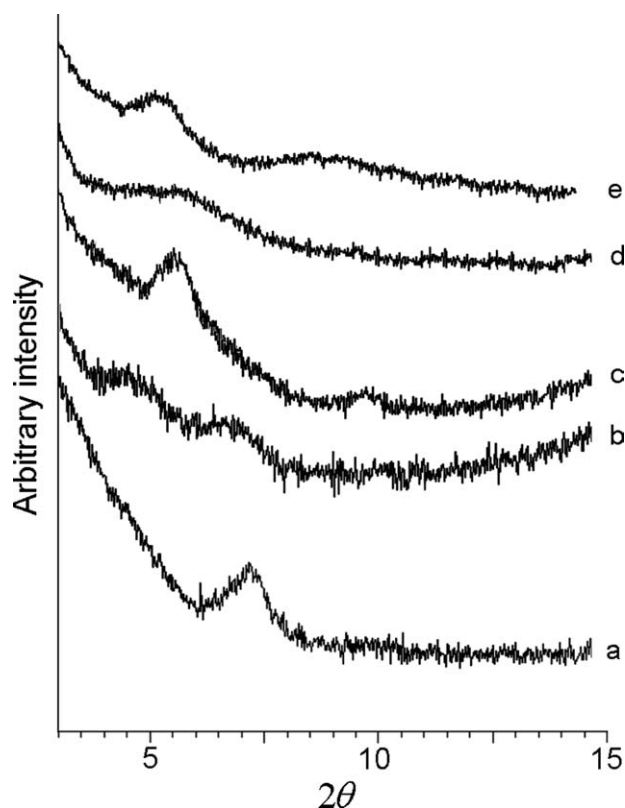


Figure 2 XRD patterns for (a) pure montmorillonite, (b) HDPE/PEG/OMMT 5 wt % nanocomposite, (c) HDPE/PEG/OMMT 10 wt % nanocomposite, (d) PP/PPG/OMMT 5 wt % nanocomposite, and (e) PP/PPG/OMMT 10 wt % nanocomposites.

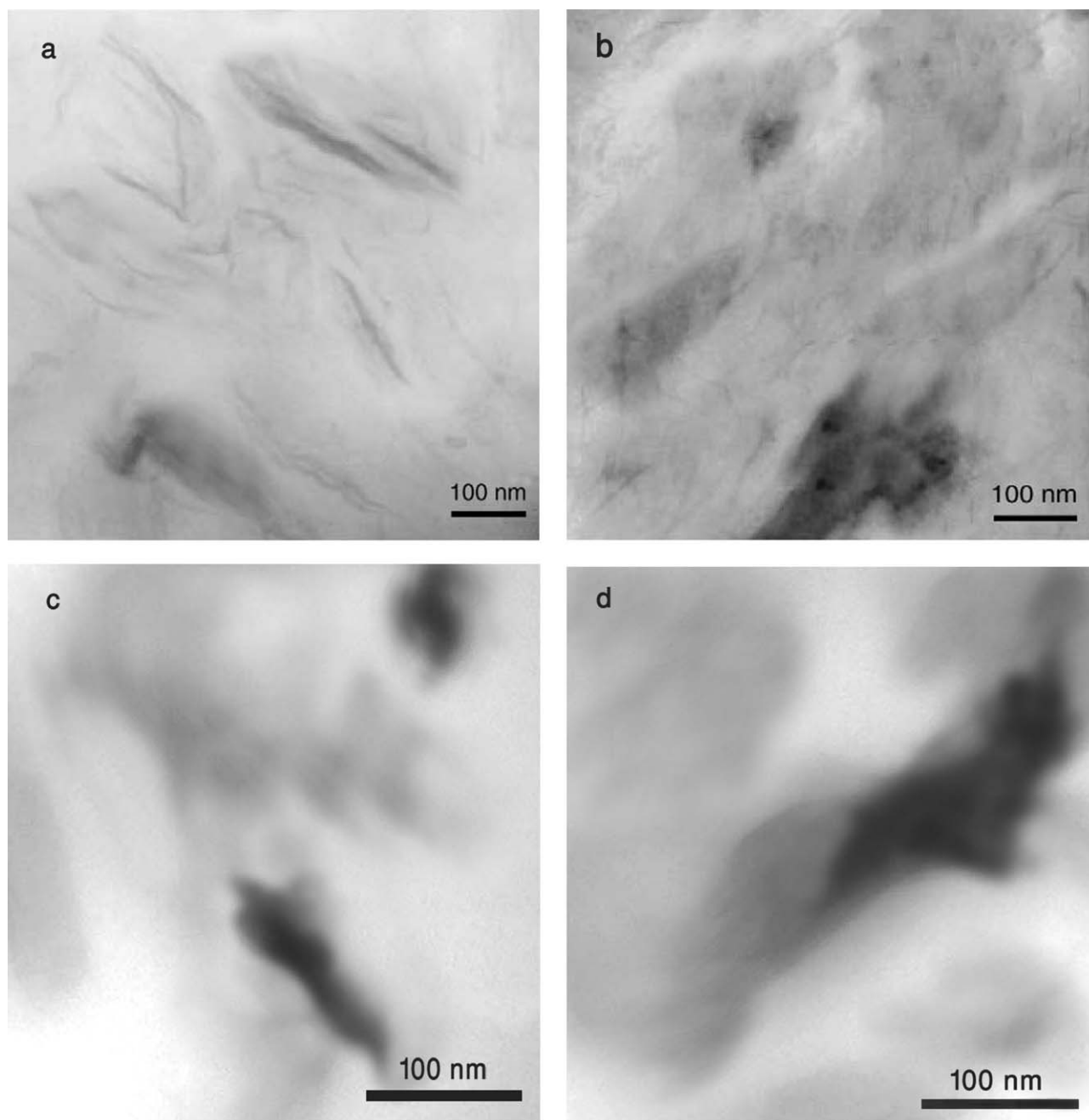


Figure 3 TEM images for (a) HDPE/PEG/OMMT (5 wt %) nanocomposite, (b) HDPE/PEG/OMMT (10 wt %) nanocomposite, (c) PP/PPG/OMMT (5 wt %) nanocomposite, and (d) PP/PPG/OMMT (10 wt %) nanocomposites.

exfoliated structure can be perceived as an overall result of slightly smooth curves of XRD patterns, in which the corresponding peaks of OMMT have been nearly disappeared. The hydrophilicity of HDPE and PP grafted with PEG and PPG, respectively, and the chain length of the organic modifier in the clay structure completely impact the extent of exfoliation and intercalation of layered silicates. Although in the high clay content, where the layered silicates find less considerable space to be dispersed, layered silicates show more tendency to aggregate between the galleries, but the addition of compatibilizer has a

significant effect on smooth dispersion of layered silicates due to the bonds in which one of the endings of compatibilizer's chain holds the responsibility for increasing the space between the layers into the gallery. As a result, the structure in 10 wt % of clay in our samples is observed as intercalated.

Figure 3 displays TEM images of HDPE and PP nanocomposites. As it can be inferred, the addition of PEG and PPG into the polymer matrices improves the dispersibility of clay layers. In reverse, the addition of clay by 10 wt % results in agglomeration of some layers as the space of galleries is not

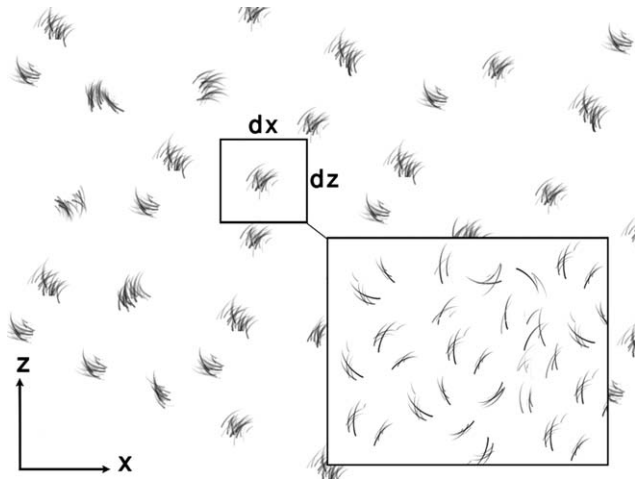


Figure 4 A simple schematic describing the condition for eq. (5).

considerably increased by complete intercalation of polymer chains. Therefore, the structure of samples containing 10 wt % of clay is rather intercalated than exfoliated, what is acquired by individual dispersion of clay layers into the HDPE and PP matrices. By taking this point into account, we are able to concentrate on the homogeneous modeling of all nanocomposites and presume that the thermal conductivity coefficient is similar in all directions for synthesized nanocomposites (Fig. 4).

Mathematical modeling of polymer/clay nanocomposites

There are various effective medium approaches (EMA) like the Maxwell-Garnett (MG) approximation to analyze the thermal transport behavior in heterogeneous media such as thermal conductivity of some composite structures.^{21,22} The validity of the conventional EMA has been proved, and the MG-EMA has been known to be reasonable for matrix-based composites with small filling ratios. The purpose of this study is to present a new model based on developed EMA formula for polymer/clay nanocomposites media and compare the results with our recent experimental data to determine the range of validity of the conventional theory for nanoclay-based materials.

By reviewing the multiple scattering approach of Nan,²¹ we consider a variable thermal conductivity of composite medium from point to point. Regarding this variation, thermal conductivity of the media is represented in the form of $K(\psi) = K_1 + K_2(\psi)$, where K_1 indicates a constant part of a homogeneous medium and $K_2(\psi)$ is an arbitrary fluctuating part. If we use the Green function, G , for the homogeneous medium defined by K_1 and the transition matrix, T , for the entire nanocomposite medium, the temperature gradient distribution can be acquired:

$$K_n = K_1 + \langle T \rangle (I + \langle GT \rangle)^{-1} \quad (2)$$

Where I is the unit tensor and $\langle \rangle$ denotes spatial averaging while the matrix, T , is described as:

$$T = \sum_i T_i + \sum_{i,j \neq i} T_i G T_j + \dots \quad (3)$$

Where the first term is the sum of the T matrices of i particles and the successive terms denotes the interaction between particles. As the accurate calculation of T is demanding, a simple calculation by neglecting the interparticle multiple scattering can be achieved by general approximation as:

$$T \cong \sum_i T_i = \sum_i K_{1,i} (I - G K_{1,i})^{-1} \quad (4)$$

By correctly considering of three factors; (1) random dispersion of layered silicates into the matrix, (2) effective interface between matrix and layered silicates in nano dimension for the energy transportation across the bulk of the samples, and (3) taking the matrix phase as a homogeneous medium, we can have: $K_1 = K_p$, where K_p is the thermal conductivity of the polymer. These assumptions result in a MG type EMA of the theory that can be described as follows:

$$\frac{K_n}{K_p} = \frac{3 + 2f[\beta_x(1 - L_x) + \beta_z(1 - L_z)]}{3 - f(2\beta_x L_x + \beta_z L_z)} \quad (5)$$

$$\beta_x = \frac{K_x - K_p}{K_p + L_x(K_c - K_p)}$$

$$\beta_z = \frac{K_z - K_p}{K_p + L_z(K_c - K_p)}$$

where K_x and K_z are the thermal conductivities of the layered silicates along transverse and longitudinal axes, respectively; K_c is the thermal conductivity of layered silicates; f is the volume fraction of layered silicates; and L_x and L_z are geometrical factors dependent on the layered silicates aspect ratio, r , and given by:

$$L_x = \frac{r^2}{2(r^2 - 1)} - \frac{r}{2(r^2 - 1)^{3/2}} \cosh^{-1} r \quad (6)$$

$$L_z = 1 - 2L_x$$

For layered silicates in nano dimension, the aspect ratio is around 100 that for high r over than 100 we have $L_x = 0.5$ and $L_z = 0$. As a result, eq. (5) is reduced to:

$$\frac{K_n}{K_p} = \frac{3(K_x/K_p + 1) + f[2(K_x/K_p - 1) + (K_x/K_p + 1)(K_z/K_p - 1)]}{3(K_x/K_p + 1) - 2f(K_x/K_p - 1)} \quad (7)$$

TABLE I
Thermal Conductivity Coefficient of HDPE and PP
Nanocomposites at Different Temperatures

Nanocomposite	T (°C)	Clay (wt %)	K (W/mK)	
HDPE	20	0	0.1509	
		5	0.1792	
		10	0.2957	
	30	0	0.1649	
		5	0.1952	
		10	0.3267	
	40	0	0.1789	
		5	0.2112	
		10	0.3577	
	50	0	0.1929	
		5	0.2272	
		10	0.3887	
	PP	20	0	0.1054
			5	0.1330
			10	0.2139
30		0	0.1184	
		5	0.1490	
		10	0.2389	
40		0	0.1314	
		5	0.1650	
		10	0.2639	
50		0	0.1444	
		5	0.1810	
		10	0.2889	

The thermal conductivity of layered silicates like MMT is mostly controlled by water content, although, obviously, the type of soil is also important. For the clay we used, the thermal conductivity in W/mK is 1.0 for moisture of 10%. We can assume that the thermal conductivity coefficient of layered silicate segments (Fig. 4) is the same in both x and z directions as they are randomly dispersed into the polymer matrix. So, we have:

$$\begin{aligned}
 K_x &= K_z \\
 \frac{K_n}{K_p} &= \frac{3 + f(2K' + K'')}{3 - 2fK'} \quad (8) \\
 K' &= \frac{K_c - K_p}{K_c + K_p} \\
 K'' &= K_c - K_p
 \end{aligned}$$

To derive the eq. (8), it is presumed that $K_z = K_c$ as we formerly proved K_x , K_z , and K_c are all the same for polymer/clay nanocomposite systems with

random dispersion of nanolayers; moreover, the effect of temperature on the thermal conductivity coefficient of polymer matrices can be included in eq. (8) as the following:

$$K_p(T) = K_p + MT \quad (9)$$

Where K_p is the thermal conductivity coefficient of pure polymer at room temperature and M is the slope of a line obtained by drawing $K_p(T)$ values with temperature. eq. (8) can be simply given by:

$$\frac{K_n}{K_p} = 1 + Kf \quad (10)$$

$$K = 2K' + K''$$

Equation (10) is derived by eliminating the term $2fK'$ as the volume fraction of clay in polymer/clay nanocomposites are usually less than 0.05. The thermal conductivity coefficient of all nanocomposites in different temperatures is given in Table I. The results from eq. (10) and their comparison with our experimental data are given in Figure 5. As seen, the simple equation derived from the conventional model predicts the thermal conductivity coefficients of both HDPE and PP matrices. The effect of temperature on the coefficients is also observed; in addition, Figure 6 indicates that the presented model can predict the coefficient for high ratio of K_c/K_p . Other models are just valid for nearly spherical particles ($r \approx 1$), but not for systems like polymer/clay nanocomposites ($r \gg 1$).²³

The model presented here can be used merely for matrix-based composites, in which layered silicates are surrounded by the matrix either with compatibilizer or without it. The MG-EMA formula is for very low range of layered silicates into the matrix in which the layers are randomly dispersed into the matrix so that the results are diverted from the real amounts for continuous networks.

The established model in this study can satisfactorily predict the thermal conductivity coefficient of HDPE and PP nanocomposites for low range of clay content into the matrices. In general, the thermal conductivity for an exfoliated structure decreases in comparison with an intercalated structure. Instead, this coefficient remarkably increases by the clay content. This increase is amplified whatever the clay content is raised. A similar observation has been recognized for N6/clay nanocomposites.¹⁸ In fact, the Maxwell model can not correctly predict the thermal conductivity for high clay content as well as an

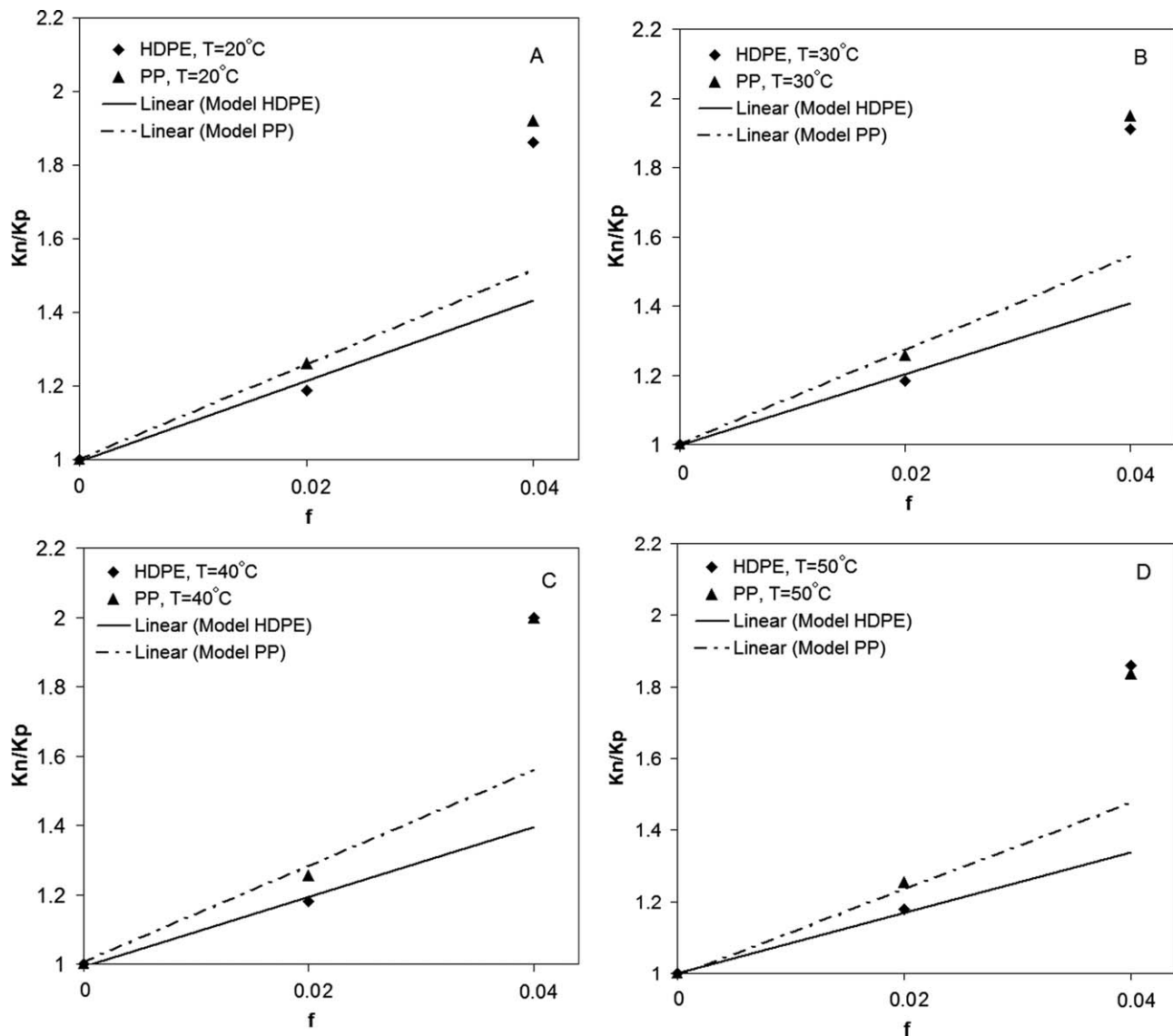


Figure 5 The comparison of K_n/K_p derived from the established model and the experimental data at (a) 20°C, (b) 30°C, (c) 40°C, and (d) 50°C.

intercalated structure of polymer/clay nanocomposites where the layered silicates indicate more appetite to be accumulated.

Thermogravimetric analysis

Elaborating on the current observation for better understanding of the natural effect of nanoclay on the thermal properties of polymer nanocomposites along with the thermal conductivity, we should first balance two diverse effects of clay layers on the polymer matrix; first, the layered silicates hold a barrier action in the matrix because of the mineral essence of layers by which the thermal stability of polymer nanocomposites is enhanced; and second, in reverse, the alkylammonium cations in the organoclay could suffer decomposition following the

Hofmann elimination reaction whose products would catalyze the degradation of polymer matrices. The TGA results for nanocomposites with 5 and 10 wt % of clay are shown in Figure 7. The balance between those two mentioned factors will determine the total effect of layered silicates into the polymer matrices. Concentrating on the thermal stability of polymer/clay nanocomposites provides a better understanding of nanocomposites' thermal properties so that we can compare our observation with what occurs for thermal conductivity to ascertain the effect of layered silicates on overall thermal properties.

As can be seen in Figure 7, the thermal stability of both HDPE and PP nanocomposites increases with the clay content, and the amount of clay in this property has a positive effect. The barrier action of

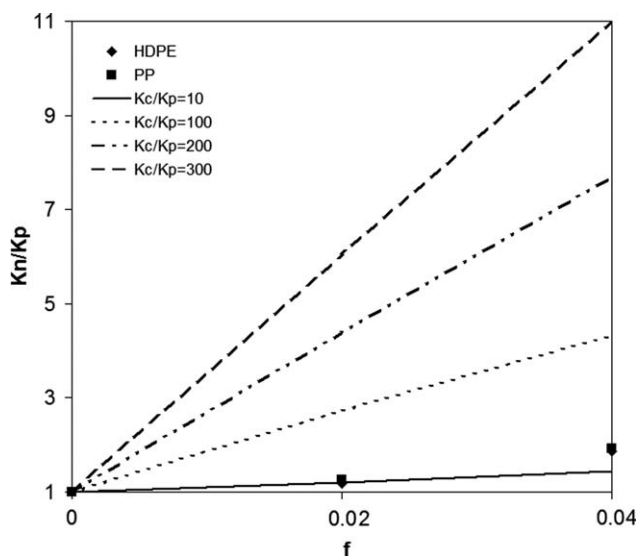


Figure 6 The prediction of eq. (10) for higher thermal conductivity of filler into the polymer matrix.

layered silicates has been the dominant factor, which has improved the thermal stability; furthermore, the products of Hofmann elimination reaction have not been as much considerable as the barrier action. The

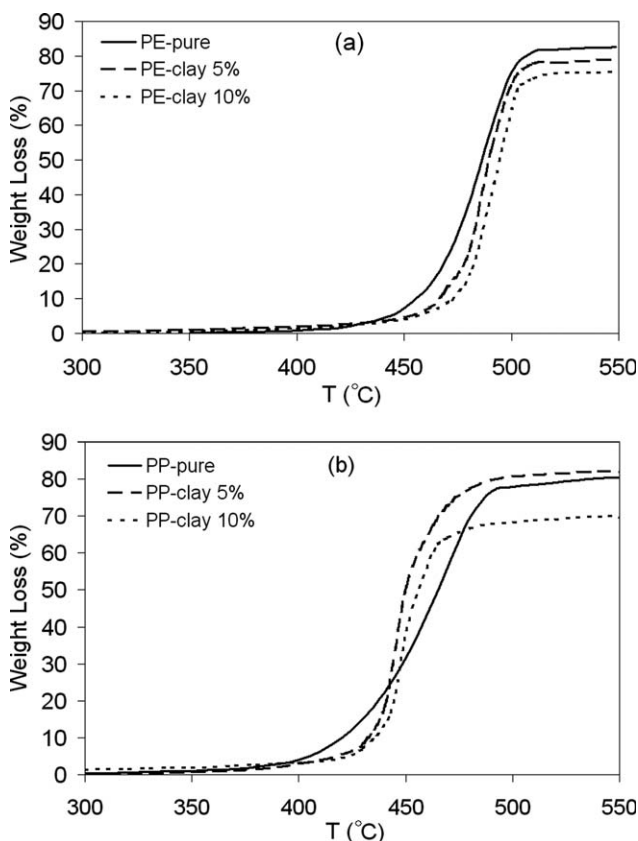


Figure 7 TGA results for (a) HDPE/PEG/OMMT nanocomposites and (b) PP/PEG/OMMT nanocomposites with 5 and 10 wt % of clay content.

TABLE II
TGA Result for PE and PP Nanocomposites

Samples	$T_{0.1}$ (°C)
PE- pure	454.80
PE- clay 5%	465.63
PE- clay 10%	472.62
PP- pure	420.16
PP- clay 5%	432.56
PP- clay 10%	436.70

$T_{0.1}$, the temperature in which the weight loss is 10%, for pristine polymers and their nanocomposites is presented in Table II. This factor increases about 1.72 and 3.91% for HDPE nanocomposites with 5 and 10 wt % of clay, respectively. This increase is observed about 2.95 and 3.93% for PP nanocomposites with 5 and 10 wt % of clay, respectively.

CONCLUSIONS

In this study, the OMMT was added into the HDPE and PP matrices which were incorporated with PEG and PPG as compatibilizer, respectively. The melt blending was used to synthesize all nanocomposites. The intercalated and exfoliated structures were homogeneously achieved as can be perceived from the XRD and TEM results. The addition of two compatibilizers into their own matrices favorably improved the dispersion of layered silicates into the matrices. A novel model was derived from an EVA formula to predict the thermal conductivity coefficient of HDPE and PP nanocomposites. This model presented excellent results for low range of clay content into the both matrices. It was also based on homogeneous dispersion of layered silicates and showed a deviation to predict the K values for higher clay contents. The K values increased remarkably for nanocomposites with 10 wt % of clay owing to their intercalated structure. The presented model could also predict the coefficient for high ratio of K_c/K_p ; nevertheless, this model can be solely used for matrix-based composites with low range of layered silicates. The TGA result indicates that the nanocomposites with 10 wt % of clay show a distinct improvement in thermal stability with respect to their pure matrices. This improvement is observed in $T_{0.1}$ which increases about 1.72 and 3.91% for HDPE nanocomposites and about 2.95 and 3.93% for PP nanocomposites with 5 and 10 wt % of clay content, respectively. Consequently, the addition of layered silicates into the HDPE and PP matrices remarkably improves the thermal conductivity and thermal stability of their nanocomposites in the high clay content due to the intercalated structure. The incorporation of the OMMT into the HDPE and PP matrices significantly impacts the thermal behavior

of synthesized nanocomposites due to the particular arrangement of layered silicates which more plays as a barrier action and shows a fair conductivity with the stacks of intercalated layers.

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