

Isothermal Crystallization Kinetics of Kevlar Fiber-Reinforced Wood Flour/High-Density Polyethylene Composites

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ABSTRACT: Kevlar fiber [KF; poly(*p*-phenylene terephthalamide)] can be used as a reinforcing element to improve the mechanical properties of the resulting wood thermoplastic composites. This study was devised to investigate the effects of incorporation of KF on the isothermal crystallization kinetics of high-density polyethylene (HDPE) in the resulting composites using differential scanning calorimetry. Avrami model was applied to describe the isothermal crystallization process, and the fold surface-free energy was calculated according to the Hoffman–Lauritzen theory. Comparative study of neat HDPE, wood flour/HDPE composite (WPC), virgin KF-reinforced WPC (KFWPC), and grafted Kevlar fiber (GKF)-reinforced WPC (GKFWPC) showed that the overall crystallization rate, the activation energy, the equilibrium melting temperature, and the fold surface-free energy of the

WPC were apparently changed due to the addition of KF; the crystallizability exhibited an order of KFWPC > GKFWPC > HDPE > WPC. The incorporation of virgin KF may cause the heterogeneous nucleation to induce a change in the crystal growth of HDPE from the tridimensional to bidimensional or to the mixed patterns. Avrami exponent values of the composites decreased with time, confirming the change of crystallization behavior. This study demonstrates that both the KF and GKF can act as nucleating agents to improve the crystallization rate of HDPE. © 2012 Wiley Periodicals, Inc. *J Appl Polym Sci* 000: 000–000, 2012

Key words: wood plastic composites; high density polyethylene; Kevlar fiber; isothermal crystallization; nucleating agent; crystallization rate

INTRODUCTION

Wood plastic composites (WPCs) have been gaining increasing applications in building, automotive, and infrastructure fields in light of their unique material properties and high environmental friendliness.¹ However, the insufficient mechanical properties of WPCs, due to the inherent incompatibility between hydrophilic wood and hydrophobic polyolefin matrix, impede their applications as structural and engineering materials. Consequently, efforts have been made such as use of coupling agents to improve the interfacial compatibility and adhesion,^{2,3} hybridiz-

ing synthetic fibers as reinforcing elements,⁴ incorporating elastomers to improve the impact strength.⁵

For composites based on semicrystalline polymer matrices, their crystallization behavior has a predominant effect on the ultimate properties of the resulting composites.⁶ Addition of reinforcing fiber can result in considerable change of both crystal morphology and crystallinity of thermoplastic polymer matrices,^{6,7,8–12} and so it is of great importance to investigate the crystallization behavior of fiber-reinforced polymer composites. The study on the crystallization of four different types of flax fiber (green flax, dew retted flax, Duralin[®]-treated flax, and stearic acid-sized flax) reinforced polypropylene (PP) composites has shown that the fiber surface roughness can affect the morphology of the PP transcrystalline layer, which may significantly improve the flax-PP interfacial bonding.⁸ In the case of wood flour (WF) as the reinforcing element, the crystallinity of polymer matrices highly depends on wood species, size, chemical composition, chemical treatment, surface polarity, and topography.^{9,10} Compared to PET, glass, and sisal fibers, aramid fibers are more effective in promoting transcrystallinity of PP.¹¹ Kevlar fiber (KF) does not exhibit any nucleation ability for isotactic polypropylene (iPP) in

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a quiescent melt. However, different nucleating abilities were observed by pulling the fiber embedded in the matrix, depending on the fiber-pulling rate, the duration of pulling, and the temperatures used for fiber pulling.¹²

Previously, we have reported the positive effects of incorporating a small amount of KF in WF/high-density polyethylene (HDPE) composites on the tensile, flexural, and impact properties of the resulting composites.¹³ To date, there have been few reports on the isothermal crystallization behavior of KF-reinforced WF/HDPE composites. The objective of this work is to compare the isothermal crystallization kinetics of neat HDPE and its composites [with WF, WF and KF, WF and grafted Kevlar fiber (GKF)] using differential scanning calorimeter (DSC).

EXPERIMENTAL

Materials

HDPE (5000S resin, density 0.954 g cm⁻³, and melt flow index 0.7 g/10 min) from Daqing Petrochemical Co., China, was used as the matrix. Kevlar-29 fibers (6 mm in length and 12 μm in diameter) were purchased from Du Pont, USA. WF that passed through a sieve of 40-mesh size (425 μm) but was retained on a 70-mesh (212 μm) sieve was supplied by Harbin Yongxu, China. KF co-grafted with the mixture of allyl chloride, and γ-chloropropyltrimethoxysilane was prepared in our laboratory as described previously.¹³

Sample preparation

To avoid any effects of heating history during processing on the polymer crystallization, all samples, including neat HDPE, WF/HDPE composite (WPC), KF-reinforced WPC (KFWPC), and GKF-reinforced WPC (GKFWPC), were prepared under the same processing conditions. WF was dried at 105°C for 24 h to remove any moisture. HDPE was respectively mixed with WF, WF and KF, or WF and GKF in a high-speed mixer for 8 min, and, subsequently, the mixture was extruded through a twin-screw/single-screw extruder system to form sample sheets. Neat HDPE sheets were also prepared as reference using the same mixing and extrusion process as the other samples. The processing parameters have been described previously.¹³ The weight ratios of HDPE/WF/KF/GKF were 40/0/0/0 for HDPE, 40/60/0/0 for WPC, 40/57/3/0 for KFWPC, and 40/57/0/3 for GKFWPC, respectively.

Differential scanning calorimeter measurement

The isothermal crystallization kinetics was investigated using a Perkin-Elmer Diamond DSC (Perkin-

Elmer Company, USA). Temperature and enthalpy calibrations were performed by using indium standards. Samples of ~ 5 mg were heated to 443 K at a heating rate of 10 K min⁻¹. This target temperature was held for 10 min to eliminate any influence of thermal history. The melts were then rapidly cooled at the rate of 100 K min⁻¹ down to the isothermal crystallization temperatures (T_c) of interest: 391, 393, 395, and 397 K, respectively. The temperature was maintained at the T_c until the completion of polymer crystallization. Afterward, the samples were reheated to 443 K at a rate of 10 K min⁻¹ to determine the crystalline melting temperatures (T_m) and crystallinity (X_{cr}) of polymer during the endothermic process.

The crystallinity of HDPE component was determined according to the following equation:

$$X_{cr} = \frac{\Delta H_f \times 100}{\Delta H_f^0 \times W} \quad (1)$$

where ΔH_f and ΔH_f^0 are the enthalpy of fusion of the measured sample and 100% crystalline HDPE sample, respectively, and W is the weight fraction of HDPE in the composite. The enthalpy ΔH_f^0 value for 100% crystalline polyethylene is taken as 290 J g⁻¹ as previously reported.¹⁴

RESULTS AND DISCUSSION

Melting and crystallization behavior

Addition of WF or/and KFs to neat HDPE caused a marginal reduction in the crystalline melting temperature (T_m) compared to the neat HDPE (Table I), which is in agreement with the results reported previously.^{15,16} The lower T_m of the HDPE matrix in the Kevlar composites may be attributed to the improved nucleation of HDPE on the KF surfaces, which shortened the time required for HDPE crystallization, therefore limiting the isothermal thickening of HDPE crystals and decreasing their melting temperature.⁶ For the composites without KF, WF does not act as a nucleating agent for the HDPE matrix, and large size of WF can lessen the adsorption of WF to the HDPE matrix due to steric hindrance,¹⁷ therefore limiting the perfection of crystallization of HDPE.

With increasing crystallization temperature, the melting temperature of all samples is increased. This behavior may be explained by the formation of more perfect crystals during crystallization at higher temperature. After the addition of WF, the enthalpy of fusion (ΔH_f) significantly decreased (Table I), because WF not only has no nucleating effects,¹⁵ but also severely inhibits the crystallization of the HDPE matrix, which can be deduced from the results that the crystallinity (X_{cr}) in the WF composites is only half that in the neat HDPE. After replacing a portion

TABLE I
Melting and Crystallization Kinetic Parameters of Neat HDPE and Composites Obtained from DSC Exotherms

Samples	T_c (K)	Thermal properties										
		n		K (min^{-1})	$T_{1/2}$ (s)	T_m (K)	ΔH_f (J g^{-1})	ΔE (kJ mol^{-1})	X_{cr} (%)	T_m^0 (K)	K_g (K^2)	σ_e (J m^{-2})
HDPE	391	2.41		3.041	32.4	404.4	146.00	80.1	50.3	407.1	2.279×10^4	1.39×10^{-2}
	393	2.99		1.336	48.0	404.6	141.32		48.7			
	395	2.51		0.299	84.0	405.0	134.12		46.2			
	397	2.94		0.012	238.2	405.6	121.33		41.8			
WPC	391	3.40	1.54	6.752	30.6	403.0	30.02	82.1	26.0	409.4	3.386×10^4	2.00×10^{-2}
	393	3.17	1.64	1.152	51.0	403.9	28.79		24.8			
	395	2.67	1.52	0.278	84.6	404.5	27.62		23.8			
	397	2.77	1.62	0.012	259.2	405.1	30.74		26.5			
KFWPC	391	2.46	1.22	8.819	21.6	403.6	61.26	64.7	52.8	405.0	1.312×10^4	7.54×10^{-3}
	393	2.56	1.24	4.016	30.0	403.8	50.48		43.5			
	395	2.41	1.29	0.976	52.2	403.9	45.30		39.0			
	397	1.77		0.227	112.8	404.4	46.99		40.5			
GKFWPC	391	2.91	1.42	7.907	25.8	403.5	40.94	69.0	35.3	406.3	1.704×10^4	1.01×10^{-2}
	393	3.15	1.37	2.585	39.6	404.0	38.03		32.8			
	395	2.51	1.45	0.389	75.6	404.3	36.34		31.3			
	397	2.38	1.62	0.094	139.2	404.7	32.48		28.0			

of the WF with the virgin KFs, the ΔH_f increased, which may be explained by the increased X_{cr} due to the KFs acting as nucleating agents. Replacing the same portion of the WF with the GKFs also causes an increase in ΔH_f , however, not as much as with the virgin KFs, which can be attributed to the improved interfacial interaction between the grafted KFs and HDPE matrix.

The effect of KF reinforcement on the crystallization behavior and thermodynamics of the HDPE matrix was evaluated by analyzing the isothermal

crystallization kinetics of neat HDPE and its composites in the temperature range of 391–397 K. The crystallization of the HDPE matrix for all samples test was considerably affected by crystallization temperature (T_c). As the T_c increased, the exothermic peak positions shifted along the time axis (Fig. 1). Both the induction time and the width of exotherms increased. This suggests that at higher crystallization temperature and longer crystallization time, the crystallization rate decreased. This is due to the fact that high-crystallization temperature can cause greater

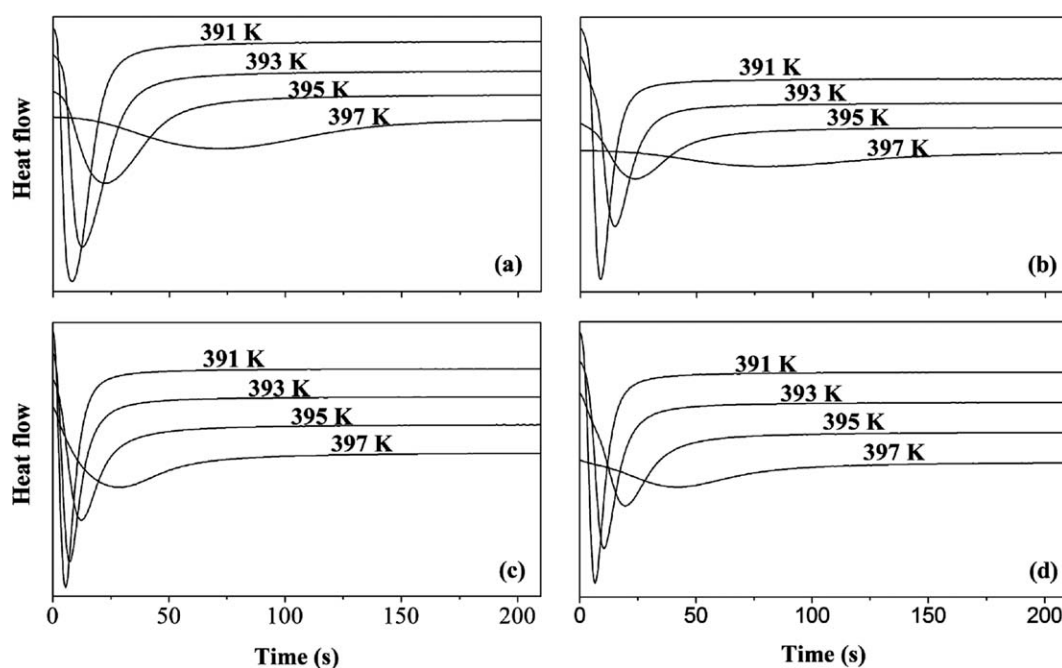


Figure 1 The DSC traces of samples isothermally crystallized at the specified temperature: (a) neat HDPE; (b) WPC; (c) KFWPC; (d) GKFWPC.

thermal motion of HDPE molecular chain, which leads to the difficulty in formation of crystal nucleus or the instability of the formed crystal nucleus. At the same crystallization temperature (e.g., 395 K), the crystallization exothermic peaks of KFWPC appeared earliest, followed by GFWPC, neat HDPE, and WPC, suggesting the same order of crystallization rate. This indicates that virgin KFs in the composites result in an improved heterogeneous nucleation. When the KF surface was co-grafted with allyl and 3-trimethoxysilylpropyl, grafted KF can still function as a heterogeneous nucleating agent in the composites; however, the nucleating effect was obviously reduced compared to the virgin KF. The use of silane-coupling agent can establish chemical bonding between KF fiber and HDPE, thereby limiting the thermal motion of HDPE segments. This may explain the lower crystallization rate of grafted KF. For WF, there are still some arguments on if it is a nucleating agent for HDPE. The studies from Mucha and Królkowski¹⁶ and Zou et al.¹⁷ have revealed that WF does not act as a nucleating agent for the HDPE matrix, but some others do not draw such a conclusion.^{9,10,18,19} In this study, WF does not act as a nucleating agent for HDPE, because the large size of WF may limit the melt flow of polymer and restrict its rearrangement due to steric hindrance. Consequently, further investigations are needed to clarify the role of the size, chemical composition, surface polarity, and surface topography of wood particles in the nucleation activity and crystallization kinetics of polymers.

The relative crystallinity (X_t) was calculated as the ratio of the exothermic peak area at time t and infinite time²⁰:

$$X_t = \int_0^t \left(\frac{dH}{dt} \right) dt / \int_0^{t_\infty} \left(\frac{dH}{dt} \right) dt \quad (2)$$

where dH is the enthalpy of crystallization released during an infinitesimal time interval dt . The plots of X_t as function of time t at various crystallization temperatures are shown in Figure 2. It can be seen that characteristic sigmoid isotherms shift to the right with increasing isothermal crystallization temperature, and the crystallization becomes slower.

Isothermal crystallization kinetics

The isothermal crystallization kinetics were further examined using the Avrami equation, where the relative crystallinity is given as follows^{21–23}:

$$X_t = 1 - \exp(-Kt^n) \quad (3)$$

where X_t is the relative crystallinity, t is the time, K is the kinetic rate constant related to nucleation and

growth parameters, and n is the Avrami exponent, which depends on both the geometry of the growing crystals and the nucleation process.

The above equation can be rearranged by taking its double logarithm:

$$\ln[-\ln(1 - X_t)] = n \ln t + \ln K \quad (4)$$

As a result, the values of n and K can be respectively calculated from the slope and the intercept of the straight line obtained by plotting $\ln[-\ln(1 - X_t)]$ against $\ln t$. It must be noted that the Avrami analysis is applied in the X_t range of 0.10–0.80. The crystallization half-time $t_{1/2}$ is defined as the time at which the extent of crystallization is 50%, and it depends on crystallization rate constant K and the Avrami exponent n :

$$t_{1/2} = (\ln 2/K)^{1/n} \quad (5)$$

The Avrami plots of $\ln[-\ln(1 - X_t)]$ versus $\ln t$ for HDPE and composites are shown in Figure 3. The experimental data of the composites (WPC, KFWPC, and GFWPC) did not appear to fit the Avrami equation, because the Avrami plot was not linear through the entire isothermal crystallization period. However, as the Avrami plot was divided into two parts at the degree of conversion of ~ 0.5 , each part of the curve individually exhibited linear behavior and can be almost fitted by the Avrami equation. In the initial part, the Avrami exponent n of WPC, KFWPC, and GFWPC was in the range of 2.4–3.4, which normally corresponds to a tridimensional (spherulitic) growth with heterogeneous nucleation.²⁴ Addition of virgin KF caused a reduction in the n value (1.8–2.6), suggesting that the nucleation pattern of HDPE was changed from tridimensional to bidimensional or the mixed patterns.²⁵ Compared to the initial part, the second part exhibited a lower n value from 1.2 to 1.6. The isothermal crystallization kinetics can be proposed as: in the early stage, HDPE crystal grows in a tridimensional pattern around the nuclei. It must be noted that the WF and KF fibers are more than 60 wt % of the composites. With increasing crystal size, the available space can be limited, and the HDPE spherulites may impinge with each other and with the reinforcing WF and KF. As a result, the dimension of crystal growth is thus reduced. Therefore, the form of HDPE crystal in the composites may exhibit either rod-like, disk-like, or/and spherulite crystals.²⁶

The value of K , which is related to nucleation rate and growth processes, decreased with the increased T_c for all composite formula (Table I). At the same T_c , incorporation of Kevlar fiber caused an apparent increase in the K value, suggesting that KF (both the virgin and grafted) promotes the crystallization of HDPE. In

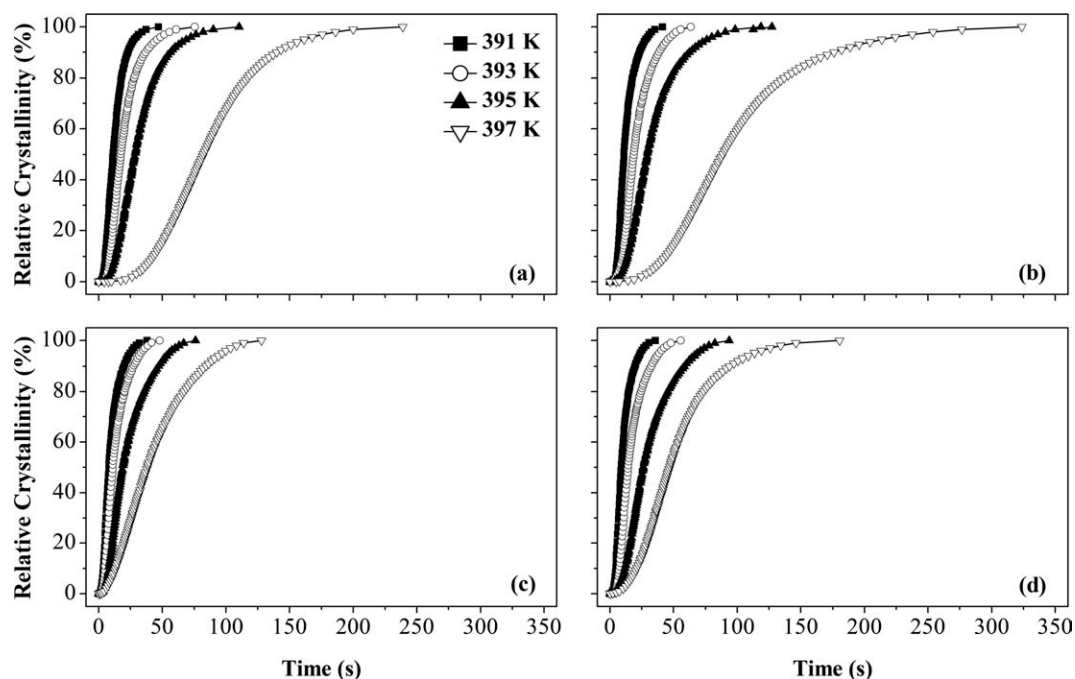


Figure 2 Plots of X_t versus t for the isothermal crystallization of samples at the specified temperature: (a) neat HDPE; (b) WPC; (c) KFWPC; (d) GKFWPC.

general, $1/t_{1/2}$, taken as a measure of the overall crystallization rate, increased with increasing T_c , indicating a longer crystallization time and a reduced crystallization rate. The crystallization rate ($1/t_{1/2}$) of HDPE in the KFWPC was greater than the neat HDPE and WPC at the same T_c , which may be attributed to better nucleation effects for HDPE on the KF surfaces.

To compare the crystallization ability, undercooling ($T_m^0 - T_c$) is taken into consideration, because the crystallization rate of a polymer depends mainly on its undercooling.²⁷ The undercooling and crystallization rate constants calculated from Avrami model are shown in Figure 4. All samples exhibited an increase in K value with increasing undercooling,

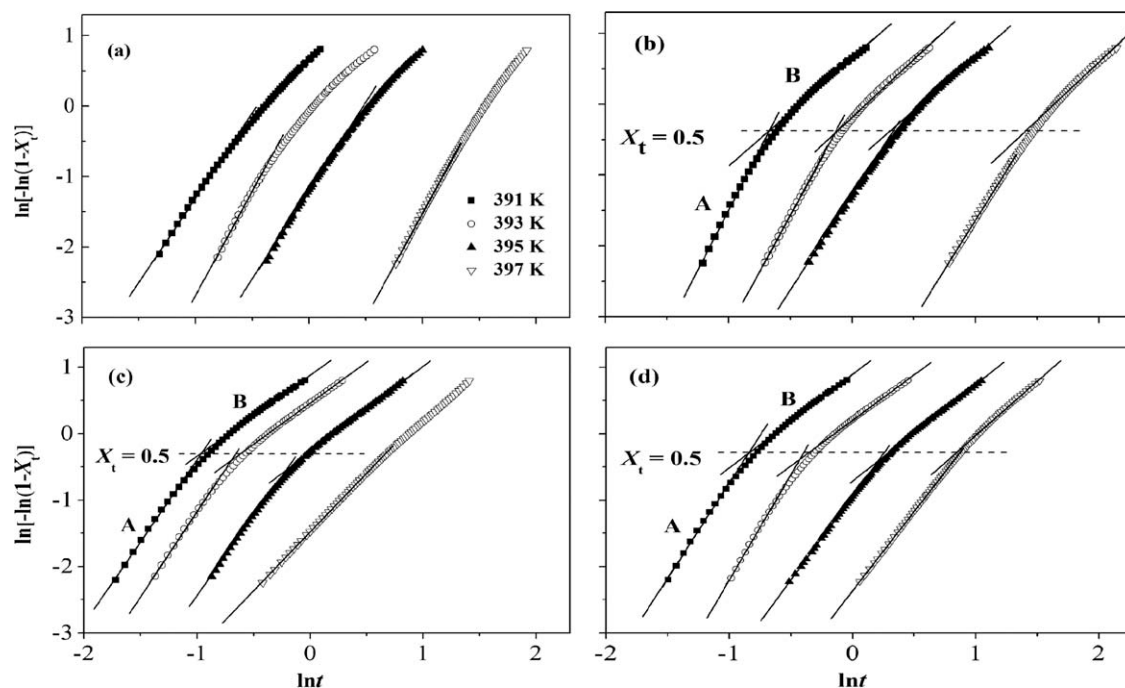


Figure 3 Plots of $\ln[-\ln(1 - X_t)]$ versus $\ln t$ for the isothermal crystallization of samples at the specified temperature: (a) neat HDPE; (b) WPC; (c) KFWPC; (d) GKFWPC.

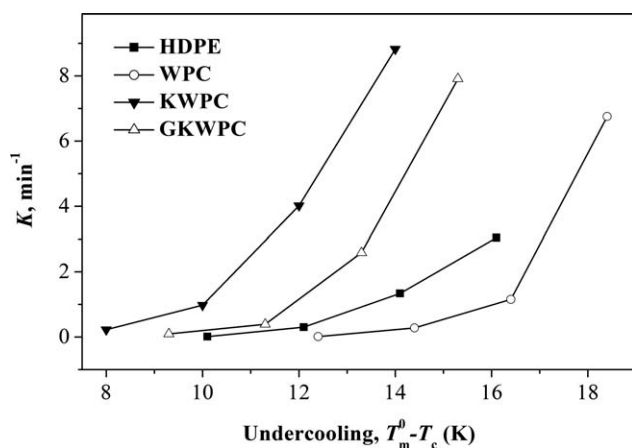


Figure 4 Relation of undercooling and rate constants.

suggesting a higher crystallization rate with greater undercooling. At a given rate constant, K , undercooling decreased in the following order: KWPC > GKWPC > neat HDPE > WPC.

Activation energy for crystallization

The activation energy for the overall isothermal crystallization process was determined based on an assumption that the Avrami crystallization rate constant can be described by an Arrhenius-type equation²⁸:

$$K^{1/n} = K_0 \exp(-\Delta E/RT_c) \quad (6)$$

where K_0 is the temperature-dependent pre-exponential factor, R is universal gas constant, and ΔE is the activation energy for the primary crystallization process. The activation energy consists of both the transport- and nucleation-activation energy. Plots of $(1/n)\ln K$ versus $1/T_c$ for the samples were linear only at the high-crystallization temperature (Fig. 5). Similar results were also observed in HDPE/CaCO₃ composites.²⁹ The slope (activation energy of the crystallization process) was obtained from the linear plots

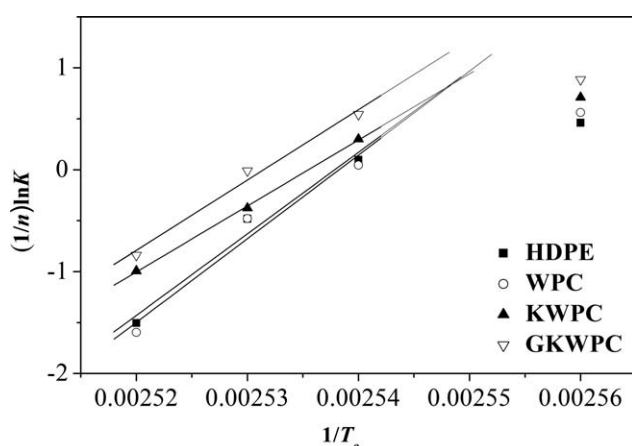


Figure 5 Plots of $(1/n)\ln K$ as a function of $1/T_c$.

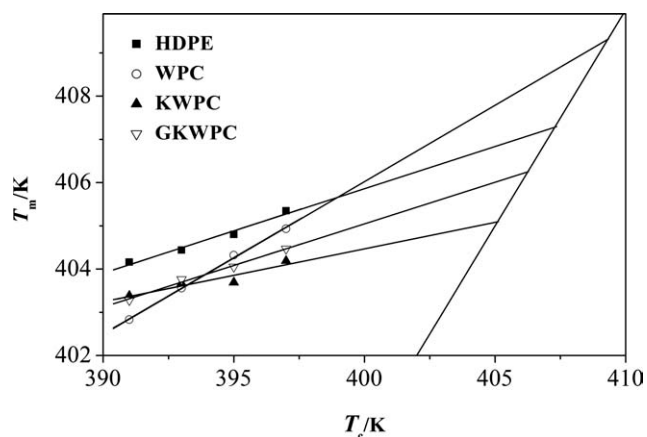


Figure 6 Hoffman-Weeks plots for neat HDPE and composites.

of $(1/n)\ln K$ versus $1/T_c$, and the values were 80.1, 82.1, 64.7, and 69.0 kJ mol⁻¹ for neat HDPE, WPC, KWPC, and GKWPC, respectively (Table I). The high ΔE value indicates low-crystallization ability. For both the KWPC and GKWPC composites, the ΔE was apparently lower than those of neat HDPE and WPC. The low ΔE for KF-based composites may be due to high-heterogeneous nucleating ability of KF. Similar results were also observed in aramid fiber/PP composites, where aramid fiber had the strongest heterogeneous nucleating ability compared to all other examined fibers (PET, glass, and sisal fibers).¹¹

Equilibrium melting temperature

The equilibrium melting temperature (T_m^0) of a polymer is an important thermodynamic parameter of crystallizable polymers, as it is the reference temperature for the driving force of crystallization,^{30,31} which is defined as the melting temperature of a perfect crystal formed by infinite molecular weight chains.²⁹ According to the Hoffman-Weeks theory,³² T_m^0 may be deduced by plotting the observed apparent melting temperature (T_m) against the crystallization temperature (T_c). The equilibrium melting point is obtained by an extrapolation of the resulting straight line to intersect the line of $T_m = T_c$. Figure 6 shows the plots of the T_m versus T_c for the neat HDPE and composites, and the equilibrium melting temperatures are given in Table I. The values of T_m^0 are 409.4, 407.1, 406.3, and 405 K for WPC, neat HDPE, GKWPC, and KWPC, respectively. This order is opposite to overall crystallization rate ($1/t_{1/2}$). This may be explained that crystal formed at a low rate may have better structure.²⁹

Hoffman-Lauritzen theory

The regime concept for polymer crystallization was first developed by Hoffman and Lauritzen.³³ They used the rate of secondary nucleation and the rate of lateral

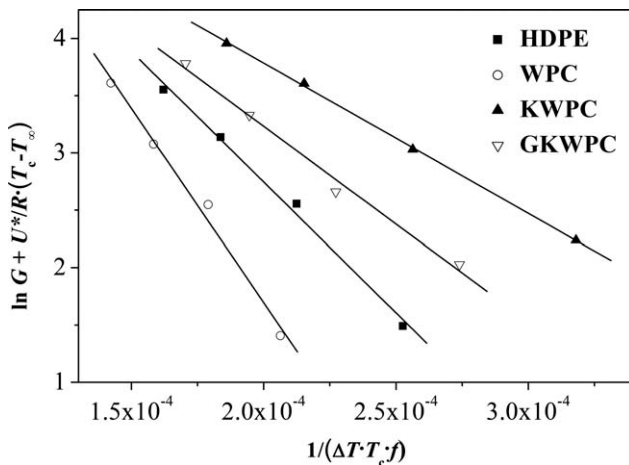


Figure 7 Hoffman–Lauritzen plots for isothermal crystallization of HDPE and composites.

surface spreading to describe the growth rate of polymer lamellar crystals. On the basis of Hoffman–Lauritzen theory, the overall crystallization rate, G , defined as $G = 1/(t_{1/2})$ is a function of the undercooling (ΔT):

$$G = G_0 \exp \left[-\frac{U^*}{R(T_c - T_\infty)} \right] \exp \left[-\frac{K_g}{T_c \Delta T f} \right] \quad (7)$$

where G_0 is a pre-exponential term, R is the universal gas constant, and U^* (6280 J mol^{-1}) is the diffusion activation energy for the transport of crystallizable segments at the liquid–solid interface.³⁰ T_c is the crystallization temperature, and $T_\infty = T_g - 30 \text{ K}$ is the hypothetical temperature, below which viscous flow ceases. T_g is the glass transition temperature of HDPE and taken as 163 K .³⁴ $\Delta T = T_m^0 - T_c$ and $f = 2T_c / T_m^0 - T_c$ is a correction factor. K_g is the nucleation parameter that can be related to the product of the lateral and folding surface-free energies ($\sigma\sigma_e$). Equation (7) can be further expressed as

$$\ln G + U^*/[R(T_c - T_\infty)] = \ln G_0 - K_g/(T_c \Delta T f) \quad (8)$$

The Hoffman–Lauritzen plots of $\ln G + U^*/[R(T_c - T_\infty)]$ against $1/(T_c \Delta T f)$ show good linearity for the neat HDPE, WPC, KFWPC, and GKFWPC (Fig. 7). The slope (K_g) is listed in Table I. The T_c of HDPE herein is in the range of regime III (the secondary nucleation rate is much higher than the lateral surface spreading rate),³⁵ and the K_g ($2.279 \times 10^4 \text{ K}^2$) is close to the value ($5.41 \times 10^4 \text{ K}^2$) previously obtained from regime III.³⁵ The obtained K_g value can be further used to determine the fold surface-free energy (σ_e)³⁶:

$$K_g = \frac{rb_0\sigma\sigma_e T_m^0}{k\Delta h_f} \quad (9)$$

where b_0 is the distance between two adjacent fold plains, σ and σ_e are the lateral and fold surface-free

energies, respectively, k is the Boltzman constant, and Δh_f is the heat of fusion per unit volume of crystal. The values of b_0 , σ , k , and Δh_f are 4.11 \AA ,³⁷ $1.07 \times 10^{-2} \text{ J m}^{-2}$,³⁸ $1.38 \times 10^{-23} \text{ J K}^{-1}$, and $2.93 \times 10^8 \text{ J m}^{-3}$,³⁵ respectively. The value of r is dependent on the regime of crystallization, and it is theoretically given as four for regime III. The fold surface-free energy (σ_e) was estimated by Eq. (8), and the values are listed in Table I. The value of σ_e follows the order of $\text{WPC} > \text{HDPE} > \text{GKFWPC} > \text{KFWPC}$. Compared to the neat HDPE, the incorporation of WF resulted in an increase in the value of σ_e , suggesting a reduction of the crystallization rate of HDPE. The lowest σ_e , that of virgin KF reinforced WF/HDPE composite, may be explained by the strong nucleating effects of KF. The value of σ_e for GKFWPC is between that of HDPE and KFWPC, which is consistent with the crystallization rate.

CONCLUSIONS

The isothermal crystallization kinetics of the neat HDPE, WPC, KFWPC, and GKFWPC was investigated by the DSC and analyzed by the Avrami model and Hoffman–Lauritzen theory. The values for the overall crystallization rate ($1/t_{1/2}$), the crystallization rate constant (K), the activation energy (ΔE), the equilibrium melting temperature (T_m^0), and the fold surface-free energy (σ_e) suggest that the crystallization rate of samples decreases with an increase of crystallization temperature, and incorporation of Kelvar fiber changed the growth pattern of HDPE crystal and efficiently promoted the crystallization of HDPE in the resulting WF/HDPE composite system. The decrease in the Avrami exponent values of the three composites further confirms the change in the crystallization pattern. Accordingly, this study demonstrates that KF and GKF can act as nucleating agents to improve the crystallization rate of HDPE, and WF does not act as a nucleating agent for HDPE.

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