

Thermal Conductivity of Glass Fiber Reinforced Polypropylene under High Pressure

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SYNOPSIS

The thermal conductivities of molten polypropylene and its glass fiber composites were measured by the compensating hot wire method. The testing apparatus employed was designed and tested in our laboratory. The measurements were carried out with temperatures ranging from 170 to 230°C and pressures from 1 to 2000 kg/cm². The results show that the thermal conductivity increases with increasing pressure and glass fiber content, but is almost independent of temperature. The thermal conductivity data were fitted satisfactorily with a proposed empirical equation for polypropylene and Lewies-Nielsen equation for the composites, respectively.

INTRODUCTION

Studies on the thermal conductivity of polymers and their composites in the molten state are not only of the fundamental interest but also helpful in the computer simulation of polymer processing. In the investigation of thermal conductivity of polymers, most experiments were directed toward the effects of temperature,¹⁻⁴ molecular weight of polymer,^{5,6} and filler,⁷⁻⁹ etc. Only a few studies had been made on pressure effect.¹⁰⁻¹²

Among the methods of the thermal conductivity measurement reported in literature, the transient hot wire method¹¹⁻¹³ seems to be the most attractive one for present study. The major advantage of this method is that the measurement can be done quickly (1-10 s) so as to prevent the sample from thermal degradation. The compensating hot wire method,^{14,15} an improved method of transient hot wire method, employs two wires with different length in different branches of a Wheatston bridge so that the end effect of the wire can be eliminated by the subtracting effect of the bridge.

In the present study, an apparatus based on the

compensating hot wire method has been developed and employed to measure the thermal conductivity of polypropylene and its composites in the molten state under high pressures and temperatures. Investigations are focused on the effect of temperature, pressure, and glass fiber content on the thermal conductivity.

EXPERIMENTAL

Preparation of Samples

The glass fiber (Ta Lai Glass Co., E type, with average length = 6 mm, and diameter = 18-20 μm) and isotactic polypropylene (Yung Chia Chem. Ind. Co., Youngsox 1080, $M_w = 108,000$ and MWD = 3.9) were used for the preparation of the polypropylene composite. The polypropylene and untreated glass fiber were dried in an oven which was maintained at 80°C for a 1-day period. The polypropylene was then compounded twice with various amounts of glass fiber through a twin screw extruder and was then cut into pellets.

Measurement of Thermal Conductivity

The thermal conductivities of the prepared samples were determined by the testing apparatus which was

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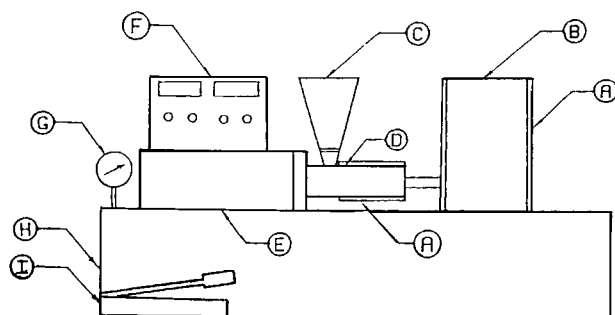


Figure 1 The schematic diagram of the thermal conductivity testing apparatus. (A) Electrical heater; (B) test cell; (C) sample feeder; (D) extruder; (E) electrical oil pump; (F) temperature controller; (G) pressure gauge; (H) base; (I) hand oil pump.

designed in our laboratory based on the compensating hot wire method. The apparatus consists mainly of a pressure system, an extruder, an electrical heating system, and a test cell as shown in Figure 1. The changes of the Wheaton bridge voltage were measured by a multimeter (HP3478A) and recorded by a personal computer. The sampling rate is about 35 data points/s. In the test cell (shown in Fig. 2), two platinum wires of 1 mm in diameter, 10 and 5 cm in length, respectively, and with the temperature resistance coefficient about $3.8 \times 10^{-3} \Omega/^\circ\text{C}$, were equipped parallelly in the centers of a combined double-cylinder sample cell, with each cylinder 2 cm in diameter. The sample was melted by a temperature-controlled electrical heater and was slowly extruded through feeding channel into the test cell by a piston.

The measured voltage changes of the Wheaton bridge were used to calculate the temperature changes of the wires during the heating; then the thermal conductivity of the tested sample can be calculated^{14,15} by

$$K = \frac{Q}{4\pi} \frac{d \Delta T}{d \ln t} \quad (1)$$

where K is the thermal conductivity of the tested sample, Q is the heat generated in the wire per unit length, t is time, and ΔT is the temperature change at time t with respect to the initial temperature of the sample.

The calculated thermal conductivity data of polypropylene under 1 atm were compared to the results reported in literature,^{4,6,16,17} as shown in Figure 3. It can be seen that the reported thermal conductivity data differ from each other due to the wide range of MW and MWD of the samples. It is ob-

served that the thermal conductivity decreased when MW is decreased (compare curves 3 and 6), or when MWD is decreased (compare curves 1 and 5). This is opposite to what Ramsey⁶ found for PE and PP. Since thermal conductivity measurements between different laboratories or with different techniques are seldom in better than $\pm 10\%$ agreement, further investigations should be taken in order to make a definite statement regarding the effect of MW and MWD on the thermal conductivity of polymer melts.

It is worth noting that the data reported by Wang et al.¹⁶ were measured by a well-designed probe which enabled very convenient and quick measurements.

RESULTS AND DISCUSSIONS

In the present study, measurements were carried out with temperatures ranging between 170 and 230°C, and pressures ranging between 1 atm and 2000 kg/cm², for the samples of polypropylene and its composites with 10.88 wt % and 18.93 wt % glass fiber content.

The effect of pressure and temperature on the thermal conductivity of the molten polypropylene are shown in Figure 4. The experimental results were fitted into an empirical equation, eq. (2), which is similar to that used by Sanberg¹² except an additional term, $\ln PT$, was included to account for the cross effect of temperature and pressure:

$$K = a + bT + cP + dP^2 + e \ln(PT) \quad (2)$$

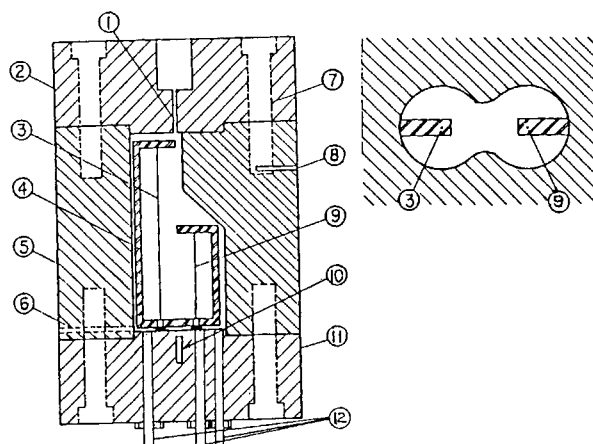


Figure 2 The side and top view of test cell. (1) Vent; (2) top cover; (3) long pt wire; (4) wire supporter; (5) test cell; (6) feeding channel; (7) high tensile bolt; (8) top temperature sensor; (9) short pt wire; (10) bottom temperature sensor; (11) bottom cover; (12) circuit connectors.

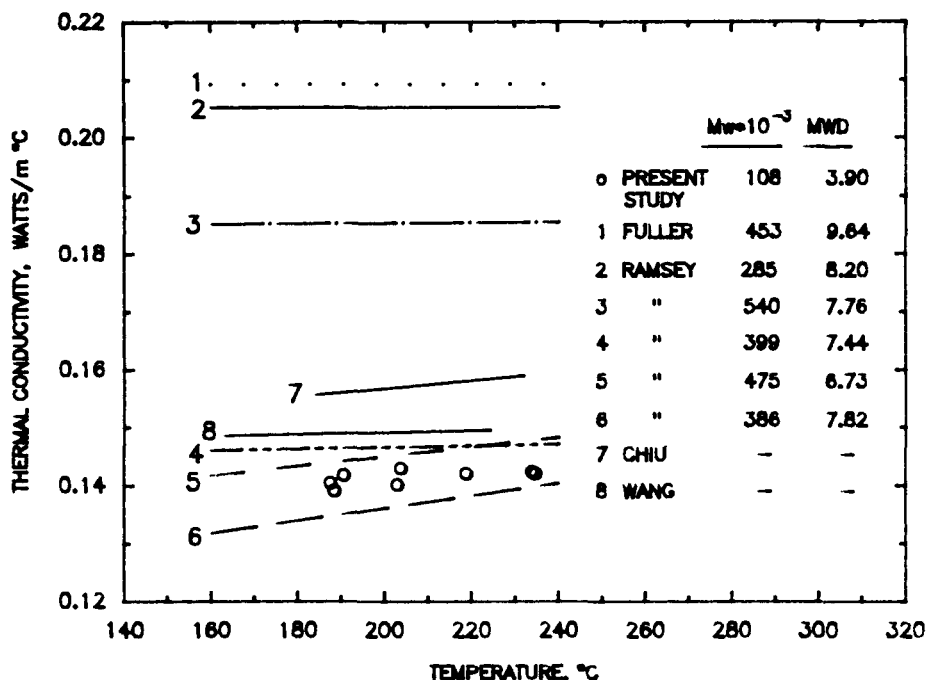


Figure 3 Comparison of experimental results of thermal conductivity of polypropylene under 1 ATM.

The fitted coefficients in eq. (2) are listed in Table I, and the standard deviation between experimental and the fitted data is $3.87 \cdot 10^{-3}$ Watts/m°C.

For the polypropylene composites with 10.88 and

18.93 wt % glass fiber content, the experimental results are shown in Figures 5 and 6, respectively. The solid lines are the results of fitting the experimental data to the Lewies-Nielsen equation¹⁸:

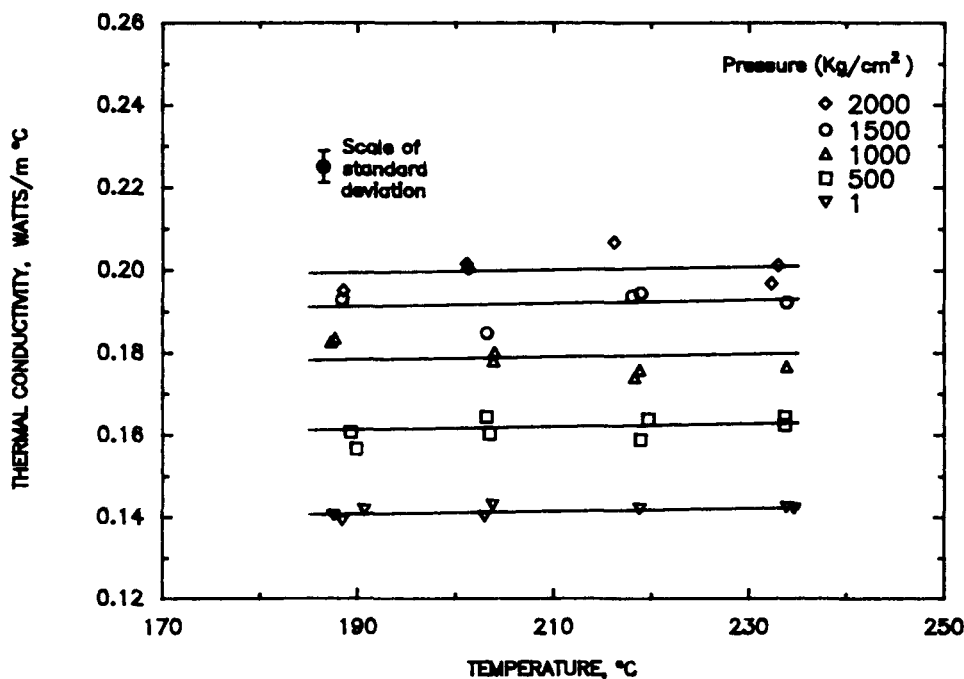


Figure 4 Thermal conductivities of polypropylene.

Table I Fitted Coefficients in Eq. (2)^a

Coefficient	Value
<i>a</i>	1.345×10^{-1}
<i>b</i>	3.539×10^{-5}
<i>c</i>	4.690×10^{-5}
<i>d</i>	-8.584×10^{-9}
<i>e</i>	1.021×10^{-4}

^a Units: *K* in W/m °C, *P* in kg/cm², and *T* in °C.

$$K_e = K_c \left[\frac{1 + AB\phi}{1 - B\phi\Psi} \right] \quad (3)$$

$$B = \frac{K_d/K_c - 1}{K_c/K_c + A} \quad (4)$$

$$\Psi = 1 + \left(\frac{1 - \phi_m}{\phi_m^2} \right) \phi \quad (5)$$

where *K_c* = thermal conductivity of continuous phase. (i.e., polypropylene), *K_d* = thermal conductivity of discrete phase (i.e., glass fiber), *K_e* = effective thermal conductivity of composite, *φ* = volume fraction of discrete phase, *φ_m* = maximum packing fraction, and *A* = shape factor. The maximum packing fraction *φ_m* is chosen to be 0.52 for fibers randomly dispersed in the polypropylene.¹⁷ The thermal conductivity of glass fiber was calcu-

lated by eq. (6) which was obtained through extrapolation from the experiment results of Ott¹⁹:

$$K_g = 1.01 + 1.814 \times 10^{-3} T - 1.983 \times 10^{-5} T^2 \quad (6)$$

where *K_g* is the thermal conductivity of glass fiber (in W/m °C), and *T* is the temperature (°C).

The specific volumes of glass fiber and polypropylene were calculated from eqs. (7)²⁰ and (8),²¹ respectively,

$$V_g = 0.3916 + 1.8824 \times 10^{-6} (T - 25) \quad (7)$$

$$V_p = V_0(T) \{ 1 - C \ln[1 + P/B(T)] \} \quad (8)$$

where *V_g* = specific volume of glass fiber (cm³/g), *V_p* = specific volume of PP (cm³/g), *V*(*T*) = 1.1606 × exp(6.7 × 10⁻⁴ *T*), *B*(*T*) = 4.177 × 10⁻³ exp × (-4.177 × 10⁻³ *T*), and *T* = temperature (°C).

The measured effective thermal conductivity data, corresponding to different volume fractions of glass fiber content, were fitted to eqs. (3)–(5) with the shape factor *A* as an adjusting parameter. The value of *A* was determined by the simplex method²² to be 2.256 for the best fit of the experimental data to the Lewies-Nielsen equation. The standard deviations between the experimental data and the fitted curves are 6.55 × 10⁻³ and 4.38 × 10⁻³ W/m °C

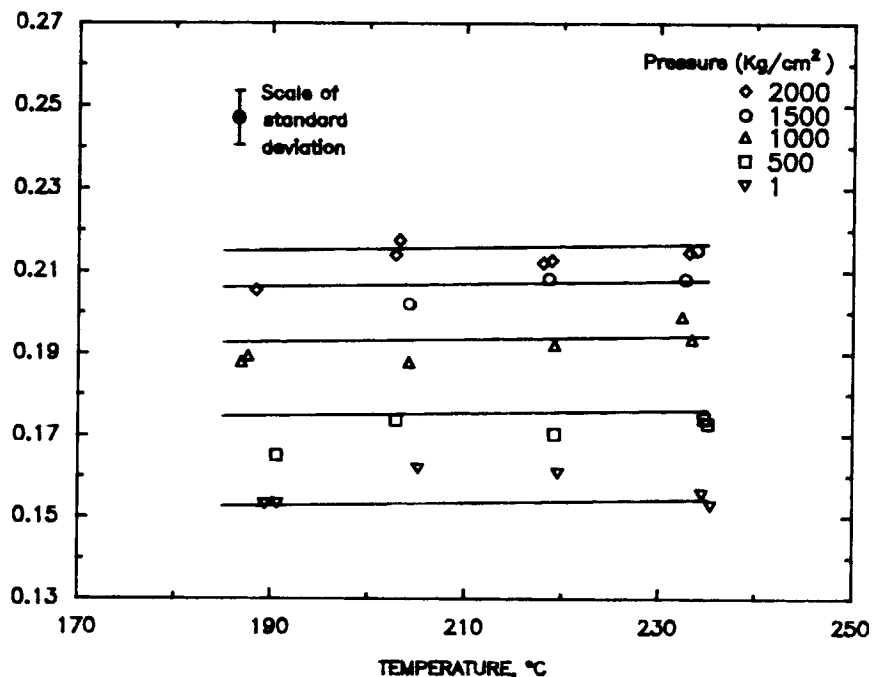


Figure 5 Thermal conductivities of the composite with 10.88 wt % glass fiber content.

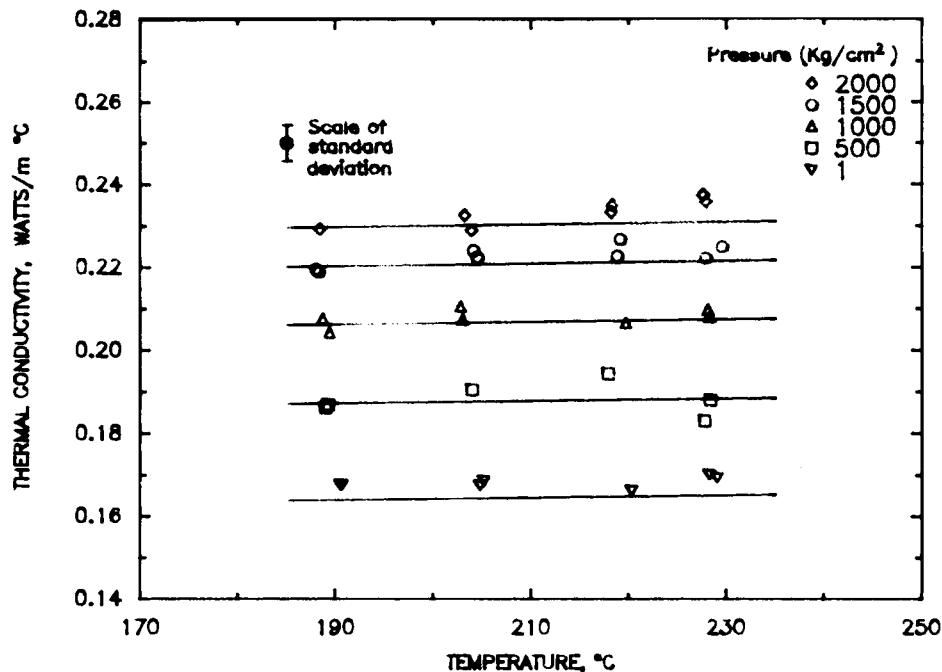


Figure 6 Thermal conductivities of the composite with 18.93 wt % glass fiber content.

for the composite with glass fiber content 10.88 and 18.93 wt %, respectively.

It can be seen from Figures 4–6 that the thermal conductivities of molten polypropylene and its composites increase slightly with increasing temperature. The temperature effect is so small that it may be considered that the thermal conductivity is almost independent of temperature. This is in agreement with literature reports^{4,6,16,17} for the measurements at 1 atm, as shown in Figure 3.

The effect of pressure and glass fiber content on the thermal conductivity are more profound than that of the temperature. In order to elucidate this phenomenon, the thermal conductivity for polypropylene and its composites at constant temperature of 230°C were plotted against pressure, as shown in Figure 7. It can be seen that the addition of glass fiber has an effect of increasing the thermal conductivity; the thermal conductivity increases almost linearly with pressure when pressure is low, and the effect levels down at high pressure. The ratio of the thermal conductivity at 2000 kg/cm² to that at 1 kg/cm² is roughly 1.4 for molten polypropylene and its composites as well. It was stated by Wynter³ that Lohe²³ had measured the thermal conductivity of molten polyethylene which is a semicrystalline polymer as polypropylene is. The results of Lohe showed that the thermal conductivity of molten polyethylene at 300 kg/cm² increased about 5% with respect to that

at 1 kg/cm². The results of the present study is in accord with that of Lohe, as described by Wynter. The slight temperature dependence of thermal conductivity of polymer melts may be explained by competing effects (both of small magnitude) of the increased segmental mobility at increased temperature to transfer heat more quickly and of the decrease in density at increased temperature such that the chain-to-chain heat transfer is less efficient. With increased pressure, the polymer chains are packed more tightly together to give more efficient thermal transfer.²⁴ Therefore, the thermal conductivity of PP increases profoundly with increasing pressure.

CONCLUSIONS

The thermal conductivities of molten polypropylene and its glass fiber reinforced composites were measured by the compensating hot wire method. The results show that, in the melt state, the thermal conductivity is almost independent of temperature and is strongly affected by the pressure and the glass fiber content. An empirical equation was proposed for the thermal conductivity of polypropylene as a function of temperature and pressure. The Lewies-Nielsen equation was applied to thermal conductivity of composites of polypropylene. Both equations fit the experimental data satisfactorily.

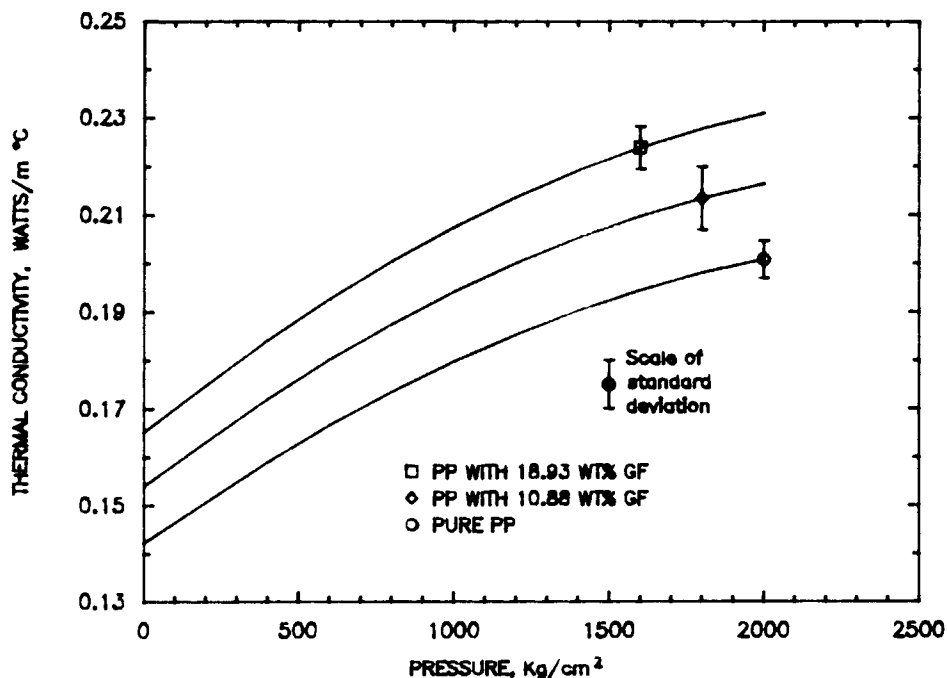


Figure 7 The effect of pressure on the thermal conductivities of polypropylene and its composites at constant temperature of 230°C.

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