

THERMAL CONDUCTIVITY OF HIGH-PRESSURE AND LOW-PRESSURE POLYETHYLENE IN THE TEMPERATURE RANGE FROM 3 TO 300 K*

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Thermal conductivity is measured in two polyethylene samples (*A* – high-pressure product, of lower crystallinity and density 0.931 g/cm³; *B* – low-pressure product, of higher crystallinity and density 0.959 g/cm³) at room temperatures as well as in the liquid nitrogen and liquid helium ranges.

The results obtained in the range from room temperature to that of liquid helium suggest that two temperature intervals of thermal conductivity are present. A comparison is made with the available data of others relative to measurements at room temperature and in the liquid helium range.

1. Introduction

Polyethylene belongs to the category of plastics which, according to the technology employed in processing the raw material, can vary in crystallinity and, consequently, in density. Plastics, owing to their excellent electrical and thermal insulating properties, are increasingly put to use as cryotechnical material for the construction of cryogenic equipment and low-temperature research stands. With regard to this situation, we proceeded to an investigation of high-pressure polyethylene of density 0.931 g/cm³ (sample *A*) and low-pressure polyethylene of density 0.959 g/cm³ (sample *B*)¹.

2. Experimental

The samples had the shape of perpendicular parallelepipeds, about 7 cm long, 1 cm wide, and 0.4 cm thick. The thermal conductivity of polyethylene was measured in the ranges of room temperature and those of liquid nitrogen as well as liquid helium.

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¹ The polyethylene samples were provided by the Warsaw Plastics Factory.

Temperature was determined, in the room temperature range, with an accuracy of $\pm 0.5^\circ\text{C}$ by means of Cu-constantan thermocouples scaled with a mercury thermometer. At nitrogen temperatures, this was done to within $\pm 0.05^\circ\text{C}$ with thermocouples which had been re-scaled resorting to the tabulated temperature dependence of the pressure of saturated nitrogen vapour. The thermocouples consisted of copper wire of diameter $40\ \mu$ and constantan wire of diameter $30\ \mu$. The thermocouple weldings were imposed at a depth of several mm in the sample, the distance between the weldings of two thermocouples used for determining the temperature gradient amounting to 3.62 cm for sample *A* and 4 cm for sample *B*. Resistance of the furnace, which consisted of $30\ \mu$ constantan wire, was $6\ \text{k}\Omega$ for sample *A* and $9\ \text{k}\Omega$ for sample *B*.

Temperature in the liquid helium range was determined by means of carbon resistors². The distance between the carbon thermometers on the samples was the same as in the case of the thermocouple weldings. The thermal conductivity coefficient was determined by the stationary method and calculated from the definition of Fourier's law in the form of a difference equation. The measuring technique is that described in short in Ref. [1] and in greater detail in Refs [2-5].

Measurements of the thermal conductivity for the two polyethylene samples in the range of room temperatures are shown in Table I.

TABLE I

Sample <i>A</i>		Sample <i>B</i>	
Temp. [K]	<i>K</i> [W/cm K]	Temp. [K]	<i>K</i> [W/cm K]
281.2-284.0	6.27×10^{-2}	288.3-292.5	8.05×10^{-2}
288.2-291.2	6.66×10^{-2}	291.3-296.1	8.15×10^{-2}
293.8-296.6	6.75×10^{-2}	295.5-299.7	8.35×10^{-2}
298.2-301.0	6.80×10^{-2}	299.3-303.3	8.50×10^{-2}

From Table I, the thermal conductivity of the two samples is seen to increase with rising temperature in the range from 280 to 300 K, exhibiting in the case of sample *B* (of higher crystallinity) values by more than 30 per cent larger than in that of sample *A*.

The results obtained when measuring the thermal conductivity of polyethylene in the range of liquid nitrogen temperatures are assembled in Table II.

TABLE II

Sample <i>A</i>		Sample <i>B</i>	
Temp. [K]	<i>K</i> [W/cm K]	Temp. [K]	<i>K</i> [W/cm K]
75.30-77.50	5.36×10^{-3}	73.90-79.20	6.48×10^{-3}
73.05-75.45	5.24×10^{-3}	72.55-77.85	6.39×10^{-3}
71.20-73.40	5.09×10^{-3}	70.55-75.70	6.25×10^{-3}
67.55-69.75	4.92×10^{-3}	69.20-74.30	6.12×10^{-3}

² The resistors were made at the Laboratory of the Institute of Physical Problems of the Academy of Sciences at Moscow.

From Table II, the thermal conductivity of the samples is immediately seen to grow with rising temperature in the liquid nitrogen range; here, too, it is larger for sample *B* than for sample *A*.

The results for the liquid helium range are given in Table III.

TABLE III

Sample <i>A</i>		Sample <i>B</i>	
Temp. [K]	K [W/cmK]	Temp. [K]	K [W/cm K]
3.895–3.970	5.10×10^{-4}	3.920–4.020	5.40×10^{-4}
3.795–3.885	4.50×10^{-4}	3.880–3.990	4.90×10^{-4}
3.700–3.820	3.90×10^{-4}	3.820–3.950	4.15×10^{-4}
3.595–3.760	2.96×10^{-4}	3.770–3.910	3.85×10^{-4}
3.470–3.680	2.32×10^{-4}	3.720–3.870	3.60×10^{-4}
3.345–3.610	1.82×10^{-4}	3.520–3.710	2.84×10^{-4}
3.235–3.520	1.71×10^{-4}	3.450–3.670	2.45×10^{-4}
2.635–2.955	1.52×10^{-4}	3.130–3.370	2.25×10^{-4}
2.585–2.920	1.45×10^{-4}	2.710–2.970	2.07×10^{-4}
2.470–2.820	1.39×10^{-4}	2.490–2.770	1.93×10^{-4}

These results are plotted in Fig. 1 for comparison with those of Reese and Tucker [6]. Agreement is seen to be satisfactory in the range from 2.5, to 3.5 K for our sample *A* with the results published [6] for polyethylene of specific density 0.964 g/cm^3 . Above 3.5 K the $K(T)$ curve runs more steeply both in the case of our sample *A* and in those of the samples of Ref. [6].

The sample *B* of this investigation exhibited a markedly larger thermal conductivity between 2.5 and 3.5 K than sample *A*. In the temperature range 3.5–4 K, the two samples revealed an almost identical $K(T)$ curve.

The lines plotted in Fig. 1 represent the theoretical shape of the thermal conductivity based on Callaway's theory [7], with the numerical values of the constants occurring in the theoretical formula brought into accordance [6] with the experimental data.

The polyethylene samples studied by us, exhibit an increase in the slope angle of $K(T)$ with rising temperature above 3.5 K. This is in qualitative agreement with the results of Salinger [8].

Fig. 2 shows the present results as compared with those of thermal conductivity measurements [9] in two polyethylene samples prepared from rods 1 inch in diameter and 10 inches in length; agreement is seen to be satisfactory.

Fig. 3 shows our results in comparison with those obtained [9] when measuring the thermal conductivity of two other polyethylene samples in the form of rods 0.5 inch in diameter. Our results are disposed slightly above the experimental curves of Kolouch and Brown [9].

Since the four samples of Ref. [9] came from two distinct sources, the technologies used in preparing them were most probably different.

Fig. 4 contains the results obtained by us when measuring the thermal conductivity of the already mentioned two polyethylene samples at room temperatures as well as in the

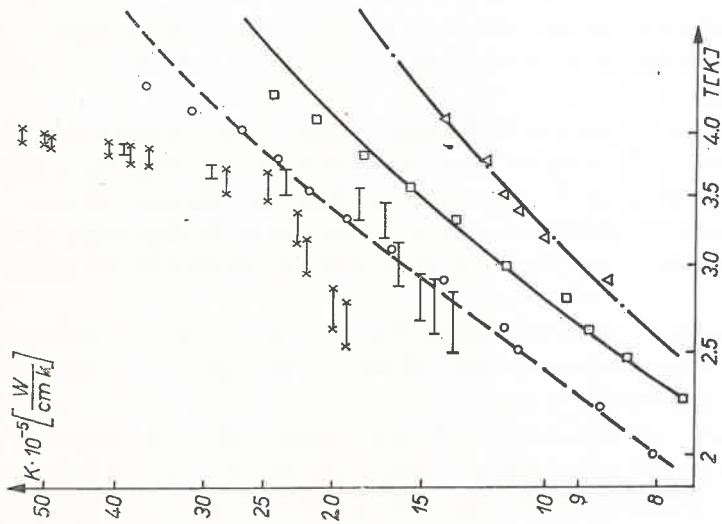


Fig. 1

Fig. 1. Temperature dependence of the thermal conductivity of the two polyethylene samples studied by us in the liquid helium region. Our results: --- sample A of density $\rho = 0.931 \text{ g/cm}^3$, x---x sample B of density $\rho = 0.959 \text{ g/cm}^3$. Results of Reese and Tucker: \circ - $\rho = 0.964 \text{ g/cm}^3$, \square - $\rho = 0.938 \text{ g/cm}^3$, Δ - $\rho = 0.917 \text{ g/cm}^3$
 Fig. 2. Comparison of the curves of thermal conductivity vs temperature $K(T)$ in the liquid helium region for our samples A and B and the results of Kolouch and Brown (+ - $\rho = 0.971 \text{ g/cm}^3$, Δ - $\rho = 0.933 \text{ g/cm}^3$) obtained for pressure-processed samples

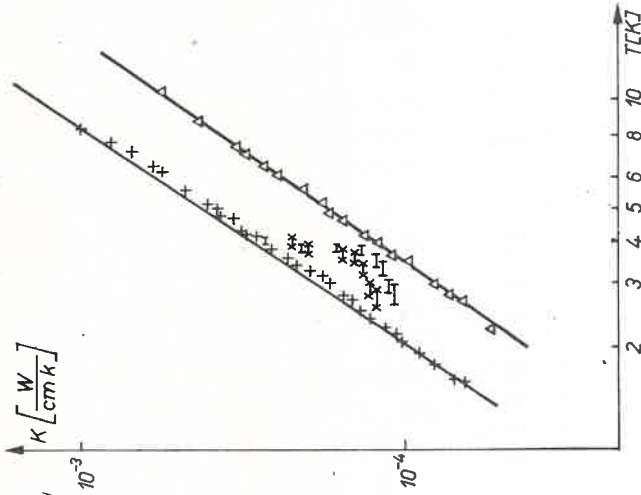


Fig. 2

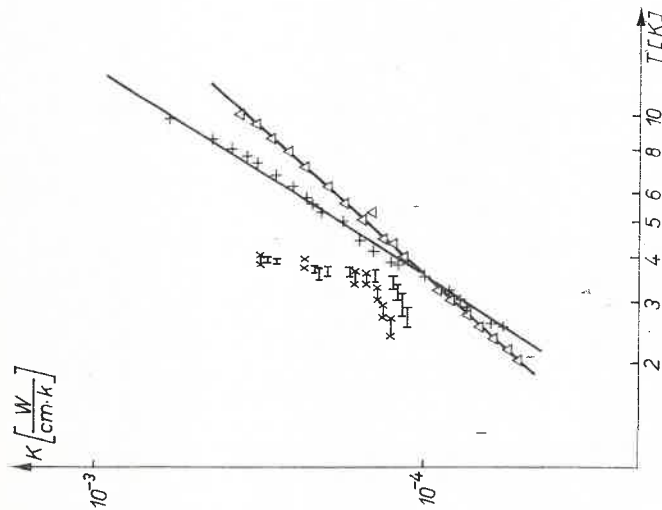


Fig. 3.

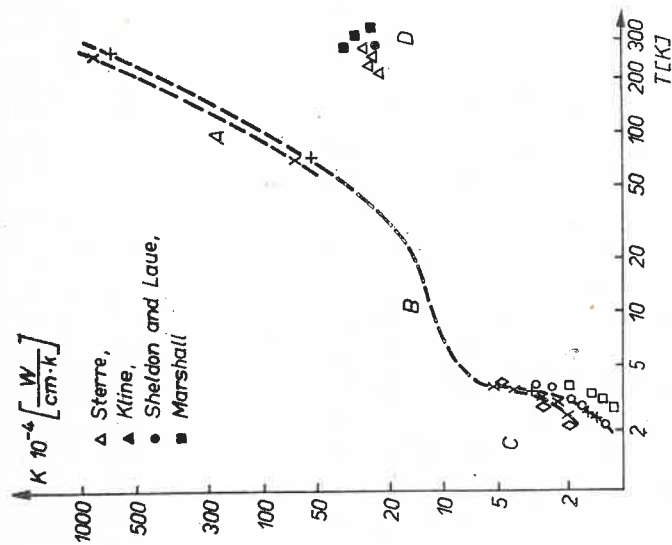


Fig. 4.

Fig. 3. Comparison of the curves of thermal conductivity vs temperature $K(T)$ in the liquid helium range for our polyethylene samples A and B and the results of Kolouch and Brown ($+ - \rho = 0.956 \text{ g/cm}^3$, $\Delta - \rho = 0.914 \text{ g/cm}^3$) obtained for industrially compressed products

Fig. 4. Thermal conductivity vs temperature measured by us in polyethylene samples A and B in the temperature range from room temperature to liquid helium temperature. C - liquid helium region: $+ -$ sample A ; $O - \rho = 0.964 \text{ g/cm}^3$, $\square - \rho = 0.938 \text{ g/cm}^3$, data of Reese and Tucker; $\diamond - \rho = 0.971 \text{ g/cm}^3$, data of Kolouch and Brown. B - presumable temperature dependence of thermal conductivity in the range from helium to nitrogen temperatures, A - temperature dependence of thermal conductivity in the range from nitrogen temperatures to room temperature. D - literature data for the region of room temperature

regions of liquid nitrogen and liquid helium temperatures. The graph contains moreover the results measured by the authors of Refs [10–13] at room temperature and of Refs [6] and [9] at liquid helium temperature. The literature fails to provide data for the thermal conductivity of polyethylene in the region of liquid nitrogen.

3. Conclusions

It appears from the results of Fig. 4 that the curve of K vs T exhibits an inflection in the temperature region separating the regions of liquid helium and liquid nitrogen, *i. e.* that the temperature dependence of conductivity undergoes a change.

From the viewpoint of physics, this means that at the temperature of liquid helium we are dealing with other mechanisms of dissipation of the thermal flow than those acting at and above the temperature of liquid nitrogen. This can be due to the circumstance that above the liquid nitrogen region (region *A* in Fig. 4) the mean free path of phonons is lesser than the mean linear dimension of the crystallites in the polyethylene samples under investigation, whereas in the region of liquid helium (*C* in Fig. 4) the mean free path of the phonons exceeds the effective mean size of the crystallites. On the other hand, the region of transition *B* is characterized by a mean free path of the phonons which is comparable with the effective size of the crystallites.

Our measurements of thermal conductivity of polyethylene in the liquid helium region agree qualitatively with the data available in the literature [6], [9] (Figs 1 and 2).

An inflection is found to appear in the temperature dependence of thermal conductivity $K(T)$ at a temperature of about 3.5 K, the steepness of $K(T)$ increasing with rising temperature (Figs 1, 2 and 3).

A comparison of the density of our samples and that of the samples discussed by others [6], [9] shows that a univalent relationship between the density and degree of crystallinity of the samples occurs only with regard to polyethylene produced according to the same technology. Samples of similar density but from different sources (and thus probably obtained by different technological procedures) are found to differ significantly as to their degree of crystallinity.

It still remains to be explained why the room temperature thermal conductivity of our samples exceeded by more than one order of magnitude the values found by others [10–13] in the same conditions.

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