RESULTS OF PHYSICAL TESTS ON POLYMER MATERIALS AT CRYOGENIC TEMPERATURES

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Summary

The mechanical and thermal properties of polymer materials have been studied in a temperature range from 4.2°K to 300°K. Some of these polymers have been irradiated up to 2 x 10^8 rad in liquid nitrogen and liquid neon and the same properties were subsequently tested. A brief description of the various testing techniques is given.

1. Introduction

The progress in low temperature physics, as well as the work on superconducting and cryogenic magnets and cryogenic techniques in general, e.g. refrigerators, pipelines, etc., necessitate a thorough investigation of the properties of various materials at low temperatures.

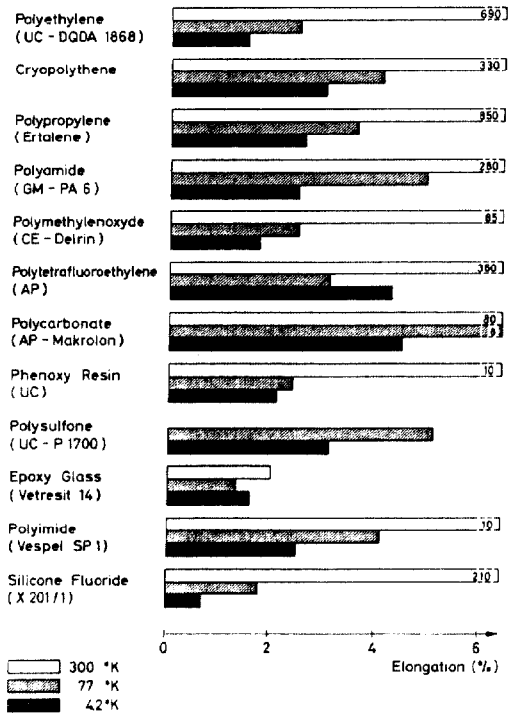
Tests have been made on some polymers which may be used as electrical insulators and constructional materials in magnets for high energy physics applications, and the results of measurements of some of their mechanical and thermal properties are presented. All materials studied are commercially available but were subjected to stringent quality control.

2. Experimental Techniques and Test Results

2.1 Mechanical Properties

a) Elongation

These tests are performed using an Instron tensile tester onto which a liquid helium cryostat has been fitted. The strain in the sample has been recorded from the output of a displacement transducer to better than 10^{-2} mm. The samples are of the dumb-bell type[1] with dimensions of 113 x 22 x 2 mm^3. The elongation at the break point for a number of promising polymers is shown in Figure 1. It is often only 1 % or less of the value at ambient temperature. Nevertheless, some polymers still exhibit an elongation of 2 % and more at 4.2°K. They primarily include heat resistant polymers, such as polyimides, fluorinated polyolefines, etc.



Elongation at Break of Polymer Materials

FIGURE 1

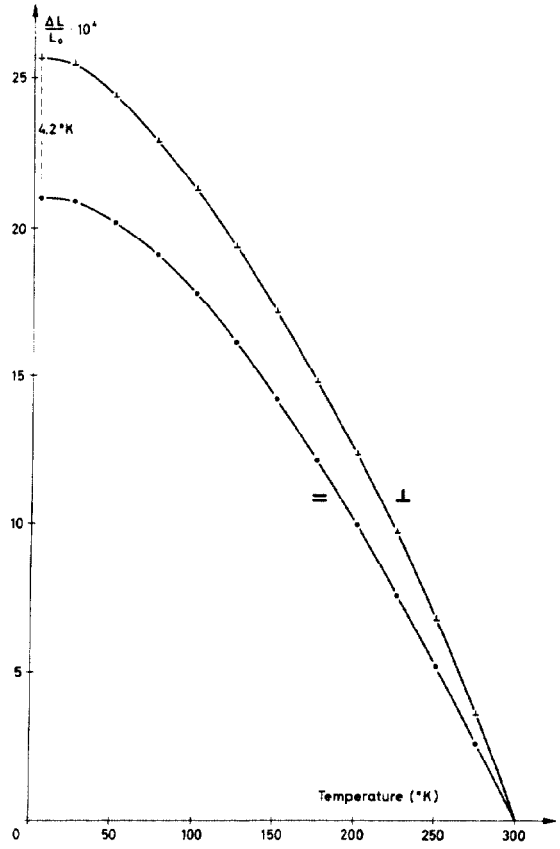
b) Linear Thermal Contraction

The linear dimensional changes of filled and reinforced materials are measured in a quartz dilatometer. It consists essentially of a fused quartz tube into which the sample is inserted. A quartz rod is then placed on top of the sample. The relatively large changes in length with temperature associated with most unfilled polymers are determined with reference to an invar-calibrated, stainless steel dilatometer. The change in length of the sample on cooling was measured with a displacement transducer. Tests were performed on samples of 90 x 20 x 6 mm^3 either in liquid helium, liquid nitrogen or gaseous helium atmospheres. It was possible to detect relative changes in length of approximately 0.0025 mm for the quartz dilatometer and 0.05 mm for the stainless steel dilatometer.

TABLE 1

Thermal contraction of polymer materials

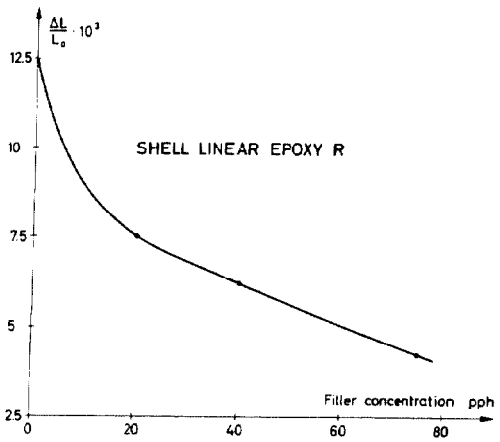
Materials	$\frac{L_{300} - L_T}{L_{300}}$			
	T = 4.2°K	T = 77°K	T=150°K	T=200°K
ELASTOMERS				
Silastic 400		0.051		
Viton RX		0.013		0.009
Neoprene L6811		0.011		0.010
Adiprene ISR1		0.021		
PLASTICS				
PTFE-GM3	0.020	0.017		
PA6-GM4	0.012	0.011		
PA6-BKV304 (Graphite filled)	0.0061	0.0055		
Makrolon-AP1	0.010			
Cryopolythene (high density)	0.021	0.018		
PE-DQDA1868 (low density)		0.030		
PPO-GM1	0.013	0.012		
RESINS				
Vespel SP1	0.0051			
Vespel SP21 (15 % graphite)	0.0049			
Various unfilled epoxies	0.014±4			
Filled epoxies	0.005±2			
Stycast 2850GT-X1	0.0041	0.0036	0.0028	0.0021
Stycast 2850GT-X2	0.0046	0.0037		
Stycast 2850FT-X3	0.0030	0.0027	0.0021	0.0015
Stycast 2850FT-X4	0.0044	0.0040	0.0030	0.0022



Thermal Contraction of Epoxy Laminate ISR 1

FIGURE 2

The results are summarized in Table 1 and Figures 2 and 3. Dimensional changes occurring as the temperature drops are important in pure polymer materials (Table 1 - 5.1 % for silastic 400 rubber between 300°K and 77°K). Thermal contraction is not as marked in reinforced and filled plastics (Fig. 2 and Table 1, resp.) and in addition, it depends to a great degree on the type[2] and quantity of the filler (Fig. 3). It should be noted that, on contraction, the filled and reinforced polymer materials may exhibit considerable anisotropy, depending on the type and direction of the incorporated



Thermal Contraction vs Filler Concentration "Quartz Powder" (300°K → 4.2°K)

FIGURE 3

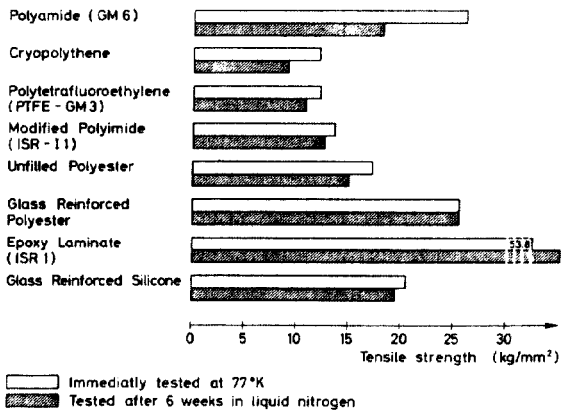
reinforced material. Contraction parallel to the direction of the glass fibres is less marked than that perpendicular to them (Fig. 2). The thermal contraction of commonly used polymers at 4.2°K is on average 10 ± 4 % higher than at 77°K.

c) Thermal ageing and cycle tests

Different polymer materials were immersed for six weeks in liquid nitrogen and mechanically tested afterwards (Table 2).

The effect of fast cool down, i.e. within two minutes (300°K to 77°K), and repeated thermal cycling between these two temperature limits was also studied for polymer materials of dimensions 90 x 20 x 4 mm³ (Table 3).

TABLE 2



Thermal Ageing Tests on Polymer Materials

TABLE 3

Influence of Thermal Cycling Tests on the Tensile Strength of Polymer Materials between Room Temperature and 77°K

(% of Initial Tensile Strength)

Materials \ Cycles	0	50	100	200	400
	Tensile strength kg/mm²	%	%	%	%
Polyamide GM6	25.7	90	82	66	59
Cryopolythene	12.0	95	90	86	75
Polytetrafluoro-ethylene(PTFE-I1)	12.2	--	--	--	85
Modified polyimide (ISR-I1)	13.5	100	100	90	80
Unfilled polyester	17.2	85	--	--	--
Glass reinforced polyester RX1	25.5	100	100	95	90
Epoxy laminate ISR1	53.8	105	110	105	105
Glass reinforced silicone RX1	26.4	95	90	80	80

2.2 Thermal Properties

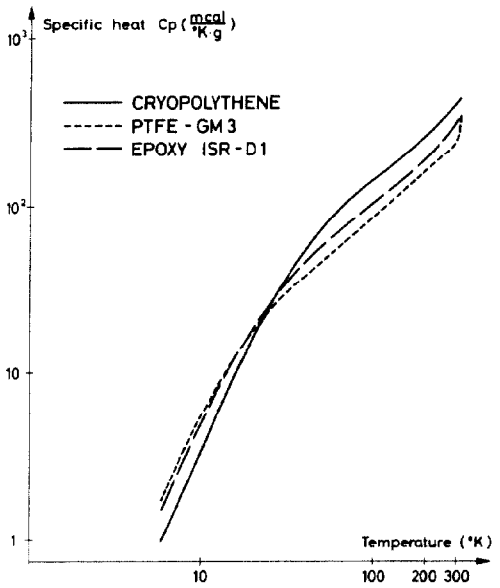
a) Specific Heat

A differential adiabatic calorimeter equipped with two sample holders, one for the reference, the other

for the sample proper is used. Absolute powers of 0.1 to 300 mW applied continuously to each sample and regulated in order to maintain a linear heating rate (1°K/min), strictly identical on both sides, are recorded. The difference of the applied powers allows the determination of the specific heat of the test sample. The accuracy of the measurement is about 5 % depending on the thermal diffusivity of the samples.

The samples measure 9.9 mm Ø and 10 mm length for tests between 4.2°K and 300°K, and 4.55 mm Ø and 10 mm length for tests between 100°K and 300°K. The first test samples are mounted at room temperature while the latter ones are mounted at liquid nitrogen temperature in order to evaluate the post-irradiation heating effects. Two tests under vacuum have been made per material[3].

The results are given in Figure 4. The effect of the chemical composition in Adiprene ISR5 and cure conditions in Epoxy ISR-F4 on the specific heat are shown in Figure 5. The solidification region is situated around 290°K for PTFE-GM3 (Fig. 4) and around 220°K for Adiprene ISR5 (Fig. 5).

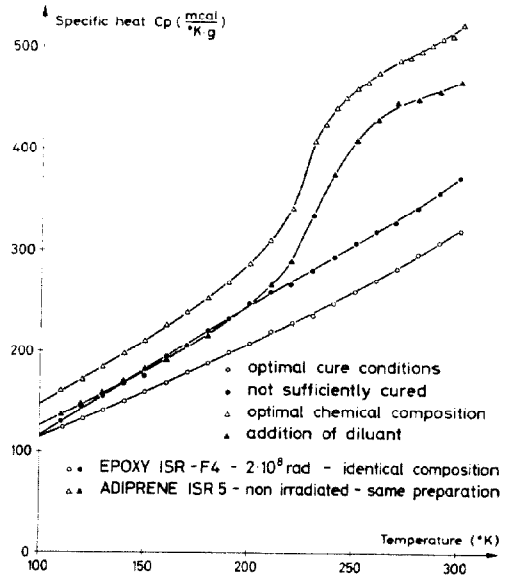


Specific Heat vs Temperature for Polymer Materials

FIGURE 4

b) Thermal Conductivity

The samples are rods of 10 mm Ø by 30 mm length and are mounted into the measuring cell at room temperature. The heat conduction is measured in the temperature range from 6°K to room temperature in a vacuum of 10^{-5} mmHg. The thermocouple junctions were in contact with thin copper rods which were pushed through the cross-section of the sample. The temperature measured was, therefore, the average cross-sectional sample temperature. The distance between the junctions of the two thermocouples used for determining the temperature gradient was 10 mm (the middle 10 mm of the sample length). This temperature difference was determined with an accuracy of ~ 0.04°C by means of Au-Fe/chromel thermocouples. The absolute temperature of the sample was measured with a precision of about 0.1°K



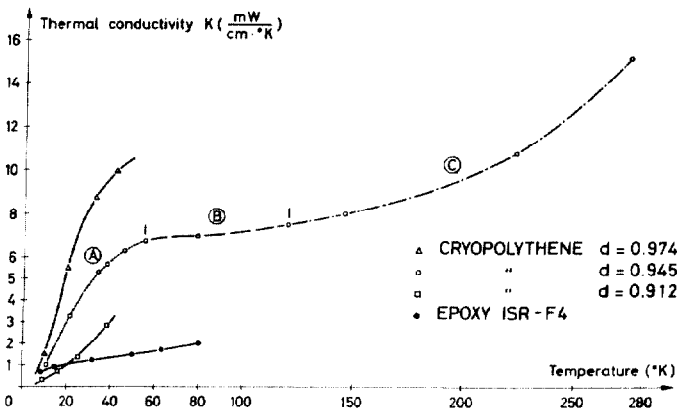
Specific Heat vs Temperature for Epoxy ISR-F4 and Adiprene ISR 5

FIGURE 5

by means of a germanium resistor between 6°K and 40°K and with a platinum resistor between 40°K and 280°K. The applied power of the furnace was in the range 1 - 10 mW with a precision of 10^{-3}. The precision of the thermal conductivity values for these polymers is ~5 %.

The test results of Cryopolythene of various densities and of Epoxy ISR-F4 are shown in Figure 6.

For Cryopolythene, the thermal conductivity increases with increasing density: this phenomenon is related to the greater crystallinity of the higher density grade (Fig. 6). Samples of similar density but obtained by different technological procedures are found to have different thermal conductivity values. The thermal conductivity of Cryopolythene is higher than that of Epoxy ISR-F4 at low temperatures, in particular in the liquid nitrogen region (Fig. 6).



Thermal Conductivity vs Temperature for Cryopolythene and Epoxy ISR - F4

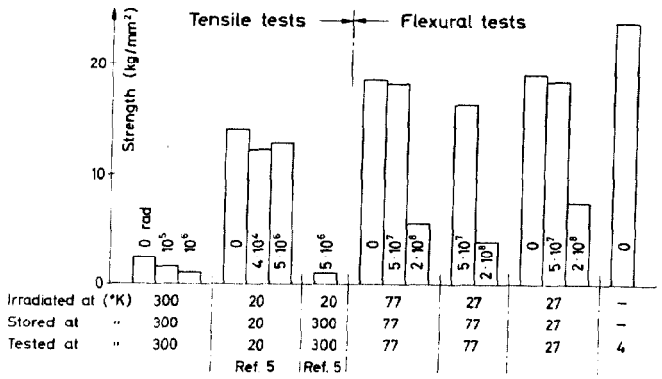
FIGURE 6

3. Radiation Resistance

All irradiations were performed in the "Melusine" water pool reactor of C.E.A., Grenoble, at a dose rate of about 5 x 10^7 rad/h. Neutron fluxes were measured with threshold detectors and gamma dose rates with a calorimeter. The polymer materials have either been exposed in liquid nitrogen and/or liquid neon[1].

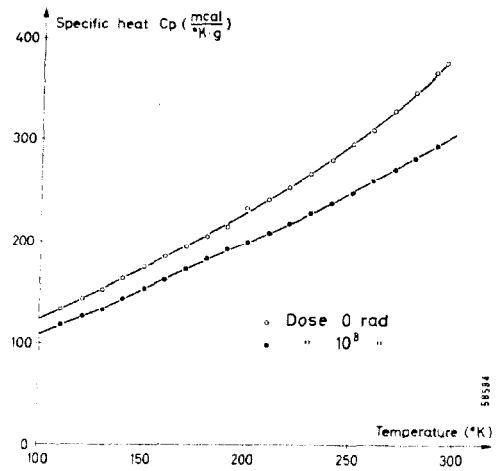
The effects of radiation at 77°K on the mechanical properties of various polymer materials are given in References 1 and 4. Table 4 shows the radiation effect on PTFE-GM3 as a function of radiation doses at different temperatures. This polymer irradiated and tested in liquid nitrogen and/or neon still had good mechanical properties after irradiation to 1 x 10^8 rad. On raising the sample temperature to ambient, severe degradation of the properties occurred. Similar effects have been reported on phenolics and teflon studied at liquid hydrogen temperature[5,6] (Table 4).

TABLE 4



Radiation Resistance of Polytetrafluoroethylene (PTFE-GM3)

Irradiated thermoplastics give a lower thermal contraction than the unirradiated ones. The maximum difference noted was 20 % for some unfilled plastics at a dose of about 5 x 10^7 rad. Epoxies and in particular mineral filled and glass reinforced ones did not show any change at this dose level.



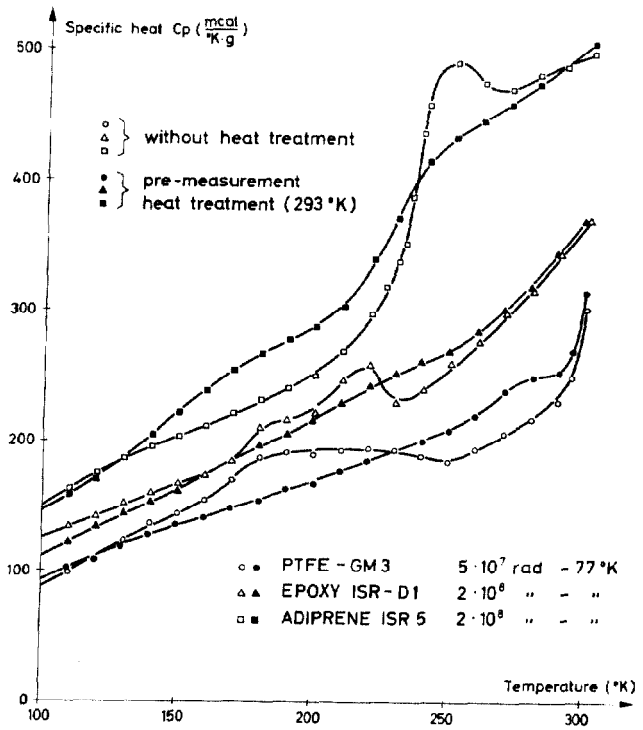
Effect of Radiation on the Specific Heat of Epoxy ISR-F4

FIGURE 7

A decrease in specific heat over the tested temperature range (100°K - 300°K) has been observed for the irradiated Epoxy ISR-F4 (Fig. 7).

The irradiated polymers showed anomalies in the Cp vs T curves if measured without a prior heat treatment. A post-irradiation heat treatment at 300°K anneals the polymer and removes these anomalies (Fig. 8).

No changes in thermal conductivity were observed for the Epoxy ISR-F4 irradiated at 1 x 10^8 rad and tested between 6°K and 77°K.



Effect of Post-Irradiation Heat Treatment on the Specific Heat of Polymers

FIGURE 8

4. Conclusion

Desired coefficients of contraction to cold can be obtained by the judicious choice of the polymer, the filler and the quantity in which it is used (Fig. 3 and Table 1). The ability to match the contraction of a polymer to a metal is useful in applications where differential contraction between polymer and metal must be minimized. The $\Delta L/L_o$ of the filled epoxy 'Stycast 2850 FT-X3' (30 x 10^{-4}) is nearly the same as OFHC-copper (32 x 10^{-4}).

A decrease in the mechanical behaviour of the unreinforced plastics was observed during the thermal ageing and cycling tests. Glass reinforced materials were not sensitive to these tests (Tables 2 and 3).

The variation of specific heat with temperature is similar for the various polymer materials studied. The specific heat is about three decades lower at liquid helium temperature compared to the value at room temperature (Fig. 4).

The material composition and manufacturing techniques seem to have an important effect on the physical properties of polymer materials at low temperatures (Fig. 5).

The heat conductivity of pure polymer materials reduces as the temperature is lowered. The materials which exhibit the greatest ductility are also the best heat conductors, e.g. compare PTFE-GM3 against Epoxy ISR-F4 (Fig. 6).

The mechanical properties of PTFE-GM3, irradiated and tested in liquid nitrogen, were found to be similar to those in liquid neon (Table 4).

Acknowledgements

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