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Thermally Stimulated Current Characteristics of Polypropylene With an Without Glass Fibres

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Summary

Small discs of melt-pressed polypropylene (PP), drawn polypropylene sheet (natural draw ratio 5.5) and short glass fibre-reinforced polypropylene sheet were polarized in a d.c. field at 100°C for 1 hr. Thermally stimulated depolarization current (TSC) characteristics of the polarized discs were studied from 50 to 170°C. The isotropic sample shows two peaks around 75 and 110°C and these peaks shift to 100 and 140°C respectively for the drawn sample. The TSC peaks of glass fibre-reinforced PP sheet are close to those of the isotropic sample but the current emitted is much higher even though it was polarized at a lower field. It is proposed that the large interface areas between the glass fibres and polypropylene matrix can accumulate charge and lead to the observed enhancement of the depolarization current.

Experimental

Polypropylene sheets were compression moulded from ICI's 'Propathene' GWM-22 at 190°C and quenched in water, at room temperature. The sheets were crystalline and had essentially random orientation, as seen by X-ray diffraction. A specimen of 2 cm gauge length was stretched uniaxially at room temperature in an 'Universal testing Machine' to its natural draw ratio (~5.5) at a strain rate of 2-3 mm/min. The drawn sheet (thickness ~ 0.06 cm.) had a slightly higher crystallinity than that of the isotropic sheet and had high axial orientation, as seen by X-ray diffraction. The axial stiffness of the drawn sheet was about four times the stiffness of the isotropic sheet; thus stretching, reinforces the sheet along the orientation direction. A similar, or even higher, increase in stiffness can be achieved by incorporating 30% by weight (~12% by volume) of short glass fibres in polypropylene and extruding the glass fibre-reinforced polyproplylene in the form of a sheet, which has the fibres oriented along the extrusion direction and then measuring the axial stiffness along this direction. The composite material used in the present studies was Profax PC 072 polypropylene granules containing 30% by weight of short glass fibres manufactured by Hercules Incorporated and the sheet thickness was 0.062 cm.

Small discs (1 cm^2) were cut out from the isotropic polypropylene sheet (PP), the stretched sheet (PPS) and the unidirectional glass fibre-reinforced composite sheet (PPC). The discs were coated on both sides with aluminium in a vacuum coating unit. Field-induced thermally

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stimulated current measurements were then made on these metal coated discs. DC fields in the 10 KV/cm to 62 KV/cm range were applied at 100 ± 2° C for 1 hr and the samples were allowed to cool down to room temperature, in the presence of the field, under the ambient conditions, by switching off the heating. The polarized discs were stored for 30 minutes. When they are reheated, they give rise to a current which is a measure of the stored polarization within the system. The TSC studies were made on a polarized specimen at a heating rate of 4°C/min and current measurements were carried out with the help of a Keithley Electrometer 610 C, which can measure currents down to 10^{-15} ampere.

Results and Discussion

Some of the data are shown in Fig. 1. It is interesting to observe that the stretched sample, polarized under a high field of 60 KV/cm, gives rise to low depolarization currents. The TSC peaks appear to occur at about 100 and 140°C. In the isotropic sample, on the other hand, the current emitted is higher, though the sample was subjected to a lower field intensity (48 KV/cm). The TSC peaks centre around 75 and 110°C for this sample. The relatively low current emission and the high TSC peak temperature in the case of the stretched sample, are both indicative of the low polarization and the more constrained relaxation of the polarized unit in this case. It is well known that orientation of crystalline polymers can shift the temperature of maximum loss to higher temperatures. The possible reasons for this are that uniaxial stretching changes and deforms the crystal structure, thus hindering charge transport and deepening the traps. Also since the

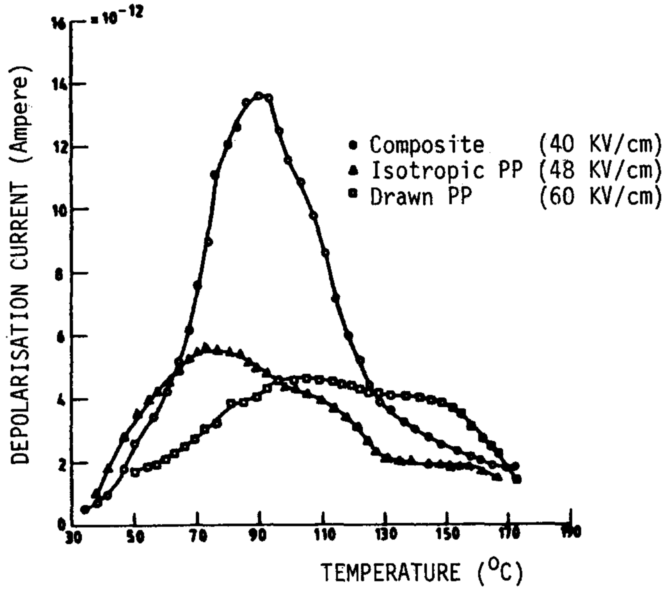

Fig. 1. The depolarisation current spectra for the three samples

intermolecular bonds are now stronger, the mobility for charge relaxation becomes less (VAN TURNHOUT, 1980). It is also known that stretched polypropylene sheets contain voids and these will also be important.

The TSC characteristics for the short glass fibre-reinforced polypropylene sample are interesting; it gives a much higher peak current for a relatively lower applied field (40 KV/cm). The TSC peak temperatures for this sample are about 90 and 105°C and are closer to the isotropic polymer sheet than to the stretched sheet, suggesting that the matrix (polypropylene) in the composite, which forms 88% of the total volume, is not as greatly affected by the glass fibres as by stretching. In the latter case, there are considerable changes in orientation and crystal morphology due to the high stretch imparted to the sheet. The high current emission in the case of the composite can partly arise from the higher polarizability of glass. However, glass fibres form only 12% of the total volume in the composite. Taken individually, the polarizability characteristics of glass fibres and polypropylene are not so different as to increase the current at the peak temperature by a factor greater than two. Also the fact that the glass fibres become more effective current raisers only at or close to the matrix peak temperature apparently indicates that the glass-matrix interface has a role to play. It is, however, noteworthy that glass of a particular composition has a dielectric loss peak at about 370°C, at 100 Hz (LEKO AND DOROKHOVA, 1965). A mechanical loss peak has been reported for another glass at about 200°C at 1Hz (JONES, 1971). The precise contribution of this relaxation process to the TSC peak of this composite sample can not be estimated for the sample under study. However, the effect of the glass fibre-matrix interface needs to be taken into account. The average length of the glass fibre in the composite is 0.025 cm. and their number in the test sample will therefore be very large ($\sim 10^5$). This additional type of polarization arises from the accumulation of the charge at the interface (SMYTH,1955) which will require the flow of current through the polypropylene and the glass fibre, a process which may require considerable time so that it may be observed only at low frequencies.

The activation energies involved in disorienting the polarisable unit have been calculated from the TSC curves (Fig. 1) using two approaches (CHEN, 1969; KHARE AND RANADE, 1975). The average values of the activation energy are 0.54 ev for the isotropic polypropylene sheet, 0.79 ev for the stretched sheet and 0.66 ev for the glass fibre-reinforced polypropylene composite sheet. This implies that the mobility is maximum in the isotropic sheet and least in the stretched sheet. In the case of the composite sample, though the mobility is lower than that of the isotropic sheet, the depolarization is higher and, as stated earlier, it can arise from the large surface of the fibre-matrix interfaces.

The equivalent frequency, f_{eq} involved in the TSC measurements, is given by (VAN TURNHOUT,1975).

$$f_{eq} = \frac{1}{2\pi} \frac{qE}{KTm^2}$$

where, q is the rate of heating, E is the activation energy, K the Boltzmann constant and Tm the peak temperature. It comes out to be about 0.0005 Hz in the present case and thus this experiment is equivalent to a low frequency dielectric measurement. The corresponding loss value ε'' at the peak temperature has been calculated (VAN TURNHOUT, 1975), and is 3.57×10^{-3} for isotropic sheet, 2.10×10^{-3} for the stretched sheet and 14.5×10^{-3} for the composite sample. The high loss in the composite, inspite of its relatively lower mobility, is again indicative of the role played in polarization and depolarization by the large surface area of the glass fibre-polypropylene interface. The dielectric loss, ε'', for the three samples was also measured at 1000 Hz and the values were found to be 4.8×10^{-3} for isotropic PP, 3×10^{-3} for the stretched sheet and 7×10^{-3} for the composite sample.

It may be stated that undrawn and drawn polypropylene sheets show a mechanical relaxation at 110^{0} and 120^{0}C respectively at 110 Hz. (JARRIGEON ET AL, 1980). In the case of the drawn sample, the peak shifts to a lower temperature (100^{0}C), when measurements are made at 3.5 Hz. This peak has been identified as a crystalline dispersion. In reinforced sheets since glass fibres act as nucleating agent, extensive crystallisation can occur around them and the imperfect boundary can accumulate charge and thus increase the depolarisation current.

In summary, the reduced segmental mobility in stretched polypropylene reduced the polarization effect though it is known to reinforce the structure mechanically. The reinforcement of polypropylene by short glass fibres not only improves it mechanically but also results in high polarization effects. The latter may be attributed to the large interface between the glass fibre and the polymer matrix which can accumulate charge and enhance the depolarization current.

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