Temperature dependence of craze shape and fracture in polycarbonate

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The shape of the craze at the tip of a loaded crack has been determined by optical microscopy for polycarbonate. The effect of temperature was examined, and measurements were made on samples of different molecular weight. In all cases the craze shape can be described to a good approximation by the Dugdale model for the plastic zone at a crack tip. The crack opening displacement depended on sample molecular weight, but was independent of temperature. Fracture toughness values deduced from the craze shape were in good agreement with plane strain fracture toughness obtained from direct cleavage fracture measurements, on the assumption that failure occurs by combined plane strain and plane stress fracture modes.

INTRODUCTION

In previous publications[1,2] it has been shown that the Dugdale plastic zone model[3] provides a good starting point for a study of the quantitative relationship between primary craze formation at a crack tip and fracture toughness in polymers. Two experimental techniques were used: first, optical microscopy on the crack tip area to determine the craze size and shape, and secondly, fracture toughness tests using compact tension (CT) specimens as described by Brown and Ward[4]. The material investigated was PMMA, Brown and Ward[1] employing two grades (a commercial grade and a plasticized grade) and Morgan and Ward[2] examining the influence of temperature on the same commercial grade. All the PMMA samples showed totally brittle behaviour under the conditions of testing used. The aim of the research now reported is to extend these investigations to a more ductile polymer, poly(bisphenol A–carbonate) (PC) to measure the fracture properties of the material when failing in a combined brittle/ductile mode. Parvin and Williams[5] have reported fracture tests on PC and interpreted these on a numerical basis by assuming that the failure is by combined plane stress and plane strain fracture modes. The same basic assumption is used in the present investigation, but the contributions of the two failure modes are now estimated experimentally from microscopical examination of the fracture surfaces and the plane strain fracture toughnesses are determined independently from the craze shape using the Dugdale plastic zone model.

EXPERIMENTAL

Materials

Measurements were made on four different grades of PC, two commercial grades and two specially moulded grades. The two commercial grades were Bayer Makrolon extruded sheet, nominally 3 mm thick and General Electric Lexan extruded rod, nominally 90 mm diameter. The two specially moulded grades, which were provided by the Ministry of Defence PERME, Waltham Abbey, were Bayer Makrolon 2803 and 2401 grades, supplied in injection moulded plaques nominally 3 mm thick. Molecular weight determinations on these polymers were carried out by RAPRA, Shawbury and the results are shown in *Table 1*.

EXPERIMENTAL

Measurement of craze shape

The measurements of craze shape were made using a specially modified Riechert microcold stage, *Figure 1*, on a Carl–Zeiss Jena reflected light microscope. Small sections of the materials were removed from CT specimens (around the crack tip area) and mounted in the stage. These could be returned to the CT test temperature (using liquid carbon dioxide) and simultaneously wedge loaded to reopen the crack and craze to breaking point. A series of photographs were taken as the wedge loading was increased and the last photograph, obtained before the crack/craze combination moved, was taken as representative of the fully loaded condition.

Fracture toughness measurements

Fracture toughness tests were carried out on CT specimens using an Instron Universal Testing Machine. An Instron Carbon Dioxide chilled environmental chamber was employed and the specimens were 50 mm square as shown in *Figure 2*.

Notching technique

The specimens were notched with a razor blade at the base of the saw cut following previous practice[1,2]. Since the materials were all much more ductile at room temperature than PMMA it was impossible to create a true brittle crack, thus all the specimens were cooled in liquid nitrogen prior to razor notching and then allowed to recover to room temperature before testing. To avoid the possibility of memory effects of this notching technique affecting results, all cracks were allowed to run at least 0.5 cm before readings were taken.

Table 1 Molecular weight characterization, with results for *COD* and craze stress

	$\bar{M}_w$	$\bar{M}_n$	Number of fringes in craze: Loaded	Number of fringes in craze: Relaxed	*COD*, δ_t(μm)	Calculated craze stress at −30°C, σ_0 (MN/m²)
Makrolon sheet	20060	9500	42	32	8.9	120
Makrolon 2803	20000	9500	42	32	8.9	120
Makrolon 2401	15000	7900	32	24	6.8	96
Lexan Rod	17000	4000	27	21	5.8	110

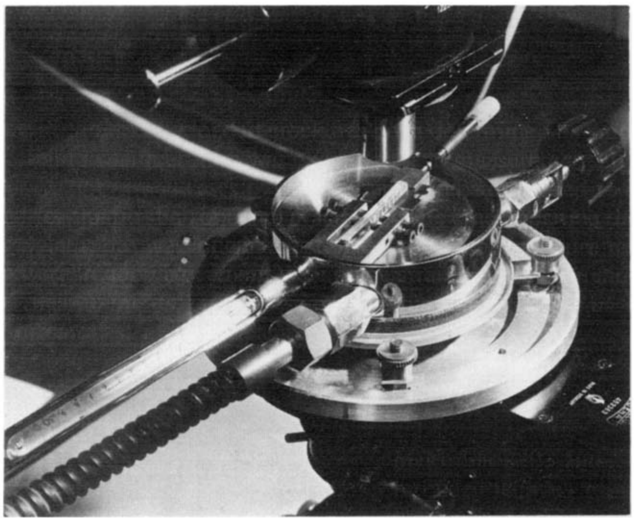

Figure 1 Photograph of experimental set-up

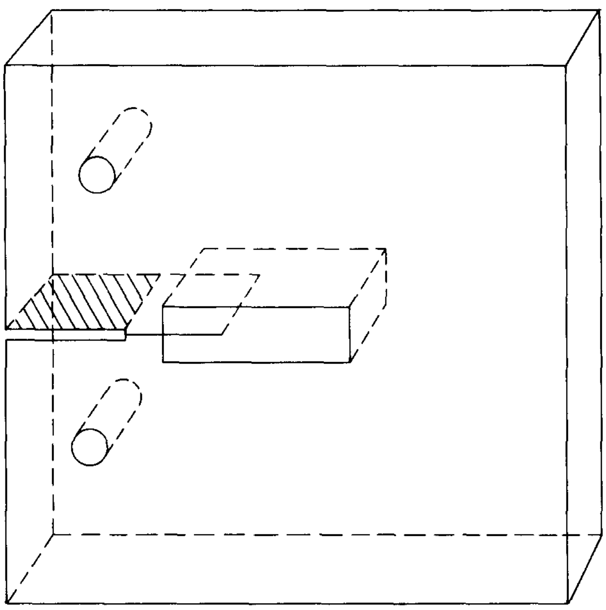

Figure 2 The compact tension specimen showing centre portion which is removed for optical microscopy

The various test series were as follows: commercial grade Makrolon 3 mm thick specimens from −30° to −70°C, Lexan specimens 6 mm thick from +22.5° to −70°C, Lexan specimens 2.5 to 10.25 mm thick at −20°C and the two specially moulded grades of Makrolon at −30°C.

The CT tests were analysed in the manner detailed previously by Brown and Ward[1], to obtain the reduced moduli of the materials tested at the various temperatures used. Crack speeds were measured as the crack progressed across the specimens and the results presented here are for a nominal crack speed of 2.0 cm/min.

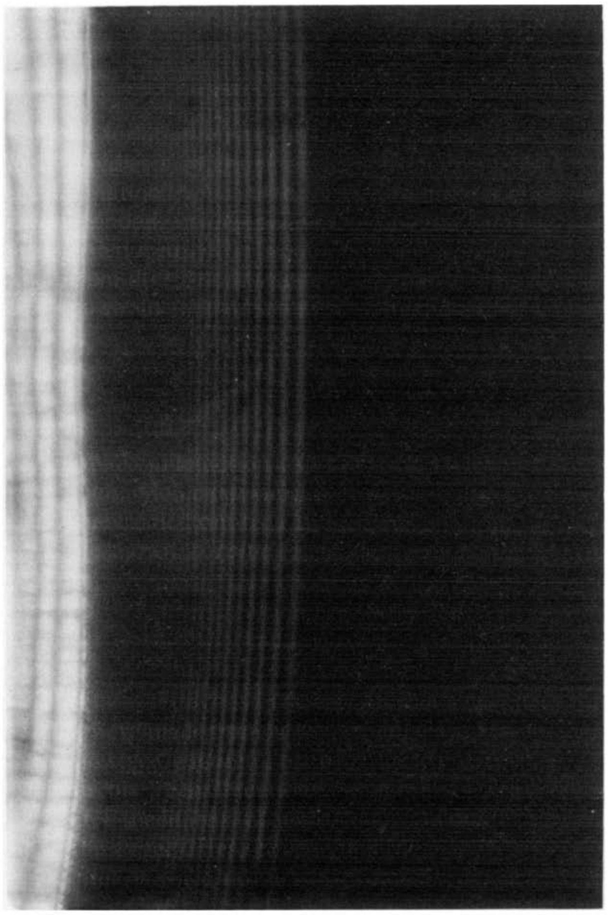

Figure 3 Photograph of the interference pattern observed in a typical wedge loaded crack in PC

RESULTS

Comparison of craze shape with Dugdale plastic zone model

The optical microscopic examination described was performed on the primary crazes of all the test specimens under the same conditions as those chosen for the fracture toughness tests so that direct comparisons of results from the two types of measurement could be made. For all specimens, craze lengths and shapes were measured, as were the crack opening displacements (*COD*), by examination of enlarged prints of the reflected interference fringes, an example of which is shown in *Figure 3*. It will be noted that in contrast to the previous work on PMMA many more fringes were present which made this direct method preferable to the microdensitometer examination of the film negative.

As an example, the loaded fringe pattern for the commercial grade Makrolon at −30°C is shown in *Figure 3*. We assume a value of refractive index of 1.27 as reported by Kambour[6]. This will change by less than 1.5% over the temperature range employed[7]. It is then possible to plot the craze shape following the technique of Brown and Ward[1]. This shape may be compared with that predicted by the Dugdale plastic zone model as developed by Rice[2,8]. The result is shown in *Figure 4*. As in the previous work on PMMA, the refractive index along the craze length is assumed to be constant. It is to be noted that *Figure 4* includes the unloaded craze shape. The fact that this is very close to being a constant proportion of that for the fully loaded craze gives good support for our assumptions.

To find the absolute value of *COD*, δ_t we require the value of the refractive index of the craze at break μ_B. Following the method of the previous paper[1], it can be shown that this is related to the refractive index of the relaxed craze μ_0 by the relationship:

$$\frac{\mu_B \lambda}{\mu_0} = \frac{n_b}{n_0} = 1 + \frac{\lambda - 1}{1.27} \qquad (1)$$

where λ is the extension ratio of the craze at break and n_b and n_0 are the number of fringes in the loaded and unloaded crazes, respectively. It was noted that for all the tests the ratio $n_b : n_0$ remained essentially constant and hence the

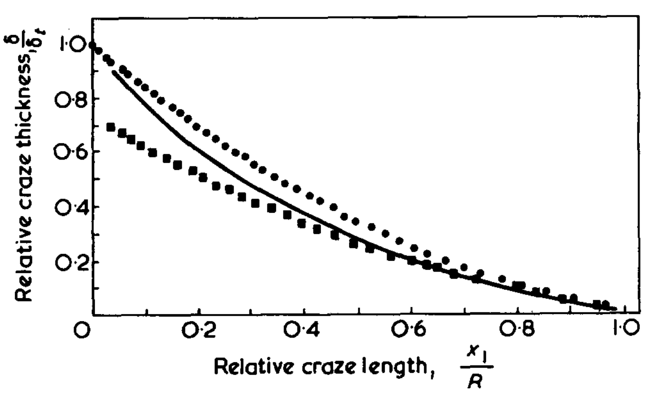

Figure 4 Craze shape as measured from interference pattern compared with the best fit to the Dugdale plastic zone model. ●, Loaded; ■, unloaded, ——, Dugdale zone analysis

value of λ remained constant at a value of 1.41.

The results of these tests are summarized in *Table 2* for the various materials at the various temperatures used. Included in this table of results are calculated values of fracture toughness G_{IC} and craze stress σ_0 calculated from the relationships given by the Dugdale plastic zone model:

$$\delta_t = \frac{8\sigma_0 R}{\pi E^*} = \frac{G_{IC}}{\sigma_0} \qquad (2)$$

where, R is the craze length and E^* is the reduced modulus (equal to Young's modulus, E in plane stress and $E/(1 - \nu^2)$ in plane strain where ν is Poisson's ratio).

The value of E^* used in these calculations is calculated from the compliance of the fracture toughness specimens as detailed in previous publications[1,2,4].

FRACTURE TOUGHNESS TESTS

The measured overall toughness of the materials tested is shown in *Table 2* and it is apparent that the values are much higher than those predicted by the Dugdale plastic zone model for the primary crazes. On examination of the failed specimens it became apparent that the fracture surfaces were not totally flat as found in PMMA[1,2], but considerable shear lips were present as shown (exaggerated for clarity) schematically in *Figure 5*. The measurements of these widths are also included in *Table 2*. From the optical microscopy of the crack tip area it could be seen that the central portions of the specimens were failing in plane strain (by brittle cleavage through crazing) whereas the mode of failure in the shear lips was by ductile yielding and eventual rupture. These observations are similar to those reported for the same materials in the impact test by the authors[9] and in tensile fracture tests by Parvin and Williams[5] where the two modes of failure are predicted and their values estimated but are not associated directly with any physical phenomena as reported here.

If two different critical strain energy release rates G_{ICE} and G_{ICS} are assigned to the plane strain and plane stress

Table 2 Collected data from craze shape measurements and compact tension fracture toughness tests

Material	Temperature (°C)	Thickness, W (mm)	Measured COD, δ_t (μm)	Measured craze length, R (μm)	Calculated modulus, E^* (GN/m^2)	Fracture toughness calculated from craze shape, G_{ICE} (kJ/m^2)	Craze stress calculated from craze shape, σ_0 (MN/m^2)	Measured overall fracture toughness, G_{ICO} (kJ/m^2)	Measured shear lip width, w (mm)	Calculated plane stress fracture toughness, G_{ICS} (kJ/m^2)
Makrolon sheet	–30	2.97	8.9	83.2	2.78	1.12	115	5.8	0.69	21
	–40	3.07	8.9	94.5	3.05	1.09	110	3.7	0.49	17
	–50	2.96	8.9	94.5	3.20	1.15	115	3.3	0.43	16
	–60	3.03	8.9	99.0	3.40	1.16	120	3.0	0.37	15
	–70	3.07	8.9	91.9	3.44	1.27	130	2.8	0.35	15
Makrolon 2803	–30	3.14	8.9	85.5	2.87	0.94	120	3.1	0.44	16
Makrolon 2401	–30	3.17	6.8	70.3	2.45	0.57	96	2.2	0.34	16
Lexan rod	+22.5	6.48	5.8	101.5	2.59	0.37	58	4.6	1.103	24
	0	5.75	5.8	70.0	2.86	0.59	93	2.3	0.454	23
	–11.5	5.68	5.8	64.0	2.81	0.64	100	1.9	0.392	18
	–20	6.10	5.8	62.5	2.89	0.67	105	1.7	0.375	15
	–30	6.04	5.8	61.6	2.95	0.70	110	1.5	0.320	16
	–40	5.73	5.8	60.4	3.02	0.72	115	1.6	0.284	18
	–50	5.81	5.8	59.2	3.14	0.77	120	2.0	0.220	32
	–60	6.08	5.8	58.0	3.22	0.81	125	2.1	0.150	55
	–70	6.03	5.8	57.8	3.29	0.83	130	2.3	0.137	67
	–20	3.40	5.8	62.5	2.89	0.67	105	2.6	0.375	16
	–20	8.25	5.8	62.5	2.89	0.67	105	1.7	0.375	15

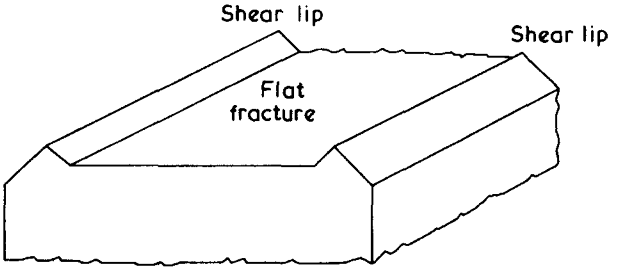

Figure 5 Schematic diagram of fracture surface in PC showing shear lips

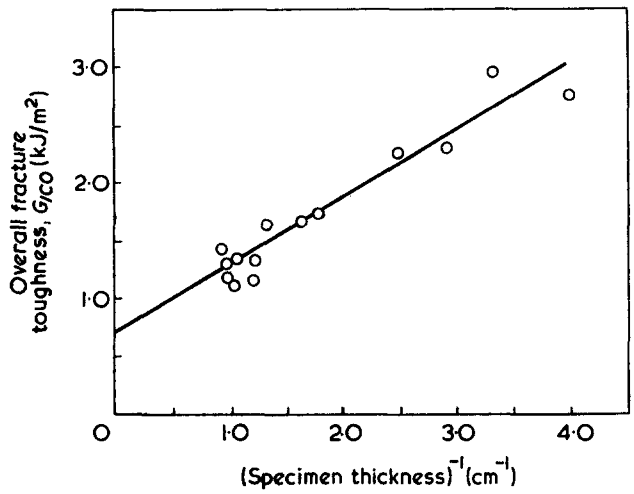

Figure 6 Overall toughness G_{ICO} as a function of specimen thickness

areas, respectively, these may be related to the overall critical strain energy release rate (G_{ICO}), as measured in the CT test, by the equation:

$$G_{ICO} \times W = G_{ICE} \times (W - w) + G_{ICS} \times w \quad (3)$$

where W is the specimen width and w the total width of the shear lips. This model is similar to that proposed by Bluhm[10] for a titanium alloy but differs in that the latter formulation assumes both modes of failure to be both stress and strain dependent.

For the Lexan specimens of various thickness tested at −20°C, the overall toughnesses are shown in *Figure 6*, plotted against inverse specimen thickness. These results will be discussed in detail below.

DISCUSSION

Table 2 shows the measured values of *COD* (δ_t) for all the specimens tested. It can be seen that δ_t remains constant for each material over the temperature ranges used (and also over the various widths used for the Lexan material). However the value of δ_t does vary considerably with molecular weight between the various materials tested. From the molecular weight determinations in *Table 1*, δ_t appears to correlate best with $\bar{M}_n$, falling as $\bar{M}_n$ decreases.

Craze stress values calculated from equation (2) are listed in *Table 2*, and shown as a function of temperature in *Figure 7*. As found previously for PMMA[2], the craze stress falls quite rapidly with increasing temperature. It seems likely that there may be differences between the craze stress for different grades, and this is now being examined in a separate investigation.

Table 2 also shows the calculated values of plane strain fracture toughness (G_{ICE}) given by the Dugdale plastic zone analysis. For easy comparison, the temperature dependence of G_{ICE} can be seen in *Figure 8*. As to be expected from the previous consideration of *COD* and craze stress, G_{ICE} increases monotonically with decreasing temperature and there is a clear molecular weight effect.

The values of overall fracture toughness measured in the CT tests are now to be compared with the values of fracture toughness calculated from the Dugdale plastic zone model (columns 9 and 7, respectively in *Table 2*). As will be anticipated, the former are much larger than the latter. We will therefore proceed by first using the results of the tests carried out at −20°C for several different width specimens, to examine the two mode fracture model originally proposed by Parvin and Williams[5]. *Figure 6* shows the values of overall measured toughness plotted against inverse specimen thickness and the line shown is the best fit to these points using a least squares fit. This line predicts a plane strain toughness value (G_{ICE}) of 0.679 kJ/m^2 (i.e. where $1/w = 0$) and a plane stress toughness value (G_{ICS}) of 16.28 (where $1/w = 26.67$ cm^{-1}) i.e. $w = 0.0375$ cm—the width of the shear lips. This value of plane strain toughness is very close indeed to the value of 0.67 kJ/m^2 predicted by the Dugdale plastic zone analysis of the results from the optical microscopy of the primary craze.

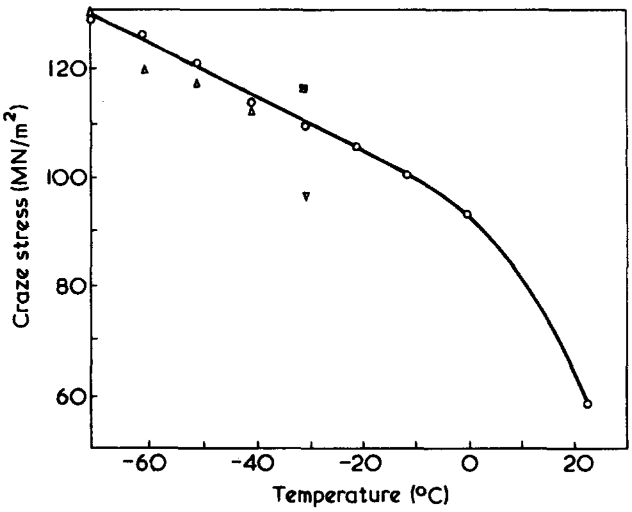

Figure 7 Calculated craze stress σ_0 as a function of temperature. ○, Lexan rod; △, Makrolon sheet; □, Makrolon 2803; ▽, Makrolon 2401

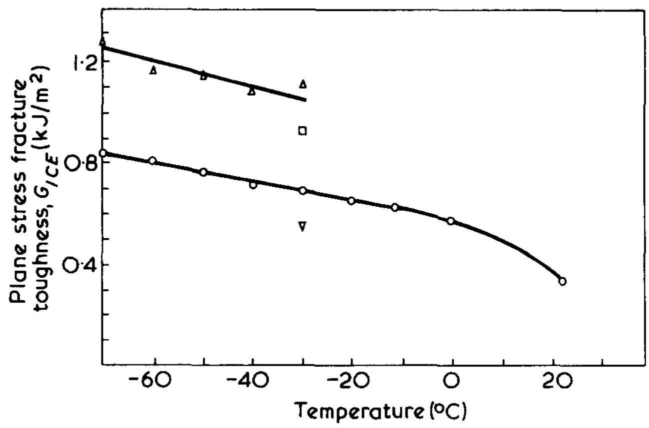

Figure 8 The temperature dependence of the plane strain fracture toughness G_{ICE}: ○, Lexan rod; △, Makrolon sheet; □, Makrolon 2803; ▽, Makrolon 2401

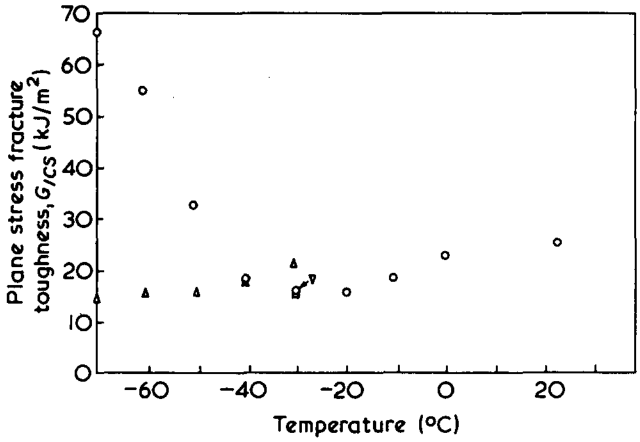

Figure 9 The temperature dependence of the plane stress fracture toughness G_{ICS}. ○, Lexan rod; △, Makrolon sheet; □, Makrolon 2803; ▽, Makrolon 2401

Further confirmation of the consistency of the two approaches is given by the G_{ICS} values for the Lexan Rod at $-20°C$. Calculations using the Dugdale Zone Analysis give values of 15.54, 15.08 and 15.42 kJ/m^2 for narrow, medium and thick specimens, respectively and the value from *Figure 6* is 16.28 kJ/m^2. Since this latter value is found by considerable extrapolation of the mean line some error is to be expected but the agreement is still very good.

With this direct confirmation of the validity of equation (3) we now proceed to use the model of additive fracture toughnesses to calculate the plane stress toughnesses (G_{ICS}) for all the specimens examined. This calculation combines the Dugdale Zone Analysis calculated value for G_{ICE}, the CT test measurements of overall toughness (G_{ICO}) and the measured specimen width and shear lip sizes. The results are also shown in *Table 2* and summarized for easy comparison in *Figure 9*.

CONCLUSIONS

The shape of the craze at the top of a sharp crack in polycarbonate is in good approximate agreement with that predicted on the basis of the Dugdale plastic zone model. The crack opening displacement has been shown to be constant for a given grade of polymer, independent of temperature. It does, however, appear to depend on the polymer molecular weight, decreasing with decreasing $\bar{M}_n$.

Values for the plane stress fracture toughness G_{ICE} calculated from the craze shape have been used in conjunction with compact tension fracture toughness tests and direct measurements of the shear lip width to verify the validity of the two mode fracture model. Values for the plane stress fracture toughness G_{ICS} were obtained and for the Lexan rod these compare well with the results of Parvin and Williams[5]. Both results show increasing toughness with decreasing temperature below about $-30°C$. However the Makrolon sheet material tested in this investigation apparently shows decreasing toughness in this region. We attribute this observed result to thin oriented surface layers on the extruded sheet which interfere with the fracture processes particularly at lower temperatures where the shear lip widths are comparatively small. Two reasons are given for this line of argument. First, in bending beam modulus measurements on the material, high modulus surface layers are apparent and secondly the Makrolon sheet and Makrolon 2803 materials (almost identical in molecular weights) show different shear lip widths at similar temperatures (as shown in *Table 2*).

ACKNOWLEDGEMENTS

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