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**FRACTURE TOUGHNESS AND CRAZING BEHAVIOUR OF
VIRGIN AND FILLER ADDED POLYSTYRENE FILMS**

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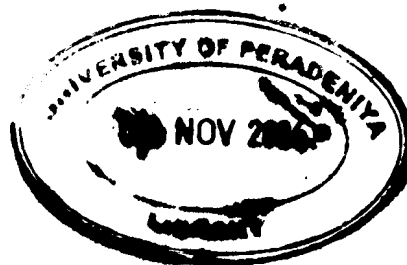
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Brittle fracture commonly observed in many glassy polymers under tension is due to the formation and breakdown of crazes. Fillers exist in a variety of systems such as organic, biological and polymeric materials. In polymer systems, nanoscale fillers not only reduce the cost of the material but also improve mechanical properties, such as hardness and tear resistance. The broad aim of this study was to investigate the fracture behavior of virgin and filler added polystyrene thin films by the trouser leg tearing test. Critical strain energy release rate (G_c) can be regarded as a material property, often equated with toughness.

The thin films, required for the tearing test, were prepared by two different methods. In the first method, a glass slide was dipped and drawn with different speeds using a motor driven lifter through a polystyrene solution. The other method was spin casting, in which the prepared solution was placed on a clean glass slide and spun with different speeds using a photo resist spinner. Toluene was used as a solvent. The virgin polymer films were prepared with a moderate solution of 14 wt-%, of polystyrene in toluene. A solution of 14 wt-%, of polystyrene and filler in toluene was used for filler added films preparation. The composition of the filler - added films was varied from 0 to 5 wt-% of fillers with respect to polystyrene content. In this study, carbon black (0.05 μm), alumina (0.075 μm) and fumed silica (255 m^2/g) fillers were used.

The slides were dried in a vacuum desiccator oven at 80°C for 12 hours for complete evaporation of toluene from the films. The samples used in this study were of rectangular shape (40 mm x 15 mm). They were stripped off from the substrate onto the surface of a



water bath. The thickness of the films was measured using a Michelson interferometer and a locally made thickness measuring instrument. A cut of 10 mm was made along the center line of the sample and two free ends were separated at constant rates using a mechanical testing machine. Tearing was done in two different directions, one parallel and the other perpendicular to the film drawn direction.

The study revealed that the G_c values of virgin polystyrene were strongly dependent on the film drawing speed. When the films were torn parallel to the film drawn direction, there was a significant increase in G_c , with increasing film drawn speed from 0.5 cm min^{-1} to 2.5 cm min^{-1} and there after almost a plateau, was observed. On the other hand, value of G_c for all the samples torn perpendicular to the film drawn direction, there was a clear decrease in G_c with increasing film drawn speeds. The value of G_c for the films which were prepared by spin casting, torn in both perpendicular directions is 0.24 kJ m^{-2} . This is almost constant for all the films which were prepared with different spin speeds and this value is comparable with the G_c value corresponding to the above mentioned plateau. The reason for such a variation in the above films could be the high degree of orientation of the polymer chains in the film drawing direction due to the very slow drawing speed. When the drawing speed was increased the degree of polymer chain orientation decreases and becomes randomly oriented.

The study also revealed that a significant decrease in G_c with the increasing filler wt-%, in the films which contained alumina and carbon black fillers, prepared by both methods. On the other hand, the films which contained fumed silica and torn in the film drawn direction showed a decrease in G_c value with increasing filler wt-%. In contrast, an increase in G_c was observed, when the fumed silica added films were torn perpendicular to the film drawn direction. The spin cast films also showed an increase in G_c with fumed silica filler wt-%. The most likely reason why such a decrease in G_c was observed in alumina and carbon black filler added films is the propagation of the fracture mostly along the filler particles. It can be speculated that there may not be any extra - ordinary interaction between the polymer and fillers. Unlike other fillers, fumed silica has a chain - like particle morphology. The decrease in G_c value in the fumed silica added films which were torn parallel to the film drawn direction may be due to the fumed silica chains and polystyrene chains aligning parallel to the film preparation direction. Tearing parallel to the direction of preferential alignment requires the rupture of fewer bonds as orientation increases and

hence, less stress is needed to tear the film in this direction. The polymer chains and fumed silica chains, in the spin cast films, were randomly oriented. These fumed silica chains give additional strength to the polystyrene.

Optical microscopic study shows the formation of craze areas ahead of the crack tip in a torn virgin polymer film. In addition to this, micro shear bands were also observed ahead of the craze region and on the sides of the fracture surfaces. The craze area ahead of the crack tip in torn polymer films showed birefringence indicating that there exists a certain degree of orientation of polymer chains in these areas. It was also observed that less amount of crazes in the alumina filler added films. Though the fumed silica added films which were torn perpendicular to the film drawn direction showed higher G_c values, the amount of craze in the films was less compared to the virgin film. On the other hand, the films which contained carbon black fillers showed very low G_c value and no crazes were observed. Unlike cracks, crazes are load bearing since their surfaces are bridged by many fine fibrils. Crazes are the major toughening mechanism in the polystyrene amorphous polymer.