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MICROSTRUCTURAL PROCESSES IN THE DEFORMATION AND FRACTURE OF POLYMERS.

Final Report.

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Fracture of shear bands in polystyrene at intersections, by tension and crazing, by shear, and by tensile fracture was studied. The fibrous material in the shear band was elongated and deformed during tensile fracture. They were completely recovered after annealing. The propagation of shear bands was observed by high speed (6000 frames/sec) cinematography. Three different obstacles were introduced, an existing shear band, a fully recovered thick band, and a region dispersed with 2-6μm rubber particles (a strip of HIPS). Except for the second one which behaved as though it was not there, the other two obstacles effectively reduced the speed

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of propagation of the shear band packet and changed their mode of operation. Transport of methanol in deformed and undeformed PMMA was compared. Transport rate was enhanced by deformation and obeyed Case I behavior while that of undeformed PMMA obeyed Case II. Desorption and resorption kinetics were all Fickian in the beginning regardless of the mechanical history before sorption.

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INTRODUCTION

Except for some partial support which started in 1973, the project at Rochester on Microstructural Processes in the Deformation and Fracture of Polymers started in 1976 for a 3 year period and then renewed for one year. In the final report dated December 1979, the achievements were summarized for DAAG29-76-G0314 and DAAG29-78-G0159. The major headings in that report were:

1. Slip processes in the deformation of polystyrene
2. Pressure and normal stress effects on shear yielding
3. Some observations of coarse shear bands
4. Intersection of coarse shear bands in polystyrene
5. Thick shear bands in polystyrene

Also in that report, there listed 6 published papers, 1 accepted paper, 3 papers in preparation, 7 abstracts for oral presentation at national meetings, 1 abstract for oral presentation at an international meeting and 3 invited local seminars.

For this one year renewal period, we continued on these studies with many new results some of which were summarized in a progress report dated December 31, 1980, a copy of which is attached. In this period, we had 2 published papers, 2 accepted papers, 1 submitted paper, 2 papers in preparation, 3 abstracts for national meetings, and 2 invited lectures and seminars. In addition to the principal investigator, we had one post doctoral investigator, one graduate assistant, and one undergraduate assistant. The important results are summarized in this final report.

FRACTURE OF SHEAR BANDS

Several modes of fracture of and at thick shear bands in polystyrene formed by compression were studied. These include fracture at shear band intersections, cracking and crazing of shear bands, crack formation inside shear bands, shear fracture of thick shear bands, and tensile fracture of thick shear bands. Some findings follow:

1. Cracks are formed at shear band intersections due to internal stresses created by mutual interactions of the shear bands. Most of the cracks were along the shear bands.

2. The tensile stress needed to open up shear bands is less than that required for undeformed material.

3. Relaxation after deformation could cause cracking between strands of fibers in a shear band.

4. Shear fracture along the shear band during compression takes place not inside the band but along either of the two interfaces between the band and the undeformed material.

5. Shear bands formed in compression showed no evidence of deformation during subsequent tension before cracking. After cracking, the strands of fibers were pulled apart and elongated by the tensile force. These elongated fibrous sheets could be recovered completely at 110 C for 3 hours to return to their undeformed state. Projected area measurements showed no change for each of these fibrous sheets before and after recovery. The shear band thickness after recovery was also the same as that before tensile fracture. Thus the fibrous sheets were not pulled out from outside the shear bands during tension. They were formed by plastic

deformation of the shear band material.

PROPAGATION OF SHEAR BANDS THROUGH OBSTACLES

High speed cinematography (up to 6000 frames/sec) as well as optical and electron microscopy was used to study the propagation and formation of thin and thick shear bands with and without obstacles in their paths of propagation. Three different kinds of obstacles were studied, an existing thick band, a fully recovered thick band and a region dispersed with 2-6μm rubber particles (a strip of HIPS). Except for the fully recovered thick band which behaved like undeformed PS, the other two obstacles effectively reduced the speed of propagation of the shear band packet and changed their mode of operation by dispersing them into thin bands and spreading them out into larger spaces. However, the original localized mode of operation resumed after the shear band packet passed through the obstacles. Some results follow:

1. When atactic PS was compressed at 0.12/sec. strain rate, a shear band packet consisting of thin coarse bands developed at stress concentrations. A single resolvable coarse band was observed to propagate between 23 and 185 mm/sec. with an average speed of 80 mm/sec. Then the number of thin coarse bands in the packet became so numerous that the packet became a thick band by a large scale shear motion which seemed to take place throughout the packet. Such shear motion had an average shear rate of about 6 mm/sec. corresponding to a shear strain rate of 24 $sec.^{-1}$.

2. When a propagating coarse band packet met an existing shear band, the packet dispersed itself into thinner bands and spread out to cover more spaces. At the same time the rate of propagation slowed down to about one half of the original speed. Individual thin coarse bands seemed to propagate in the fiber or microcrack directions in the existing band. After the thin coarse bands passed through the existing band, they joined together to form a thick packet which resumed the high speed of propagation. When the large scale shear motion took place, the shear displacement was about the same throughout the packet but the shear strain was much smaller at and near the intersection. The displacement rate, 2 mm/sec., was also smaller than the case without the existing band as an obstacle (6 mm/sec.).

3. A fully recovered shear band did not offer any observable resistance to the propagation of a new band packet, both in surface morphology and in the speed of propagation.

4. A region of HIPS which had a dispersion of 5% 2-6μm rubber particles did offer considerable resistance to the propagation of a shear band packet. First, it slowed down to about 1/5 of its speed even before approaching the HIPS region. This reduced speed caused spreading of the thin bands. Secondly, the thin bands "disappeared" into the HIPS region or they were transformed into discontinuous fine bands which sometimes clustered between the rubber particles. These fine bands recombine into thin bands which joined and gathered together into a band packet outside the HIPS region and resumed the speed of propagation.

5. These observations established the concept of a shear front in the propagation of shear bands which enables us to use

the properties of dislocations to understand the behavior of propagating shear bands in meeting obstacles such as an existing shear band and a HIPS region.

REMARKS ON SHEAR MECHANISMS

The set of experiments on the propagation of shear bands through obstacles has some similarity with such experiments on crystalline substances. For example, the slip lines in metals also grow and propagate such as reported in Al by Chen and Pond (Trans AIME 194, 1085-92 (1952)) and by Becker and Haasen (Acta Met. 1, 325-35 (1953)). The speed of propagation was not uniform. Some distribution of such speeds was reported more recently by Pond ("The Inhomogeneity of Plastic Deformation" ASM Seminar, 1971, Publ. 1973, pp. 1-16) with data on Cu and α-brass. While the smallest speed could be zero, the largest speed observed was about 0.4 - 4μm/sec. However, speed as high as 10-70μm/sec. was observed by Becker and Haasen. Instead of slip lines, the velocity of dislocations using etch pits ranges from 10^{-5} to 10^{4} cm/sec. The speed observed here in PS, 10-20 cm/sec., is sufficiently high, suggesting collective motion of atomic groups or molecules during the propagation of shear bands.

Such collective motion which travels by a front like a dislocation can interact with another front traveling in different directions. An easy way of illustrating such interaction is by the use of dislocations. The interaction produces a new front which may be immobile such as the Lomer-Cottrell lock. These immobile or slow-moving fronts could be used to explain our observation here for the slowing down of new shear bands by existing shear bands. The propagation inside the existing shear band takes place by the motion of a slow-moving new front produced by the interaction of the two shear bands.

Similar to the behavior of dislocations, the shear fronts can change their directions of propagation upon meeting obstacles such as the rubber particles in HIPS and thereby disperse themselves. Some early examples of fine slip observed in precipitation-hardened Al are reported by Carlsen and Honeycombe (J. Inst. Metals 83, 449-54 (1954-5)), Greetham and Honeycombe (J. Inst. Metals 89, 13-21 (1960-1)) and Thomas and Nutting (J. Inst. Metals 86, 7-14 (1957-8)). While the slip lines are coarse and straight in supersaturated solid solution of Al-4.5w/o Cu alloy (no precipitates) deformed at 77 K, they are fine and wavy in aged alloys (with precipitates). Such change of slip line patterns was explained by Hirsch (J. Inst. Metals 86, 13-14 (1957-8)) using the cross slip property of screw dislocations. A similar explanation can be advanced here since the concept of propagating shear front seems to have been established by the observations made in the present research.

METHANOL TRANSPORT IN DEFORMED PMMA

Gravimetric sorption studies of methanol transport in cross-linked deformed and undeformed PMMA samples indicate that the mechanical history of the polymer plays an important role in determining the transport kinetics of the system. Sorption, desorption and resorption experiments were conducted at temperatures from 35-50 C. Sorption rates are clearly accelerated for the deformed samples in the initial sorption cycle. Kinetics, in addition, are Fickian for deformed samples, while undeformed samples sorb at much slower rates, probably controlled by a relaxation process at the penetration front. Upon sorption, the compressed samples recover and swell to the same thickness as the sorbed undeformed samples. Desorption kinetics are Fickian regardless of any prior exposure to mechanical testing. Resorption of methanol in undeformed samples reveals that kinetics approach the behavior observed in the initial sorption cycle for deformed samples. All of this indicates that the deformation or rearrangement induced in undeformed samples as a result of the sorption process is similar to the rearrangement taking place during mechanical deformation. Some results follow:

1. Absorption of liquid methanol into undeformed PMMA at 35-50 C approaches Case II at lower temperatures while it is Fickian or Case I in PMMA deformed 66% by compression and annealed at 60 C for 12 hours. The time needed for saturation is about twice in the undeformed samples than in the deformed samples.

2. Samples saturated with methanol were desorbed by immersing in cyclohexanol. The early desorption kinetics are Fickian in both deformed and undeformed samples. The diffusivities are comparable to those of absorption in deformed samples.

3. Resorption after desorption again shows early Fickian kinetics in both the deformed and undeformed samples. The diffusivities are again comparable.

4. The results indicate that the critical step in Case II

transport is molecular rearrangement at the penetration front involving segmental motions probably similar to those in plastic deformation. Once rearranged either by transport or by plastic deformation, the polymer matrix is ready for methanol diffusion as revealed by the desorption and the resorption measurements.

5. Recovery may accompany swelling so that the deformed structure may return somewhat to the undeformed state.

LIST OF PUBLICATIONS

I. Published papers:

1. Morphology and Annealing Behavior of Thick Shear Bands in Polystyrene, C.C. Chau and J.C.M. Li, J. Mat. Sci. 15, 1898-1906 (1980).

2. Chemical Potential for Diffusion in a Stressed Solid, J.C.M. Li, Scripta Met. 15, 21-8 (1981).

II. Accepted papers:

1. Fracture of Shear Bands in Atactic Polystyrene, to appear in J. Mat. Sci.

2. Diffusion of Methanol in PMMA Shear Bands

III. Submitted papers:

1. Propagation of Shear Bands Through Obstacles in Atactic Polystyrene

IV. Papers in preparation:

1. Reverse Shear of Thick Shear Bands

2. Methanol Transport in PMMA: The Effect of Mechanical Deformation

V. Abstracts for Oral Presentations at National Meetings:

1. Fracture of Shear Bands in Polystyrene, C.C. Chau and J.C.M. Li, Bull. Am. Phys. Soc. 25, 378 (1980).

2. Obstacles to the Propagation of Shear Bands in Polystyrene, C.C. Chau and J.C.M. Li, Bull. Am. Phys. Soc. 25, 378 (1980).

3. Transport of Alcohols in Deformed PMMA, J.P. Harmon and J.C.M. Li, Conference on Structure and Mobility in Molecular and Atomic Glasses, The New York Academy of Sciences, December 9-11, 1980.

VI. Invited Lectures and Seminars:

1. Shear Bands in Polymeric Materials, J.C.M. Li, at a conference on "Irreversible Deformation Processes in Polymers" Asilomar, California, February 19-22, 1980, Sponsored by the Army Research Office and organized by Eric Baer, Anne Hiltner, and Roger Porter.

2. Shear Band Behavior in Polymers, J.C.M. Li, Case Western Reserve University, May 8, 1981.

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Principal Investigator: J.C.M. Li

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