## FAILURE OF TRANSPARENT PLASTICS IN LASER PULSES AT VARIOUS TEMPERATURES

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High-intensity light flashes cause internal damage in transparent plastics. Possible explanations are as follows: 1) thermal effects play the main part [1-3], 2) interaction of hypersonic waves produces the damage [4-6]. It is possible to determine the cause by varying the conditions.

Here we report results for polymethyl methacrylate (PMM) of grade PA, polystyrene (PS) of grade VPP, and polycarbonate (PC).

\$1. Methods. The materials were used as cylinders 15 mm in diameter and 60 mm long at various temperatures (-150 to 250° C), which were measured to  $\pm 4\%$  with chromel-alumel couples inserted from sides and end to depths of 10-15 mm. The center and surface were at the same temperature.

The pulses (6943 Å) were provided by a ruby rod 12 mm in diameter and 120 mm long and had lengths of 1 msec (no Q switch) and  $0.02 \ \mu$ sec (with Q switch), the respective energies being 1.0-1.5 J and about 0.5 J. The end face was perpendicular to the axis of the laser. The lens had a focal length of 40 mm and had its focal point at the center of the specimen. The specimens were sectioned and polished for examination.

\$2. Results and discussion. The working conditions varied substantially. Figure 1 shows the results with 1 msec pulses. PMM: a)  $-150^{\circ}$  C, b) 20° C, c) 80° C, d) 90° C, e) 160° C, f) 230° C; PS:'g) 100° C. Letter A denotes the focal point, while the arrow indicates the beam propagation direction. The two materials show similar modes of failure. Below the vitrification point (105° C for PMM, 100° C for PS [7]), the fracture is of brittle type, with specific disk cracks. Above the softening point (120° C for PMM and 116° C for PS), the failure failure is plastic, with small zones of burned material near the focus.

Let us consider the characteristic features of failure. As can be seen from Fig. 1 (failure in PS is similar), the greatest amount of failure is along the axis. This is probably related to the nonuniform energy distribution of the light beam. In a small region along the axis of the exposed area, failure is the result of a large number of small cracks (microcracks). The average dimension of these cracks is about 1 mm; in PS, their interiors are coated with soot (evidently the result of burnout).

The most important form of damage in PMM and PS below the vitrification temperature was the formation of large disk cracks at points along the axis, whose planes lay at about  $45^{\circ}$  to the axis. The mean size was 5 mm for PMM and 8 mm for PS. The size and number of these cracks increase with the temperature, the diameter in PMM at  $-150^{\circ}$  C being about 1 mm. PS at this temperature showed a reduction only in the number of large cracks, with little change in size. The maximum thickness of the cracks at these temperatures was about 0.001 mm. Cracks were formed ahead of the focal point, and they were most closely spaced near the focus, where the diameter was usually larger.

These cracks arise by brittle failure and have a complicated structure. Figure 2 shows the internal structure of a disk crack in PS, which shows a small dark central region A (burned material) with a diameter of about 1 mm. Then follows a mirror-smooth region B with a diameter of about 2 mm. These dimensions are roughly the same for all large disks in PMM and PS at all temperatures. Then follows region C, which consists of almost concentric smoothed ridges rising toward the edge. This entire region is covered by a thin layer of soot (more in PS than in PMM). Finally, there is an outer region of prefailure, which is not distinct in Fig. 2, with no traces of soot.

The region of axial microcracking becomes larger as the temperature increases, although the mode of failure tends to alter at high temperatures. At temperatures near the vitrification point, the microcracks become transparent bubbles about 0.5 mm in radius (appreciable plastic failure). Temperatures above the softening point produce no disk cracks, and the damage is restricted to the charred area, which is coated with a layer of soot and which is 1.5-2 mm in size, being near the focus. Around the charred areas (especially at high temperatures, Fig. 1, e and f) there are holes (bubbles), which usually have soot on the surfaces. This is probably due to expansion of char products. The largest dimension of the bubbles lie along the beam axis. If the temperature is very high, numerous transparent bubbles are formed in PS, which is related to marked change in the properties of the material.

Figure 3 shows the effects of  $0.02 \ \mu$ sec pulses in the same temperature as for the 1 msec ones: PMM: a) 0° C, b) 20° C, c) 230° C; PS: d)  $-150^{\circ}$  C, e) 60° C, f) 100° C. PMM gives the same mode of failure as with 1 msec pulses, except that there tend to be radial lines (Fig. 3, a and b). PS shows a different mode of failure, in that there are no disks below the vitrification temperature, the damage being localized in the convergent beam, with microcracks coated with soot. Figure 3e shows that there is a white halo around these cracks, the largest tending to become disks (mean size about 0.5 mm). There are also many silvery cracks. No microcracks occur above the softening point and the cone is uniformly gray. The failure in PMM and PS above the brittleness point is similar to that for the 1-msec pulses.

PC was used only at 20° C. A 1-msec pulse (Fig. 4a) did not produce disc cracks but microcracks coated with soot in the convergent cone. The failure at 0.02  $\mu$ sec was as at 1 msec (Fig. 4b), except that the microcracks were smaller. In both cases the effects were very much as for PS at 0.02  $\mu$ sec.

PMM, PS, and PC were also used at energies varying by factors of 2 or 4 in both senses. Only the size of the cracks was affected.

These results differ from ones previously reported [4], where disk cracks were ascribed to hypersonic waves, while thermal effects were considered negligible. Our results do not fit fully into that scheme [4], and it would appear that hypersonic damage does not predominate. Light pressure and electrical breakdown probably were unimportant, as in [4]. We must consider in more detail the thermal action of laser radiation in relation to the structure of amorphous polymers.

An amorphous polymer may be considered [7-11] as consisting of randomly tangled molecules. Polymers of regular structure or with large exchange forces tend to crystallize. An amorphous polymer can be considered [9] as having an ordered structure.

A feature found in all polymers is that the thermophysical properties vary from one region to another [10], the sizes of the regions ranging from a few hundred Å up to several microns. A polymer tends to have several relaxation times for melting, which are determined by the mobilities of the molecules and parts of the molecules [7].

In the above materials, PMM has the highest molecular mobility and PC the lowest. Further, materials formed in air usually contain trapped air, so internal combustion is possible.

The polymer is heated in the convergent beam, the energy density increasing towards the axis and towards the focal point. A further cause of nonuniform heating is the variation in properties, so we have microregions differing in temperature. The specific heat and the relaxation times are temperature-dependent [11], so some regions may become hot enough to evaporate or decompose (a hot plasma may be formed [1,2]), or may subsequently burn. Thermal conduction can also alter the temperature distribution. Finally, the pulse length must be considered in relation to the relaxation time for melting.\*

Consider the damage with 1-msec pulses. PMM and PS show damage of the same type. If we assume that the relaxation times are here less than the pulse length, the failure is of quasi-equilibrium type, and

<sup>\*</sup> Also, the absorption coefficient may be dependent on the beam intensity.









starts with more rapid heating and melting in certain microregions, whose size is then increased substantially by thermal conduction. Individual microregions may link up. The most likely effect is for similar



Fig. 4

regions to meet along the axis (the region of highest energy density). Evaporation of combustion produces a sharp pressure rise, and the specimen cracks in disk regions. These disks still contain gas at high pressure, which may produce transparent cracks around the disks.

High-pressure gas in the cracks was detected as follows. PMM and PS specimens were given 1-msec pulses at  $20^{\circ}$  C, and then they were heated to  $200^{\circ}$  C, which reduces the strength of the material. The disks grow considerably in thickness (from 0.001 to 0.05-0.1 mm) and the surfaces opposite the disks swell. The gas escaping from a crack has also been filmed [12]. We can thus assume that crack formation involves gas pressure.

If the relaxation time exceeds the pulse length, nonequilibrium processes occur; adjacent regions do not come to thermal equilibrium, and the transformations occur independently. PC probably has a relaxation time greater than 1 msec, so the damage is of a different type (Fig. 4a), with heating and damage in parts. The relaxation time is temperature-dependent, so melting and burning occur in the hotter parts. Cracking still occurs, but the cracks are much smaller and more numerous. The relation of pulse length to relaxation time is much the same for 0.02-µsec pulses, so the damage is the same.

The relaxation time in PMM is probably less than  $0.02 \ \mu sec$ , so the damage is as for 1-msec pulses. The relationship changes for PS, and the damage at  $0.02 \ \mu sec$  becomes comparable with that for PC. The variation in the damage with temperature is explained if the damage is thermal in origin, because the material becomes plastic and damage is related to burning in the region of highest energy density, with subsequent expansion of the combustion products. We are indebted to G. I. Barenblatt, Yu. P. Raizer, and E. Z. Fainberg for interest in the work and valuable discussions, and to V. N. Kotlerev for providing the PC specimens.

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