B. L. TSETLIN, N. G. ZAITSEVA, and Academician V. A. KARGIN

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B. L. TSETLIN, N. G. ZAITSEVA, and Academician V. A. KARGIN

ON TREE-LIKE CRACKS DEVELOPING IN PLEXIGLAS UNDER THE ACTION OF ELECTRON RADIATION*

In studying the transformations undergone, under the action of high-energy radiation, by specimens of polymethyl methacrylate (specimens of Plexiglas were tested—technical organic glass based on polymethyl methacrylate), we found that in a number of cases cracks arise and develop in these specimens, having an original tree-like form.

This phenomenon attracted our attention, since elucidating the regularities and causes of cracking of structural materials under the action of this kind of radiation may be of great importance from the standpoint of deciding the question of the suitability of these materials for service under conditions of exposure to radioactive radiations. In the course of further investigation, a number of features of the development of tree-like cracks were established, indicating that what is observed here is indeed a new phenomenon, unlike any of the known processes of crack formation in plastics.

This phenomenon is characterized by the following:

1. Tree-like cracks arise and grow only when there is a defect on the specimen —one artificially "introduced" or one that existed independently, associated with the technological history of the specimen (for example, with mechanical working).
2. The development of tree-like cracks in specimens of organic glass proceeds in time at a certain rate proportional to the dose rate. Beginning from a mechanical defect, the tree-like cracks gradually and uniformly cover the entire irradiated area of the specimen. The external appearance and the process of development of cracks in a Plexiglas specimen over time are shown in Fig. 1 *a*, *b*, and *c* (the mechanical defect was first introduced into the specimen with the aid of a center punch).
3. If several defects are present on the specimen, several "trees" develop simultaneously, the branches of which do not intersect or grow through one another. Between branches of different "trees" growing toward one another, a sharp boundary is formed (Fig. 1 *g*).
4. The cracks develop only in the irradiated part of the specimen. This is

Fig. 1

Figure 1: Fig. 1

shown in Fig. 1 *d* (the annular region of the specimen not affected by cracks was covered during exposure by a metal ring; in the course of their development the cracks went around this region as a kind of obstacle).

5. The appearance and development of tree-like cracks were observed only when Plexiglas was acted upon by fast electrons. Under the influence of X-ray radiation, cracks do not appear.
6. Tree-like cracks arise only in sufficiently thick specimens, whose thickness exceeds the thickness of the material penetrated

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* The work was carried out in 1950–1951; its results are set forth in Report No. 676 of the Institute of Physical Chemistry of the Academy of Sciences of the USSR (April 1953).

Fig. 1. Nature of the development of tree-like cracks in Plexiglas (dose rate $1.9 \cdot 10^{18}$ eV/cm^3·sec; the specimens were photographed in transmitted light):
a, b, c –propagation of cracks in a specimen with one "defect" (respectively, 10 sec, 1 min, and 15 min of irradiation);
d –system of cracks in a specimen with several "defects" (15 min of irradiation);
e –cracks in a specimen whose annular region was covered during exposure by a screen (5 min of irradiation).

electron beam. Under the operating regime of the accelerator installation used in our experiments, which served as the source of fast electrons (voltage 700 kV), the minimum thickness of the specimens sufficient for the development of cracks was 1.7-2 mm.

7. The nature and rate of the process of development of tree-like cracks are determined uniquely by the location of defects and by the dose rate, but do not depend on the internal stresses that may exist in the glassy polymer. Thus, when a plexiglass specimen that had been preliminarily annealed by many hours of heating at a temperature of 130°C was irradiated, there developed (of course, after a mechanical defect had been inflicted on it) the same network of tree-like cracks as upon irradiation of unannealed specimens. The same network of cracks also developed in a specimen that had first been strongly stretched at 130° and then rapidly cooled; any tendency toward "orientation" of the cracks in the direction of the internal stresses (the presence of such stresses was established by examining the specimen between crossed Polaroid films) was completely absent.

Fig. 2. Dependence of the propagation rate of tree-like cracks in plexiglass on temperature. Dose rate $1.9 \cdot 10^{18}$ eV/cm^3 · sec.

Fig. 2

Figure 2: Fig. 2

8. Tree-like cracks are internal cracks; they do not emerge onto the surface of the specimen. This was established as a result of direct microscopic examination of the specimens.
9. The cracks are "empty"; they do not constitute "channels" through which gases formed during irradiation of plexiglass escape into the surrounding atmosphere. This follows from the fact that, in the course of their growth, the cracks are readily stained, i.e., filled with a dye solution, in the case where the latter, during irradiation of the specimen, is in contact with the defect from which the cracks originate. This phenomenon was observed in experiments in which the irradiated specimen was placed in a flat cuvette (of depth somewhat less than the thickness of the specimen) filled with an aqueous solution of methyl violet (in this case the defects were made on the side surface of the specimen).
10. As the temperature at which the specimens are irradiated increases, the rate of crack growth decreases, as shown in Fig. 2. This indicates that relaxation phenomena play a definite role in the process of development of tree-like cracks.

It was further established that the process of development of tree-like cracks under the action of fast electrons is common to all organic glasses,* which emphasizes the interest of the phenomenon described here.

The experimental data obtained are clearly insufficient to draw any definite conclusions concerning the mechanism of the process under consideration. Nevertheless, the observations made do make it possible to draw some preliminary conclusions regarding the probable causes of this phenomenon.

It is evident that the cracks arise as a consequence of the appearance, in the irradiated material, of internal stresses that cause destruction of the specimen at its weakest points, namely at the mouths of microcracks formed when a mechanical defect is inflicted. The tree-like character of the developing cracks is connected with the fact that the stresses arising under the action of irradiation are uniformly distributed over the entire cross-

* Data on the development of cracks in various organic glasses will be described separately.

of the specimen perpendicular to the direction of the electron beam. This determines the equality, in a fully developed network of cracks, of the sums of the areas of the cracks in each element of the specimen area, regardless of the distance of this element from the defect. The nonuniformity in the growth of cracks in different parts of the specimen over time is determined by the fact that crack growth can begin only from the defective site. Developing in this way, starting from the defect, the crack then gradually spreads throughout the whole specimen and, in the course of its development, being itself nonuniform (“defective”), branches. Therefore, in those elementary regions where the absolute number of cracks is small, they are larger, whereas where there are many of them (as a result of branching), their transverse dimensions are smaller; this is what determines the dendritic form of the cracks.

To clarify the possible causes of the phenomenon under consideration, it is important to note that crack development is closely connected with the presence of a sharp boundary between the irradiated and unirradiated layers of the specimen; this follows directly from the above-described features of crack development. Evidently, at this boundary, partly because of the volume shrinkage of plexiglass as a result of its radiation-chemical destruction (it is known that in this process a large amount of gaseous products is liberated; see, for example, ([1], [2])), and partly because of the accumulation of excess electric charge, mechanical stresses arise. Low-molecular-weight products of the radiation-chemical destruction of the polymer, which form supersaturated solutions throughout the entire volume of the specimen, probably play a major role in the occurrence of these stresses. These low-molecular-weight products can be adsorbed at the mouths of microcracks present in the “defective” specimen. Molecules of such products, adsorbed near the boundary of the layer of material penetrated by fast electrons, may possess excess charges of the same sign (owing to the capture of electrons slowed down in the specimen). The electrostatic interaction of these charges probably causes further growth of the microcracks, as a result of which “fresh” surfaces for adsorption are again formed, and so on. As a result of such a process, proceeding continuously under the action of radiation, a branched system of cracks of dendritic form appears.

Institute of Physical Chemistry
Academy of Sciences of the USSR

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