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HIGUCHI, Masakazu

Research Institute for Applied Mechanics, Kyushu University: Professor

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ON THE COLOR OF THE FRACTURE SURFACE OF POLYMETHYL METHACRYLATE PLATE, COLORLESS AND TRANSPARENT IN ITSELF*

By Masakazu HIGUCHI**

The fracture surface of polymethyl methacrylate presents colors due possibly to the interference of lights occurring in the thin layer of molecular orientation, which may be produced when the fracture runs steadily across the specimen.

Essentially, according to the author's consideration, the actual surface of fracture can be regarded as a combination of the fractures of two ideal types, say, sweep type and polygenetic type, corresponding to the extremes of the fracture pattern. The colors of interference are characteristic of the fracture of sweep type, and their appearing is accordingly the more conspicuous on the surface of the fracture closer to this type.

We shall be able to utilize the colored pattern of the fracture surface as well as the markings of it for studying the fracture phenomena.

1. Introduction There are a number of studies of the markings on fracture surfaces of materials. They hold an interesting position between the practical investigation of the strength properties of engineering materials and the physical research of the strength on the submicroscopic scale. G. R. Irwin *et al.*,¹⁾ in one of such studies, showed interesting fracture patterns of plastics serving as a convenient model for the purpose of studying fracture markings in thick metal specimens. Using Plexiglass which is the material of the same family as Lucite of G. R. Irwin *et al.*, F. Zandman²⁾ gave a full microscopical investigation in "Étude de la déformation et de la rupture des matières plastiques." The parabola-like markings so clearly seen on the fracture surface of the material are justly explained by them as the boundaries of the level different domains of the surface.

So far as the author knows, however, no writer has made mention of the color appearing on the surface of fracture of the material, colorless and transparent in itself. That may be because the color usually appears only within the narrow limit near the fracture nucleus and when observed by the light of reflection. The color is not that of Newton Ring, which is observable in other part of the fracture surface as shown in Photo 14 (PL. II) and has its different origin as illustrated in Fig. 1.

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** Member of the Research Institute for Applied Mechanics, Professor of Kyushu University.

¹⁾ G. R. Irwin, J. A. Kies, A. M. Sullivan, Jour. Appl. Phys. 21, 7, 716 (1950).

²⁾ F. Zandman, Publ. Sci. et Tech. du Ministère de l'Air, No. 291 (1954).

However, by breaking notched specimens at ordinary temperatures, or at a moderately high temperature, we get the fracture surface to present bright colors with high chroma which occasionally reach the part of the later stage of breaking according to circumstances, although they are not perceptible at all when illuminated by transmitted light. It seems to the author rather unnatural that J. J. Benbow and F. C. Roesler³⁾ did not make mention of it in their paper, for the colors cannot be unrecognizable in the fracture surface obtained by such an experiment as theirs.

We may possibly add a measure for studying the mechanism of fracture of materials on a microscopic scale from the standpoint of applied chromatics, and that without the biologists' process of dyeing slides of specimens.

2. Molecular Orientation Layer It seems that the reason why some of fracture surfaces of polymethyl methacrylate present colors by reflection of light and others do not as described above is whether or not the fracture surface has a layer of regular molecular orientation of comparable depth with the wave-length of the visible light.

The fracture surfaces we get in the usual test are very rugged and look whitish on the whole as shown in Photos 1 and 2 (PL. I); and somewhere can be found the start of the fracture, in the vicinity of which the surface is comparatively flat. It is in the region that the colors are seen if any, which are however faint and appear within narrow limit in case the specimen has been broken under uniform tension as usual, and hardly attract attention.

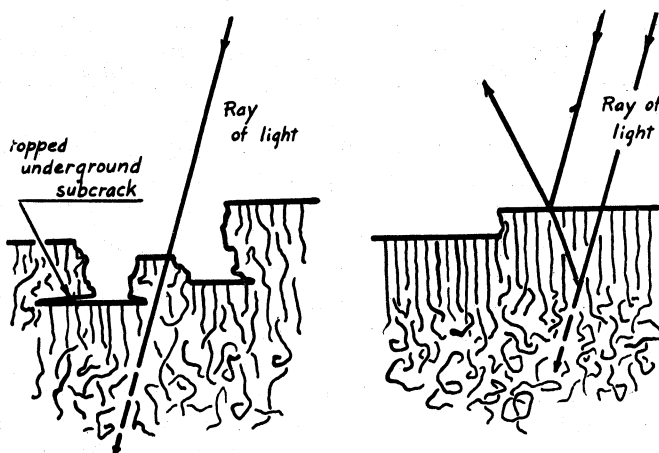


Fig. 1. Difference of the layers of molecular orientation in the cases of steadily propagating fracture (close to sweep type, right) and rather simultaneous fracture (close to polygenetic type, left).

In the polygenetic type the orientation of molecules may be irregular though extensive. Newton Ring as seen in Photo 14 (PL. II) is due to such a stopped underground crack as illustrated in the left figure, and must be kept distinct from the colors shown in PL. I.

³⁾ J. J. Benbow, F. C. Roesler, Proc. Phys. Soc., B, 70, 201 (1957).

When the tensile stress happens to concentrate particularly at some part or other beyond the extent of the occasional concentration owing to the failure of uniformity and to cause to start a fracture there, the specimen will suffer little stress in other parts by the time of fracture, even in the part of a little depth near that stress raiser nor in the layer which will become a fracture surface in the course of fracture. As illustrated schematically in the right side of Fig. 1, the surface layer of strong localized orientation of molecules will then be produced by the local stress acting just before the fracture, as against the orientation of fairly wide range and that with not a little irregularity in the case of uniform stress fracture.

The orderly layer of the surface, as has been known, has a more or less different refractive index of light from that of the matrix and the reflection of the ray of light must take place on the interface. The interference of the ray with the other ray of light reflected on the surface thus gives rise to the colors of interference.

Photos 1 to 6 (PL. I) show the contrast which is given by the difference of the sharpness of the notch which had been cut beforehand on one side of the specimen plate. Since the fracture stresses at the point of start may be considered, to a first approximation, nearly equal in any specimen, one can say that the specimens in the bottom side photos in PL. I must have broken under the lighter tensile load due to its higher concentration of stress at the bottom of notches than the specimens in the upper side photos have done. Naturally the roughnesses of the fracture surfaces relate to the breaking loads provided the tests are conducted with the same material and at the same condition of loading; it is qualitatively shown in Fig. 2. Accordingly one may say that the specimen of weak strength presents much the color if the material and the sectional area are alike.

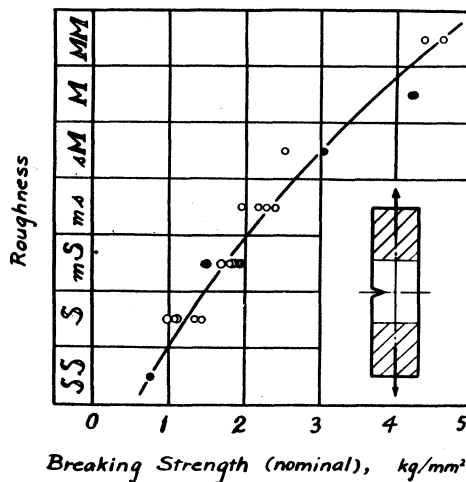


Fig. 2. Relation between the breaking stress (nominal value) and the roughness (qualitatively determined by comparison) of the fracture surface.

Specimen thicknesses are indicated by small and large circles for 3 mm and 5 mm respectively. Solid circles indicate the specimens tested at 42°C.

The monotonous relation obtained between the breaking loads and the roughnesses fails when the apparent sharpnesses of the notches are taken as the abscissas because of the indistinguishable irregularity of their sharpness; the magnitudes of breaking loads do wrong in no case.

The degrees of roughness of the fracture surfaces obtained are defined by dividing 24 specimens into three classes *M*, *ms* and *S* according to their apparent roughnesses and sorting out again intermediates into two classes *sM* and *mS*. Moreover, both extremes are added, which are the specimens from which a triangular fragment flied apart in the later stage of fracture (*MM*) and those which present especially bright colors with high chroma and have a particular smoother pattern (*SS*). This pattern has been obtained by increasing the test temperature to about 42°C, and is reproduced in Photos 7 and 8 (PL. I), placing side by side a pair of fracture surfaces which were originally opposite to each other.

3. Color Differences The characteristics of the color arrangement are twofold as recognizable in the Photos 7 and 8; the one is a parallel arrangement of the stripes colored greenish and reddish to the direction of the propagation of fracture, the other that in the transverse direction to it. These are also found in those shown in the Photos 3 through 6 when observed carefully.

There is a parallelism between the striped pattern of colors and the level differences of the surface of fracture. The reddish stripes were found by a interferometer raised several tenth microns with the specimens here shown. It will be however indicated later in a paper of the same author that the measure has not the definite connection with the difference of colors.

Nevertheless, the fact is beyond doubt that the reddish stripes are always prominent and the greenish are depressions; and the arrangement of the colors is exactly the reverse to that of the partner of the fracture surface in so far as the Photos 7 and 8 (PL. I) and the like patterns are concerned. This will prove the illustration given by F. Schwarzl and A. J. Staverman⁴⁾ after A. Smekal's explanation concerning the formation of level differences.

But such can not always be the case as will be known from the Photo 18 (PL. IV), in which all of the domains bounded by the parabola-like curves are found raised, and so with the opposite fracture surfaces. A modification of their explanation is needed for the pattern far from the ideal pattern of sweep type that the domain around the focus of a subcrack is raised owing to the possible elongation slower and hence larger than that of the zone near the boundary parabola-like curve, because the crack may propagate at the higher rate in the later stage of the subfracture.

The other characteristic of the color arrangement, that is the alternative appearing of two principal tones of color with the propagation of the main crack, shows possibly like the first the difference of the orientation layer depth measured with the optical path, although the partner of the surface also gives the identical pattern of color unlike the above-mentioned characteristic.

⁴⁾ F. Schwarzl, A. J. Staverman, Physik der Hochpolymeren (ed. H. A. Stuart) Vol. IV, p. 204.

The optical depth is related in this case to the velocity of fracture propagation at first sight. As a matter of course the depth is increased by the lower value of velocity as well as by the higher temperature. The alternations of the colors mean not only monotonically increasing or decreasing in the velocity but the hesitations of the propagation too. Nor we can say that the colors will be cyclically repeated many a time with the depth increase. Not only the brightness of the colors decreases with the depth but also the chroma does, that is, the colors fade into gray alike according to the study of H. Kubota⁵⁾ on the interference color of thin coating films.

For this reason the orientation layer of the fracture surface presenting colors is thought to be the visible light wave length deep ($400\sim700\text{ m}\mu$).

The colors fade out by ageing or otherwise by keeping the specimen several hours at a temperature of $40\sim50^\circ\text{C}$. Moreover the fading seems to take place together with the shift in the clockwise direction around the zone of white color on the CIE chart, which is attributable to the diffusion of the orientation.

4. Development of Fracture It will be interesting to study the markings on the fracture surface by color photographs. The curves shown in Photo 9 (PL. II) remind us of the stream lines about weak sources in a uniform flow of liquid. They can be said to have been developed by the steady running crack due to the concentration of the tensile stress acting only just prior to the crack formation. If the subcracks break out in advance of the arrival of the main fracture front, many of the curves widen sideways as seen in Photo 10, which is however not so much wide as those found on the surface shown by Photos 1 and 2 (PL. I); some of which are almost circular. Such pattern explains that there had been a nearly uniform distribution of stress before the fracture started.

Thus the interpretation seems to be appropriate that the difference of the patterns is not immediately the difference of the velocity of the fracture propagation, but rather that of the distribution of stresses.

Although there is a variety of their forms in the field of a microscope, the nuclei to start fracture may be classified into two or three types. Two of them are reproduced in Photos 11 and 12. But the types are not incompatible with one another, one or two of which appear sometimes on a small scale in another pattern of nucleus. Exactly speaking, therefore, the growth of the nucleus consists of three stages and their degrees of growth varies in each nucleus. Photo 11 shows the first one, wanting the second, the final part of which is regarded as the third stage where the main fracture starts, and Photo 12 is the second stage, comprising the first stage on a small scale with a greater or lesser modification. The third stage is also seen in the specimen to succeed to the hesitation front of the form of an arc.

There are seen a small number of radiants outside of the hesitation front, the detail of which is shown with another specimen in Photo 15 (PL. III). There is seen on the right a range of hills, a portion of the arc-shaped hesitation front. The eminence near the upper right becomes the start of a radial stripe; or otherwise it may be said that the parabola-shaped zone pokes its nose into the hesitation lunette.

⁵⁾ H. Kubota, Jour. Appl. Phys., Japan, 18, 4, 139 (1949). Ditto, 18, 8, 247 (1949).

The third stage develops sometimes following immediately to the first one according to conditions as seen in the Photo 7 of PL. I, where the growth of that phase happened to be arrested with a lunette region and the speed-up after a hesitation took place. It is possible for us to get the phase to develop overwhelmingly in a particular condition until the final of the fracture; and then the pattern on the whole becomes something that suggests the ark shell.

Photo 17 (PL. III) shows one of the typical patterns of the parabola-like curves in a magnification. Whereas, Photo 13 (PL. II) as well as Photo 16 (PL. III) show the patterns which are seen in the region between the nucleus to start the fracture and the stage of fracture in propagation of Photo 17. The left side portion of the pattern shown in Photo 13 presents the microscopic irregularity like that of the surfaces of the pasted papers torn off before the paste dries up, while that of Photo 17 appears fibrous within the parabola domain and innumerable tiny frizzes are seen run in the radial direction from the focus.

5. Conclusion Photo 19 in PL. IV, which is not the color photograph, shows the usual pattern of the fracture surface of polymethyl methacrylate plate. The velocity of crack propagation increases with the development of fracture as well known since H. Schardin's⁶⁾ measurements. At the same time, the stresses in the specimen naturally change their distribution with the propagation of the fracture.

When we control the fracture speed (here by making the sharp notch beforehand), we can get such patterns of the fracture surface as shown in Photo 20 and Photo 21, where each fracture must have propagated with its rather uniform speed with an identical distribution of stresses with respect to the momentary front of the fracture. The former type is akin to the fracture of sweep type, the typical model of which must be obtained when fractured under the stress distribution of the form of Dirac's delta function and must have no parabola-like level difference curves.

The latter type, on the other hand, may be called the fracture of polygenetic type in its extreme case. The ideal fracture of this type would have, if any, the infinite value of the velocity. The truth is however that no concept of the velocity of propagation can be defined from the macroscopic viewpoint.

The actual type of fracture is one of the combination of these two. The ratio of each component depends on the distribution of the stresses when the fracture starts, laying aside the influence of the temperature. Therefore N. F. Mott's⁷⁾ upper limit of the velocity of propagation of brittle fracture could seemingly be surpassed in the actual fracture which might include the prevailing component part of the polygenetic type.

The appearing of the colors of interference is characteristic of the fracture of sweep type and not recognized by its nature when illuminated by the transmitted light. The interference occurs in the surface layer of molecular orientation measuring $400 \sim 700 \text{ m}\mu$ in optical path. The colors fade out by ageing or by keeping the specimen several hours in a moderately high temperature; the fading is accompanied by the shift of the colors in the clockwise direction on the CIE chart, owing possibly

⁶⁾ H. Schardin, *Kunststoffe*, 44, 2, 48 (1954).

⁷⁾ N. F. Mott, Lecture at the Strength of Solids Symposium, Bristol University (1948); cf. G. Irwin's *Fracture Dynamics in Fracturing of Metals*, ASM (1948).

to the decrease in the degree of molecular orientation accompanying the increase in the layer depth due to the diffusion of the orientation.

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PL. I.

Fracture surfaces of Mitsubishi Acrylite (polymethyl methacrylate plate) specimens 3 mm thick and 40 mm wide at the notch bottom section.

The left ends of the specimens in the left photos are the bottoms of the parabola-shaped notches (cf. Fig. 2) of the varying bottom radii, therefrom the fractures, occurring at the respective nominal stresses, developed to the right.

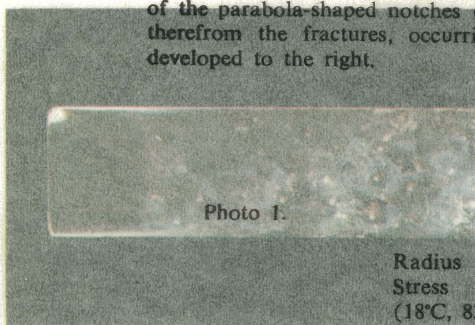


Photo 1.

Radius 3 mm
Stress 4.24 kg/mm²
(18°C, 83% R.H.)

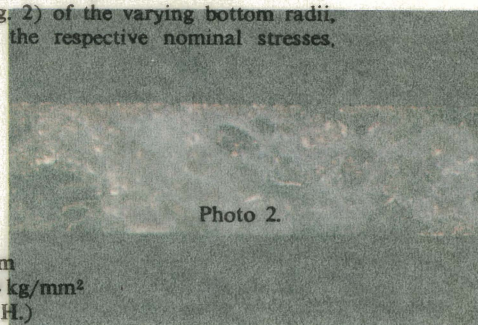


Photo 2.

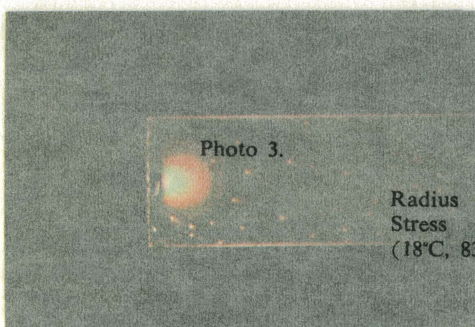


Photo 3.

Radius 0.5 mm
Stress 1.88 kg/mm²
(18°C, 83% R.H.)

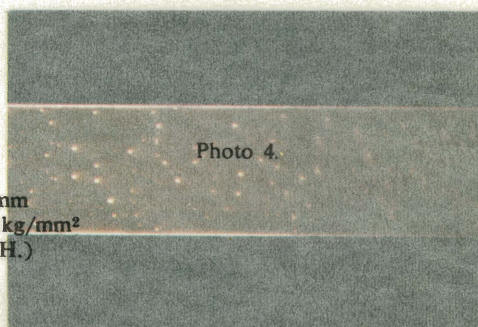


Photo 4.

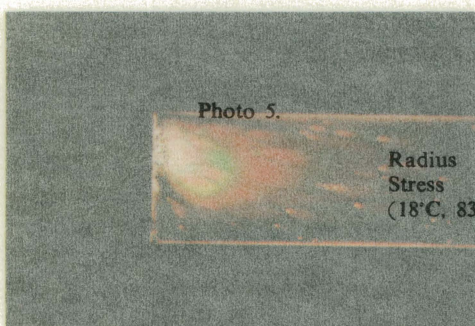


Photo 5.

Radius 0.2 mm
Stress 1.34 kg/mm²
(18°C, 83% R.H.)

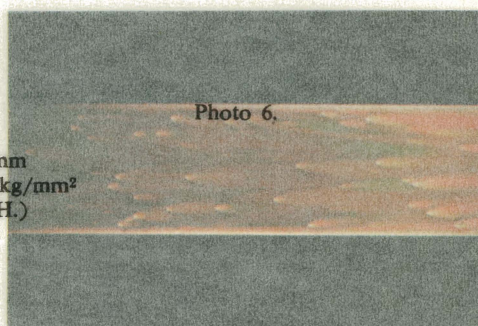


Photo 6.

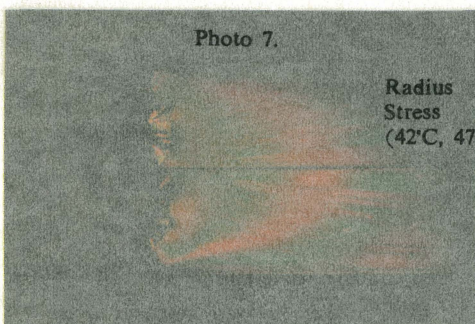


Photo 7.

Radius 0.2 mm
Stress 0.767 kg/mm²
(42°C, 47% R.H.)

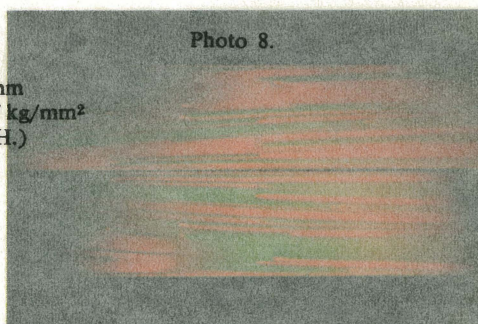


Photo 8.

PL. II.

Photos taken by the illumination of reflecting light.

Color temperature of the light source, about 3700°C.

Photo 9.

The same pattern of stream line markings as shown in Photos 7 and 8.

Fracture propagation, from the bottom to the top.

Reddish parts are slightly raised (a fraction of a micron).

Photo 10.

Parabola like markings
Fracture ran to the right.

Photo 11.

Fracture nucleus of the specimen in Photo 5.

The notch bottom is seen at the bottom side of the photo.

Photo 12.

Nucleus of the specimen in Photo 1, having an incubation region with a hesitation lunette.

Photo 13.

Transient zone from the slow rate of propagation to the higher rate.

Slow rate zone (left) reminds us of the pasted paper surface torn off before the paste dries up.

Photo 14.

The pattern observed somewhere on the fracture surface shown in Photo 2.

The colors are due to the underground stopped crack on the way of growth. Every white spot is a focus or the nucleus of a subcrack.

PL. III.

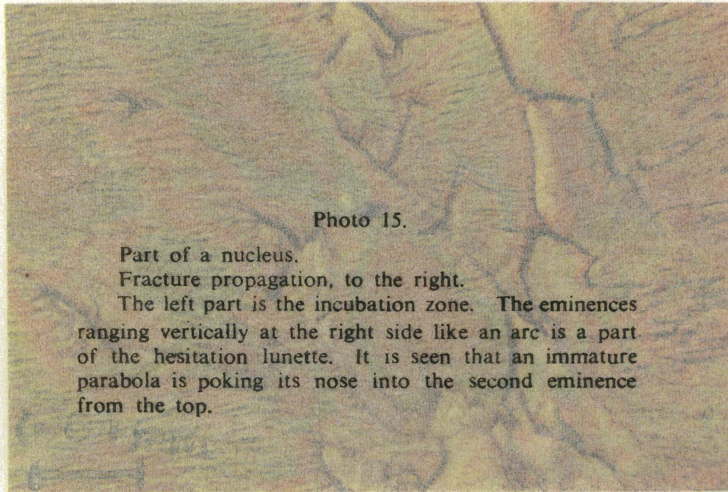


Photo 15.

Part of a nucleus.
Fracture propagation, to the right.
The left part is the incubation zone. The eminences
ranging vertically at the right side like an arc is a part
of the hesitation lunette. It is seen that an immature
parabola is poking its nose into the second eminence
from the top.

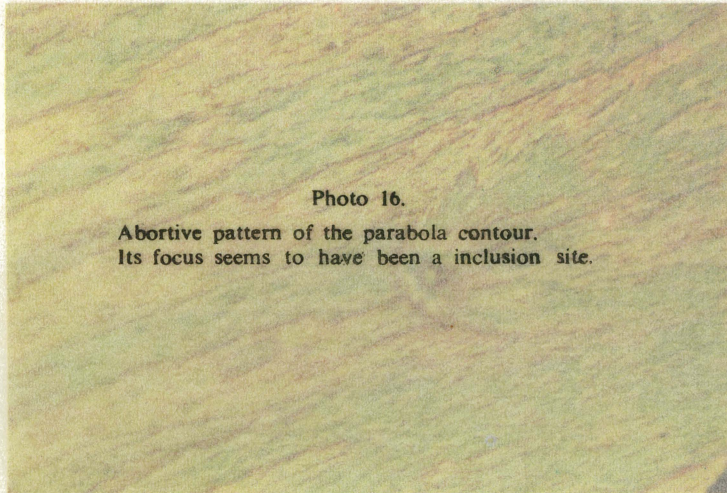


Photo 16.

Abortive pattern of the parabola contour.
Its focus seems to have been a inclusion site.



Photo 17.

Full-grown contour.

0.01 cm
1

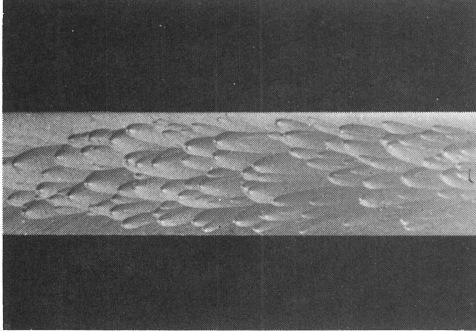


Photo 18.

Fracture surface illuminated obliquely.
Specimen thickness, 3 mm.
All the domains bounded by the parabola like curves are projected. So with the other surface opposite to this.

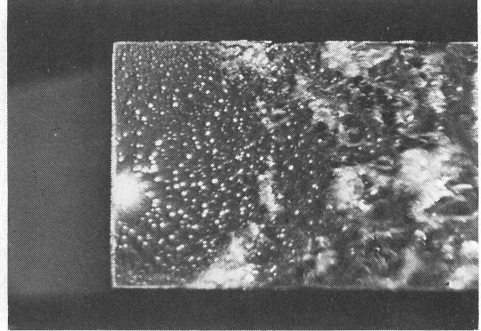


Photo 19.

Usual pattern of the fracture surface of a specimen (broken under tension without notch) of polymethyl methacrylate.
Specimen thickness, 5 mm.
Fracture propagation, to the right.

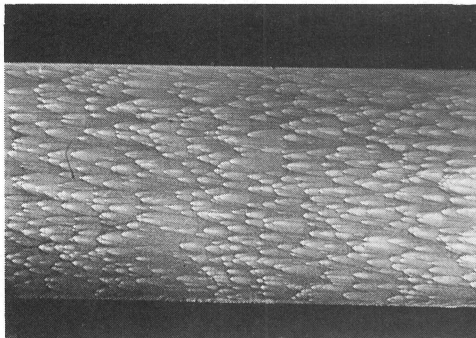


Photo 20.

Homogeneous pattern of a controlled fracture surface by means of a sharp notch, a type closer to the sweep type.
Parabola like curves are seen in multitude.

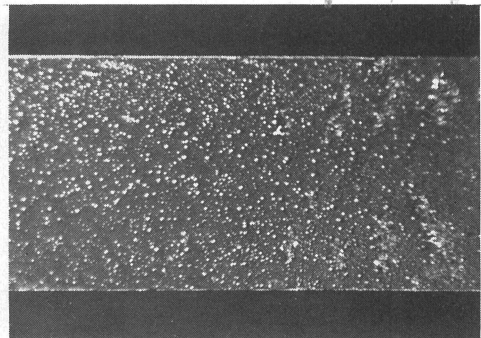


Photo 21.

Another homogeneous pattern of a controlled fracture surface by means of a dull notch, a type rather close to the polygenetic type.
Every white spot is a nucleus of a subcrack.