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A Microscopic Study of Fracture in Regenerated Cellulose Film*

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Synopsis

The fracture edge of a strip of regenerated cellulose film broken under tension can usually be divided into three regions: (A) the site of the initial failure, (B) the slow-tear region, and (C) the fast-tear region. Photomicrographs showing these regions are presented and discussed. Three different types of initial failure are described and some data presented relating the breaking strain to the type of initial failure.

Introduction

Many workers have demonstrated the usefulness of microscopy when applied to the study of fracture processes.[1,2] Most of the previous work has been restricted to materials in bulk form and there is little data relating to thin films of polymeric materials (about 0.001 in. thick). Although microscopic features due to the fracture process have been reported previously for regenerated cellulose and cellulose acetate fibers,[3-5] there appears to be no data available relating to regenerated cellulose film. This paper presents some preliminary results of an investigation designed to obtain a correlation between the fracture topography and ultimate tensile properties of cellulose film.

Experimental

Most of the data presented here were obtained for an experimental 300-gage machine cast film. The film was made using the viscose process and had 18 parts glycerol as the softener. Features identical to those which will be described have been observed on production made films so that the conclusions arrived at are not necessarily restricted to the film used for the present study.

The tensile samples (0.4 in. wide) were cut in the machine direction from adjacent areas of web. The breaking load and elongation were measured using an Instron tensile testing machine with a gage length of 5 in. and a crosshead speed of 10 in./min. The samples were conditioned for 24 hr. at 50 ± 2% r.h. and 20 ± 1°C., and were pulled under the same ambient conditions.

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The fracture samples were examined under a microscope using vertical illumination. It was not possible to examine the fracture surface because the film was too thin (nominal thickness 0.91×10^{-3} in.). In the present investigation, the fracture edge (that is, the surface of the sample in the vicinity of the fracture) was studied.

Selected samples were metal shadowed with antimony, using a zero replica technique similar to that described by Emerton and Page.[6] Shadow angles between 6 and 20° were used. The direction of shadowing was along the direction of the applied tensile load, i.e., the machine direction. The metallized samples were remounted on glass slides, using double-sided adhesive tape. The background structure, which can be seen in some of the photomicrographs, is due to this tape.

Experimental Results and Discussion

Figure 1 shows a photomicrograph of the two complementary fracture edges of a sample of 600-gage cellulose film (about 1 cm. wide) broken under tension. The fracture edge can usually be divided into three regions; the site of the initial failure (A), the slow-tear regions (BB) and (CC), and the fast-tear region (DD). The direction of the applied tensile load is indicated by the arrows labeled T.

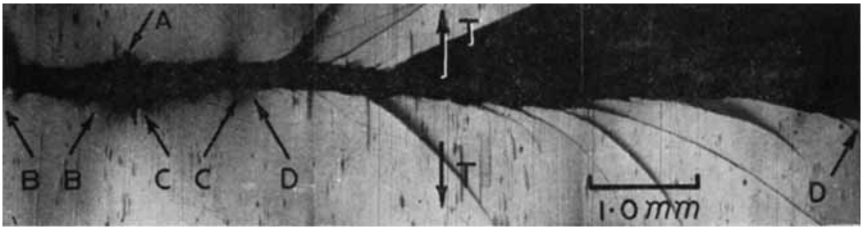

Fig. 1. Photomicrograph of the fracture edge in regenerated cellulose film: (A) site of initial failure; (BB and CC) slow-tear regions; (DD) fast-tear region. Direction of tensile load indicated by the arrow labeled T.

The fracture is initiated at some particular point (A) on the sample. The resulting crack is propagated outwards from this point. At first the speed of propagation is slow but increases rapidly as the crack length increases. These regions (B to B and C to C on the photomicrograph) are characterized by an irregular tear edge and surface cracking. At some instant fast tearing commences. In the fast-tear region (D to D) the fracture edge is relatively smooth, there is virtually no surface cracking and bifurcation of the crack occurs. This bifurcation is probably related to crack speed[7] and results in the loss of triangular-shaped pieces of material from the edges of the sample. Since this region is usually devoid of visible microscopic detail, it will not be discussed further.

The site of initial failure and the slow-tear region will be considered separately, and some data relating the type of initial failure to the ultimate tensile properties presented.

Initial Failure

Ignoring edge breaks, three distinct types of initial failure have been recognized so far. These are called (*1*) surface crazing, (*2*) internal cavitation, and (*3*) surface cavitation. They are shown in Figures 2, 3, and 4,

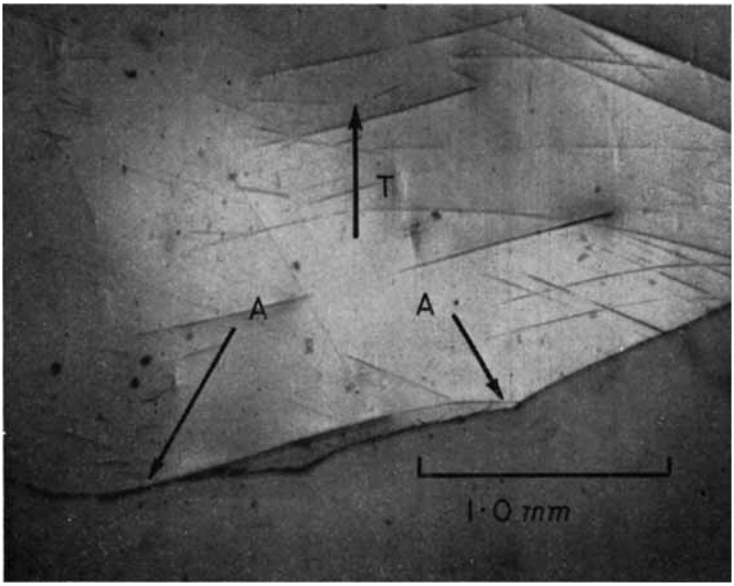

Fig. 2. Surface craze failure: (AA) region of initial failure.

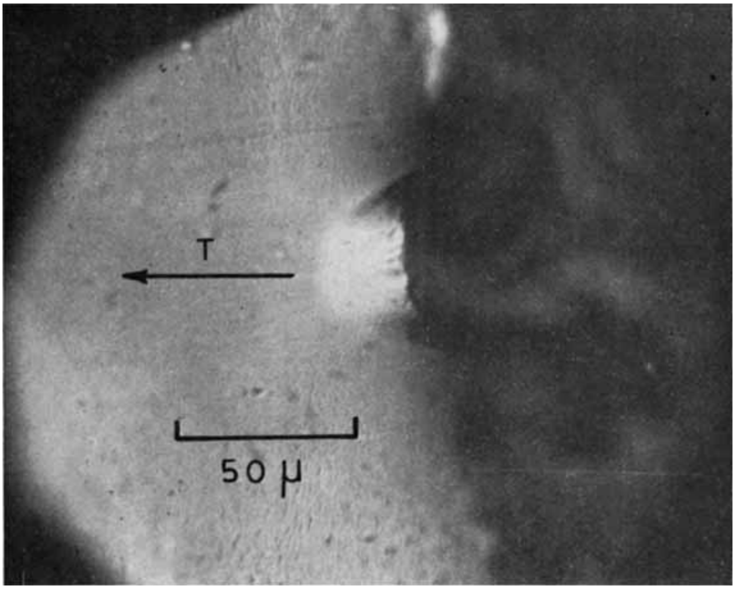

Fig. 3. Internal cavity failure.

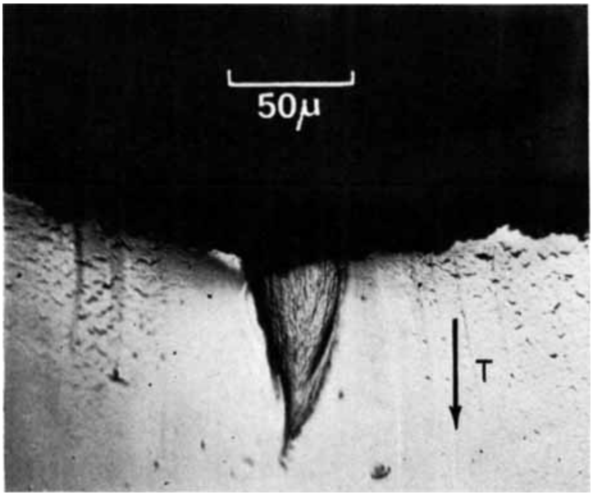

Fig. 4. Surface cavity failure.

respectively. The site of initial failure can usually be detected by studying the direction of tear propagation (as indicated by the secondary cracks in the fast-tear region) and the two slow-tear regions. The slow-tear regions are symmetrically placed about the site of initial failure.

These microscopic features should not be considered as the original cause of failure, but as manifestations of a particular process. That is, they are not the cause but the result of a particular type of initial failure.

Surface crazing is a form of localized yielding that occurs along a line in the surface of the film. It has a similar appearance to that described previously for other polymeric systems.[8] If fracture is the result of this type of process, initial failure occurs along a line, rather than at a point as in the other two cases. The initial failure appears to be due to the propagation of the craze through the thickness of the film until the reduced cross-sectional area cannot support the load. In Figure 2, initial failure seemed to occur in the region A to A.

Internal cavitation (Fig. 3) is the formation of a large, void-like feature within the bulk of the film. This is manifested by the dome-shaped feature on the surface. This type of failure does not appear to be associated with the film surface, as in the other two cases.

Surface cavitation (Fig. 4) is the formation of a crater-like feature in the surface of the film. Although similar in nature to surface cavities in rubber,[9] it has a different physical form. This type of initial failure proved to be the most interesting, not only because of its unusual form, but because it was amenable to intensive investigation.

Multiple cavitation is often observed in samples that fail by surface cavitation. This is the formation of several surface cavities with well-developed transverse cracks, as shown in Figure 5. The cavities usually lie in a line across the sample, the line terminating with the cavity that is

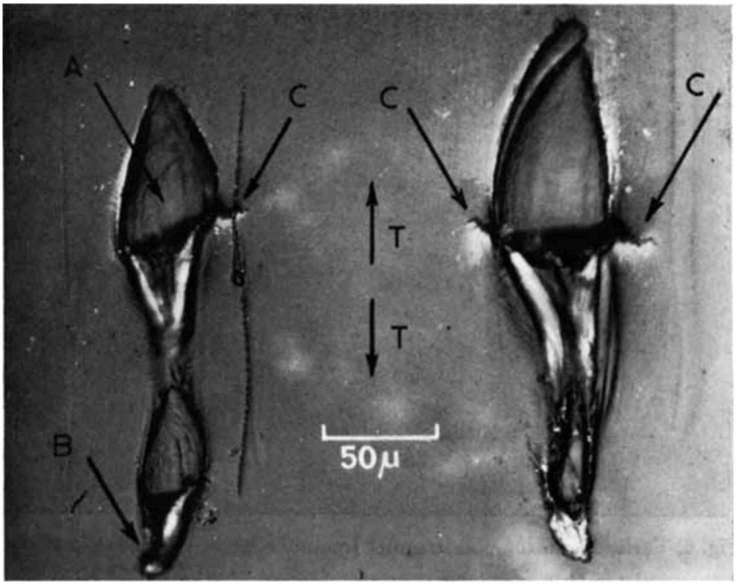

Fig. 5. Multiple-surface cavities: (A) cavity; (B) skin material; (C) transverse cracks.

the actual cause of failure. These cavities can be considered as sites of latent initial failure, so they can be used to study surface cavitation failure in its early stages.

Failure commences on or just underneath the surface of the film. The stress field causes a layer of material to be torn from the surface (refer to Fig. 5). This layer folds back onto itself and curls (B), leaving a crater (or cavity) in the surface (A). At some instant, transverse cracks (C) are formed across the cavity which, if allowed, would propagate across the sample. The cavity (A) is similar to that shown in Figure 4. When the two complementary fracture edges are compared the crater is found on one edge and the skin material on the other. With the other types of initial failure, the complementary fracture edges contain identical features.

Although it is not apparent from the photomicrograph (Fig. 5), visual observation showed that there were small surface cracks (described later) at the tips of the transverse cracks.

The Slow-Tear Region

Once a transverse crack has been formed by the initial failure process, the crack is propagated across the sample. The initial part of the crack growth (or tear) can be observed under the microscope, using samples with artificial flaws. At first the speed of propagation is very small (rather like a creep process) but increases rapidly as the tear length increases. This slow-tear process is characterized by an irregular fracture edge, as shown in Figure 6, and surface cracks which have a saw-tooth configuration. These can also be seen on each side of the surface cavity in Figure 4.

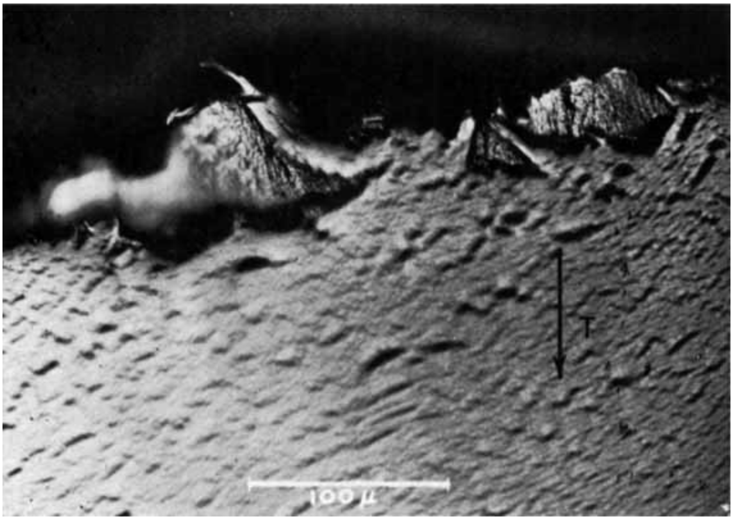

Fig. 6. Surface cracking and irregular fracture edge in the slow-tear region.

The tear propagates by the coalescing of the surface cracks which appear ahead of the tear tip. These surface cracks are formed on each surface of the sample and the tear propagates by the joining up of the two surface crack systems through the sample. This results in the irregular fracture edge shown in Figure 6.

Surface cracks similar in appearance to those observed in the present work have been reported previously in fractured cellulose acetate fibers.[4,5] They were attributed to the material in the surface of the fiber having a lower extensibility than that in the core. This would appear to be so in the present case. Pearse et al. have shown that for machine-cast films

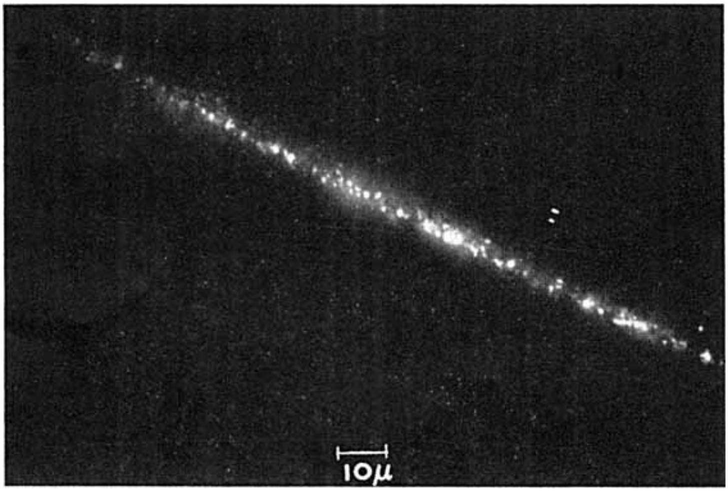

Fig. 7. Voids formed during the tearing process in the slow-tear region.

the machine direction[10] and planar[11] molecular orientation is much greater in the surface than it is in the core of the film. This increased molecular orientation would result in an increased stiffness but lower breaking elongation.

The formation of the tear, by the joining up of the two surface crack systems, is accompanied by the appearance of light-scattering centers (voids) within the bulk of the film. This is shown in Figure 7, which was obtained using dark-field illumination. Void formation in regenerated cellulose filaments by stress fields (before break) has been reported previously.[5]

The stress field ahead of the tear tip causes molecular orientation, as well as surface crack formation. This orientation can be detected by using transmitted polarized light, and is confined to the regions containing surface cracks. Void formation, molecular orientation and necking in the thickness direction, which can be observed on some photomicrographs, are all manifestations of the inelastic deformation that takes place during the slow-tear process.

Initial Failure and Elongation at Break

A histogram showing the frequency distribution of the breaking strain for different types of initial failure is shown in Figure 8. This was obtained for about 120 replicates of the 300-gage experimental film described previously. After eliminating jaw breaks, there were about 100 results suitable for analysis. In this diagram, *a* is the total distribution, *b* the distribution for craze failures, *c* for surface cavity failures, *d* for internal

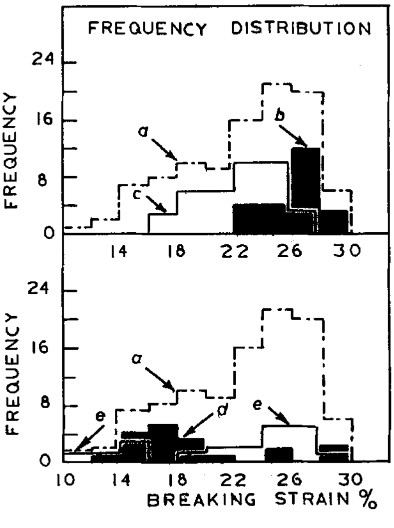

Fig. 8. A histogram showing the frequency distribution for breaking strain for an experimental cellulose film: (*a*) total distribution; (*b*) craze failures; (*c*) surface cavity failures; (*d*) internal cavity failures; (*e*), edge breaks.

cavity failures, and *e* for edge breaks. The failure was classified as an edge break if the site of the initial failure was close to the edge of the sample and such that two distinct slow growth regions could not be detected. Edge breaks can be due to stress raisers caused by imperfect cutting as well as inherent types of flaw, so that they do not necessarily reflect the true behavior of the material.

It can be seen that, for this film, the majority of the failures were due to surface cavitation and that the skew distribution was due mainly to the low strength internal cavity failures and edge breaks. Although there were insufficient data for a rigorous analysis, these results indicated that it may be possible to associate craze failure with the highest breaking strain. This implied that the maximum possible strain the sample could sustain was governed by surface craze formation. This conclusion was supported to a certain extent by the observation that, for breaking strains greater than 26%, about 50% of the samples which failed by a process other than surface crazing exhibited craze marks. For breaking strains smaller than 26%, less than 1% of such samples exhibited craze marks.

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Résumé

Le tranchant de cassure d'une languette de film de cellulose régénérée brisée sous tension peut généralement être divisé en trois régions distinctes: (A) le côté de la cassure initiale, (B) la région de déchirement lent et (C) la région de déchirement rapide. Des photomicrographies montrant ces régions sont présentées et discutées. Trois types différents de cassures initiales sont décrits et certains résultats présentés, reliant la tension à la cassure au type de brisure initiale.

Zusammenfassung

Die Bruchkante eines unter Spannung gebrochenen Filmstreifens aus regenerierter Cellulose kann gewöhnlich in drei Bereiche unterteilt werden: (A) den Ort des Ausgangsbruchs, (B) den Bereich des langsamen Reissens und (C) den Bereich des schnellen Reissens. Mikrophotographien dieser Bereiche werden gezeigt und diskutiert. Drei verschiedene Ausgangsbruchtypen werden beschrieben und einige Daten bezüglich des Zusammenhanges zwischen Bruchspannung und dem Typ des Ausgangsbruches vorgelegt.

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