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Reflection of Soft X-Rays by Organic Fibers

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Synopsis

An artifact observed in point projection microradiographs of polymeric fiber and filaments obtained with 8-A. x-rays is recorded and described. The phenomenon has been related to fiber-beam geometry and the high reflection efficiency of soft x-rays for these materials.

Introduction

The development of microfocus tubes of the Cosslett-Nixon type[1] produced a renewed interest in applications of microradiography to technical problems. Poen,[2] Isings et al.,[3] and Newman[4,5] have all demonstrated the usefulness of the instrument in studies of fibrous or fiber-containing materials. In general, their work dealt with interfiber relationships, the distribution of discrete phases in a fibrous matrix, or changes in whole fibers. Increased availability of microradiographic equipment with relatively high resolution has permitted observations on individual commercial fibers, as well as other forms of both inorganic and organic materials. In this work the analysis of artifacts produced by the interaction of the radiation and the specimens is an important element. Polymers differ considerably in x-ray absorption and other characteristics from many materials commonly involved in microradiographic investigations.

Equipment

A two-lens, electromagnetic microfocus tube, essentially of the design described by Cosslett et al.[6] and manufactured by Microxray Laboratories, Guildford, Surrey, England, was used in this work. Almost all of the data were obtained with 3-μ aluminum targets and the tube operating with an accelerating voltage of 15 kv. A few microradiographs were obtained with radiation from 5-μ copper targets at the same accelerating voltage.

Observations and Discussion

When polymeric fibers in a fairly random orientation to a divergent beam of soft x-rays are recorded on a plate, some of the fibers in the resulting microradiograph reveal a concentric structure of high contrast. The results obtained with a specimen of knitted acrylic fabric are shown in Figure

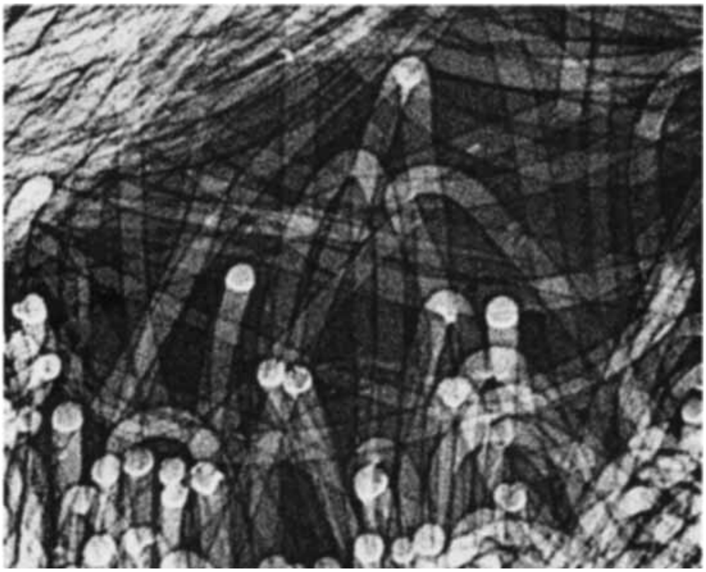

Fig. 1. Microradiograph of commercial acrylic fibers (15 μ diameter). Aluminum target, 15 kv.

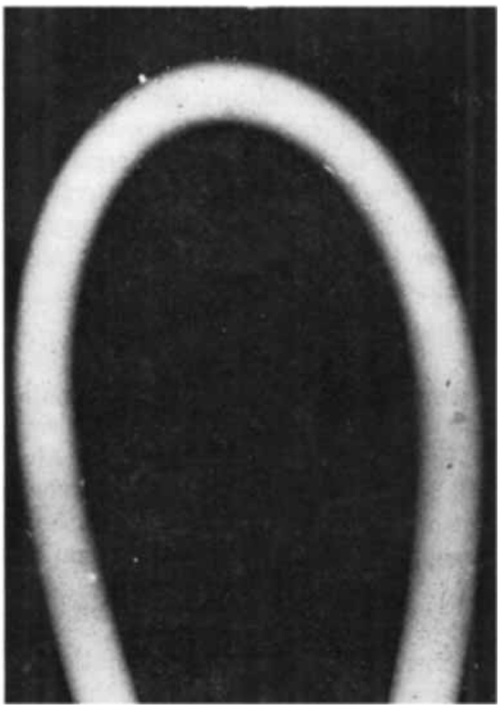

Fig. 2. Microradiograph of a bent nylon monofilament (0.38 mm.). Aluminum target, 15 kv. The long axis of the filament is normal to the x-ray beam axis.

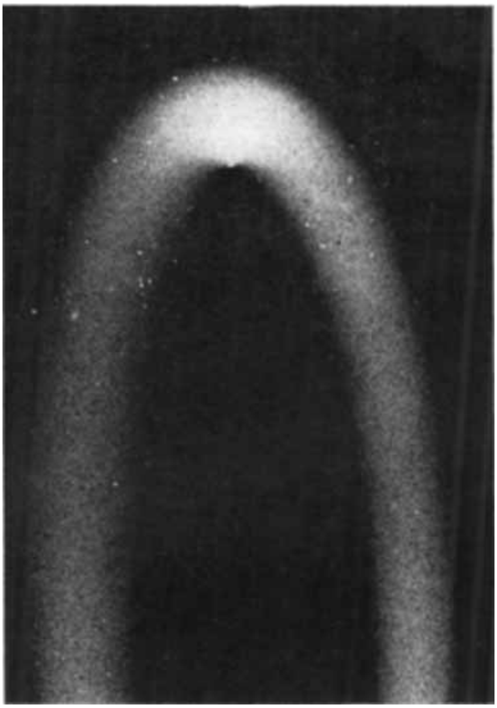

Fig. 3. Microradiograph of a bent nylon monofilament (0.38 mm.). Aluminum target, 15 kv. The long axis of the filament in Fig. 2 has been rotated slightly so that the arm which appears larger in diameter is closer to the target than the other.

1. This print and all others used in this report were made from a reversal of the original plate and therefore indicate the relative blackening of the recording surface by the x-rays. At first glance the figure appears to contain a number of fiber cross-sections with a light skin and a darker core. However, closer scrutiny reveals that these are not the ends of fibers, but portions in which the fiber is acutely bent with the long axis of the bent section nearly parallel to the x-ray beam axis. The peripheral element is never differentiated completely around the fiber but is limited to arcs of about 180° or less. This phenomenon is not restricted to a single type of polymeric fiber; it has been observed in nylon, viscose rayon, and several acrylic fibers. However, it is most contrasting and best resolved in acrylic fibers. The irregular contour of viscose does not prevent its appearance. Glass fibers give some indication of a similar structure, but it is poorly resolved and lacking in contrast.

A number of samples of acrylic fiber were observed by microradiography. These differed only in their delustering and dyeing processing. One was bright, one delustered with titanium dioxide (0.5%), a third was bright but stained with a chromium-free brown dye, and the fourth was also bright but stained with a brown dye containing chromium. These all gave the results shown in Figure 1. Similarly, the varying of the skin–core ratio in a series of viscose rayon fibers produced little change in the development of the same features in microradiographs of these fibers. It would appear that these variables can be eliminated as the basis for this phenomenon.

Filaments of the commercial fibers were too fine and flexible to orient accurately in the beam so a relatively thick nylon monofilament was substituted. These monofilaments had a diameter of 0.380 mm. Figures 2–5 are microradiographs of one bent monofilament as it was rotated in the x-ray beam. In Figure 2, the filament was oriented with the long axis in

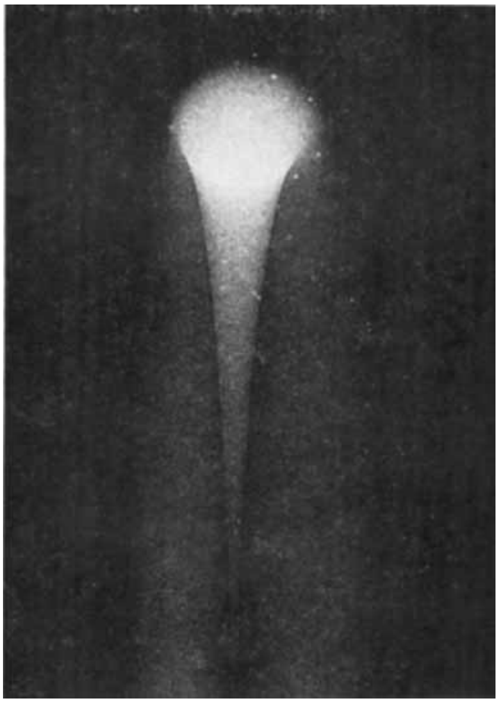

Fig. 4. Microradiograph of a bent nylon monofilament (0.38 mm.). Aluminum target, 15 kv. The long axis of the filament in Fig. 3 has been rotated farther and the two arms now overlap (light area).

a plane parallel to the target. The subsequent illustrations show the filament being rotated through 90 degrees until in Figure 5 the filament axis is in a plane almost normal to the target plane. The characteristic light-bordered cross-section again appears.

This series of microradiographs indicates the dependence of the appearance of the light border on the orientation of the specimen in the beam. Only when the curved surface of the bent filament is almost parallel to the beam axis does it become visible. Total reflection of x-rays from solid surfaces takes place at very small angles, and parts of the surface of the concave inner portion of the bend could provide the necessary geometry. That such a phenomenon might arise from total reflection effects is sug-

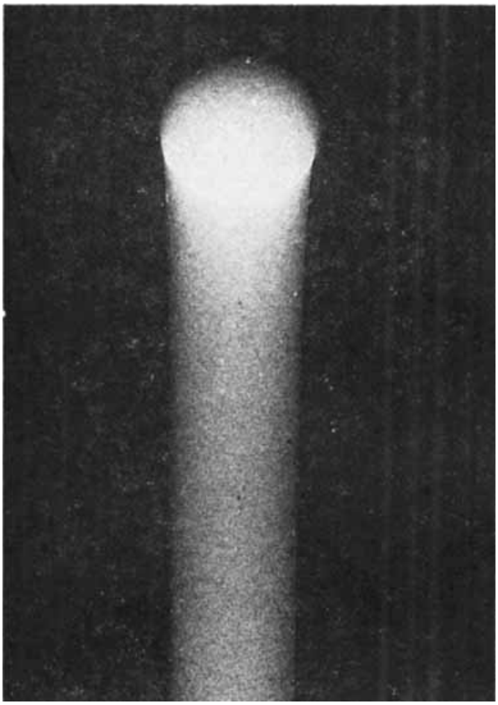

Fig. 5. Microradiograph of a bent nylon monofilament (0.38 mm.). Aluminum target, 15 kv. The filament in Fig. 4 has now been rotated through 90° and the arms are almost completely overlapping.

gested by the orientation dependence since it is at the normal fiber orientation that the largest coherent surface is available.

To see how this may arise, consider the conventional Fresnel description of total reflection.[7] The theory shows the dependence of refractive index n on wavelength and the physical constants of the material. It is convenient to introduce a quantity δ, the unit decrement of refractive index

$$\delta = 1 - n$$

The classical expression for δ at wavelength $\lambda \ll \lambda_k$ is:

$$\delta = (Ne^2/2\pi\ mC^2)Z\lambda^2$$
$$= 2.70 \times 10^{10} Z\rho\lambda^2/A$$

where ρ is the density, Z the atomic number, A the atomic weight, e the charge, N the member of atoms per cubic centimeter and m the mass of the electron. The calculated values for δ agree will with experimental results.[8,9]

Values of δ and the critical angle ϕ_c for a number of low Z materials are given in Table I.

The values for ϕ_c for organic materials are all about 1° or less. Absorp-

TABLE I
Critical Angle ϕ_c and Unit Decrement of Refractive Index δ for $\lambda = 8.3$ A.

Material	Z	ϕ_c, milliradians $\times 10^2$	$\delta \times 10^4$
Aluminum	13	2.20	2.41
Acrylonitrile	4.0[a]	1.54	1.16
Cellulose	4.1[a]	1.74	1.50
Nylon	3.3[a]	1.54	1.16

[a] Weighted calculation.

tion of the incident rays in the reflecting medium is neglected. Mohr[10] has measured the reflection efficiencies of surfaces of glass, steel and gold at angles inside the critical angle. Henke and Du Mond[11] have noted that these data would indicate that the reflection efficiency is approximately proportional to the reciprocal of the reflector density. Extrapolation of this relationship to the fibers used in these observations gives reflection efficiencies about twice that of glass for viscose rayon and three times for polyacrylics and nylon. This simple relationship may be misleading if other factors are ignored. For example, in later work, Henke[12] has noted that the total reflection cut-off is quite sensitive to the chemistry and structure of the reflecting surface when the incident radiation has a wavelength near a critical absorption edge wavelength of a major element constituting the surface.

However, it seems reasonable to assume that the "skin-core" phenomenon is a result of total reflection from the areas of the fiber surfaces making the requisite small angles with the x-ray beam. Since ϕ_c is approximately 1° it should be possible to detect changes in the width of the halo if total reflection is responsible for the effect. A series of four microradiographs were prepared by increasing the plate-to-specimen distance, the maximum increase being about sevenfold. The target-to-specimen distance was held constant, and the specimen was not moved while the whole series were recorded. Instrumental stability was insufficient for the required exposures at greater distances. The precision of the measurements was limited by the small width of the halo on the plates, never exceeding hundredths of a millimeter, and the rather diffuse edges. Despite these limitations, it was obvious that the width was increasing approximately as indicated by calculation.

These observations indicate the origin of one artifact that will presumably be commonly encountered in high resolution microradiography of textiles. They also demonstrate the high reflectivity of polymeric materials which may be useful in developing elements for efficiently controlling x-radiation.

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References

1. V. E. Cosslett and W. C. Nixon, *Nature*, **168,** 24 (1951).
2. O. S. Poen, *Microprojection with X-Rays*, Martinus Nijhoff, The Hague, 1959.
3. J. Isings, O. S. Poen, J. B. LePoole, and G. van Nederveen, *Proc. Intern. Conf. Electron Microscopy, Stockholm, 1956*, 282 (1957).
4. S. B. Newman, *Mod. Plastics*, **40** [7], 165 (1963).
5. S. B. Newman, paper presented at Symposium on Resinography, 1963; *ASTM Spec. Tech. Publ.*, No. **348,** 139 (1963).
6. V. E. Cosslett, W. C. Nixon, and H. E. Pearson, *X-Ray Microscopy and Microradiography*, Academic Press, New York, 1957.
7. R. W. James, *Optical Principles of the Diffraction of X-Rays*, G. Bell, London, 1958, p. 54.
8. E. Nähring, *Phys. Z.*, **31,** 401 (1930).
9. J. A. Bearden, *Phys. Rev.*, **54,** 698 (1938).
10. C. B. O. Mohr, *Proc. Roy. Soc. (London)*, **A133,** 292 (1931).
11. B. L. Henke and J. W. M. Du Mond, *J. Appl. Phys.*, **26,** 903 (1955).
12. B. L. Henke, *Advan. X-Ray Anal.*, **5,** 285 (1962).

Résumé

Un phénomène observé dans les microradiographies par projection de points de fibres polymériques et de filaments obtenus avec des rayons-x à 8 Å est rapporté et décrit. Ce phénomène était relié à la géométrie des fibres et du faiseau lumineux, et à l'efficacité de réflexion élevée des rayons-X mous.

Zusammenfassung

Über ein Artefakt, das bei mit 8-Å-Röntgenstrahlen erhaltenen Punktprojektionsmikroradiographien von Polymerfasern und fäden beobachtet wurde, wird berichtet. Das Phänomen wurde zur Faser-Strahlgeometrie und zur hohen Reflexionsfähigkeit dieser Stoffe für weiche Röntgenstrahlen in Beziehung gesetzt.

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