Square Bubbles in Irradiated and Annealed Lithium Fluoride Crystals

When lithium or a compound containing this element is irradiated with neutrons, the $_3Li^6$ isotope, which constitutes about 7.5 percent of natural lithium (*1*), undergoes the (n, α) reaction (*2*)

$$_3Li^6 + _0n^1 = _2He^4 + _1H^3$$

If the irradiations are carried out on solids at low temperatures, the resulting helium and tritium gases are trapped within the crystals. As an extension of a program at our laboratory on the behavior of fission gases and other gaseous nuclear reaction products (*3*), a study has been made of the behavior of irradiated LiF single crystals on thermal treatment. As would be expected, at higher temperatures, the gases agglomerate to produce voids in the solid.

The shape of these cavities is, however, quite anomalous. Instead of spherical holes, which might have been anticipated, under certain thermal treatments the cavities are nearly *square*, approximately 0.01 to 0.02 millimeters on a side, and quite thin compared with their plane dimensions. When these cavities first appear, they are characterized by brilliant interference colors, indicating that their thickness is of the order of a few wavelengths of visible light, probably no more than 0.002 mm at most. No voids, spherical or plane, were observed in unirradiated LiF even when it was annealed for as long as 72 hours at 700°C or at temperatures near the melting point of the salt, 842°C (*4*).

To observe the square holes, LiF crystals obtained from the Harshaw Chemical Co. were irradiated in the reactor at Brookhaven National Laboratory at 30°C for integrated thermal neutron exposures of 1×10^{17} and 3×10^{18} neutrons/cm². The crystals were then heated in air. There was no difference between the two samples, except for the presence of more gas and more cavities in the specimens with the longer irradiation, as might be expected (*5*).

At 500°C, no agglomeration of gas or appearance of voids could be detected even after 72 hours of treatment and with magnifications of as high as 1000; at 660°C, in 15 hours, voids were readily observable at magnification of 150; and at 726°C, 3 hours was sufficient to produce voids that were easily visible under the same magnification (*6*). Figure 1 is a photomicrograph (*7*) of a crystal subjected to this treatment, showing the square bubbles. The white areas are faces of the cavities inside the transparent crystal; cavities at the surface appear dark. The edges of these squares tend to be parallel to the (100) planes of the crystal and appear predominantly at subboundaries rather than in sound portions of the subgrains or in regions adjacent to cracks. (Note crack in Fig. 1; possibly the failure to observe voids near cracks is the result of the fact that the latter constitute convenient paths for the escape of reaction product gases.)

Once the voids have formed, prolonged isothermal treatment of the crystals produces no significant change in the relative positions, but it does result in an increase in the population density and growth of the edges of the voids. This phenomenon was examined at 726°C after 3, 9, and 24 hours, and it was found that the edge-length growth is apparently linear in time and is in fact described by the equation, determined by the method of least squares,

$$L = (2.64 \pm 0.11) + 0.141t \qquad (1)$$

for t less than 25 hours, where L is the void edge length in microns, t is the time in hours, and ± 0.11 is the standard deviation of the observed from the calculated results. (This was for a sample irradiated to 3×10^{18} neutrons/cm² at 30°C. The interference colors observed when the cavities first develop disappear as this growth process progresses, indicating that the holes are increasing in thickness simultaneously with the increase in plane area.

With a constant annealing time, at increasing temperatures, precisely the same kind of growth is observed—that is, increase in the length of void edges and thickness, and no change in the relative positions of the square holes. This behavior was examined at four temperatures in the range 700° to 820°C, and the data, for the same irradiation time and temperature as in Eq. 1, and for a uniform annealing time of 15 hours, are described by the least squares equation

$$L = (-34.00 \pm 0.51) + 0.0582T \qquad (2)$$

for $700° < T < 825°C$. In this relation, T is the annealing temperature in degrees Centigrade, L is the edge length in microns, and ± 0.51 is the standard deviation. From Eq. 2, L is 0 at $T = 584° \pm 88°C$—that is, this is the minimum temperature at which bubbles would become visible after 15 hours of annealing. At higher temperatures, the cavities lose their square shape. Thus, after 15 hours, at 820°C the holes were observed to have rounded corners; and annealing for this time just below the melting point (842°C) resulted in the more intelligible formation of cavities with circular cross sections, presumably spheres. At 726°C, rounding of the corners of the squares was also observed, after 82 hours of treatment.

The mechanism by which these thin square cavities form and the reason for their observed alignment with the (100) crystallographic planes are not understood. This phenomenon has been observed to date only with irradiated LiF, and it is not known whether any other crystals exhibit this rather surprising behavior (*8*).

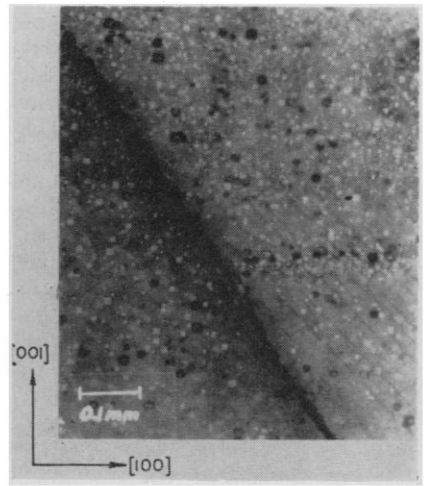

Fig. 1. Square bubbles in lithium fluoride.

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References and Notes

1. I. Kaplan, *Nuclear Physics* (Addison-Wesley, Cambridge, Mass., 1955), p. 172.
2. R. E. Lapp and H. L. Andrews, *Nuclear Radiation Physics* (Prentice-Hall, New York, ed. 2, 1954), p. 365.
3. This work was supported by the U.S. Atomic Energy Commission under contract Number W-31-109 Eng-52 and was done at the Knolls Atomic Power Laboratory, which is operated for the commission by the General Electric Company.
4. *American Institute of Physics Handbook* (McGraw-Hill, New York, 1957), section 5e, pp. 5–186.
5. Crystals exposed to approximately 2×10^{19} neutrons/cm² become extremely turbid on thermal treatment (presumably because the density of the gas aggregates) and cannot readily be examined internally by microscopic techniques.
6. Samples annealed in a vacuum at about 5×10^{-6} mm-Hg showed no significant difference in the population and size of voids from those treated in air.
7. Examination of the crystals was made with a Bausch and Lomb metallograph, and photomicrographs were taken at magnifications of 150 and 250, employing standard photographic attachments.
8. The assistance of C. W. Tucker, Jr., and Leo F. Epstein is gratefully acknowledged.
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Experimental Infection of Rats with Streptobacillus moniliformis

Although *Streptobacillus moniliformis* has been isolated frequently from wild and laboratory rats, particularly from the lung, middle ear, and nasopharynx, it has been considered a commensal microorganism of low virulence for this host (*1*, *2*). The microorganism has displayed virulence for human beings, most commonly following the bite of a rat, but