Square Bubbles in Irradiated and Annealed Lithium Fluoride Crystals

When lithium or a compound containing this element is irradiated with neutrons, the ${}_3\mathrm{Li}^6$ isotope, which constitutes about 7.5 percent of natural lithium (*1*), undergoes the (n, α) reaction (*2*)

$$ {}_3\mathrm{Li}^6 + {}_0n^1 = {}_2\mathrm{He}^4 + {}_1\mathrm{H}^3 $$

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/cm^2. 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/cm^2 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*).

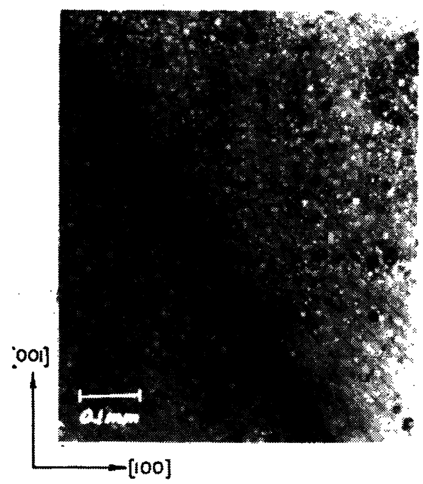

Fig. 1. Square bubbles in lithium fluoride.

PETER SENIO*
Knolls Atomic Power Laboratory,
General Electric Company,
Schenectady, New York

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/cm^2 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.
* Present address: Westinghouse Electric Corporation, Bettis Atomic Power Division, Pittsburgh, Pa.

29 April 1957

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 also after ingestion, and the infection has been characterized by fever, rash, and arthritis (*3–5*). High pathogenicity for mice has been demonstrated, and one means of identification of *Streptobacillus* is by the infection produced in mice, with resultant septicemia and joint involvement (*2, 4, 6*). The joint infection in mice is essentially an osteoarthritis and osteomyelitis, usually secondary to, or accompanying, a severe generalized systemic infection. Joint involvement accompanying systemic infections of man and animals by other microorganisms is also seen, although joint involvement alone may occur. An affinity of *S. moniliformis* for joint tissues of the developing chick embryo has been described (*7*). Attempts to infect rats with *S. moniliformis* via several routes have been unsuccessful, except for the occasional production of a localized lesion at the injection site (*2, 6, 8*).

During immunological studies with *Streptobacillus moniliformis*, a number of strains were isolated from naturally occurring lung and middle-ear infections in older laboratory rats. One recently isolated strain from a middle-ear infection (strain "C") has proved to be virulent for the laboratory rat and appears to have a specific predilection for joints, bone, and periarticular tissues. Upon intravenous injection with this strain, young rats developed a polyarthritis within 5 to 7 days, which subsided 10 to 30 days after the acute phase in most animals. The disease was produced consistently, in several experiments, in a high percentage of animals injected intravenously, but it was not produced by subcutaneous injection with the same strain.

Young Holtzmann-Fisher cross or Sprague-Dawley rats, 2½ to 3½ months old, were used. The Holtzmann-Fisher animals were offspring of rats which had been maintained on prophylactic doses of aureomycin for months, and neither parents nor offspring showed cultural or immunological evidence of natural infection with *Streptobacillus*. Cultures of *S. moniliformis* were carried routinely on rabbit blood agar, and in beef infusion broth adjusted to *p*H 7.6 to 7.8 and enriched with 20 to 30 percent sterile human ascitic fluid. These media were selected for producing the most consistent and abundant growth of the strains under investigation, after several other culture media and enrichment materials had been screened. Injections were made into the tail vein with 2.0 ml of an 18- to 20-hour beef infusion-ascitic fluid broth culture which had been inoculated from a 24-hour broth or blood agar stock culture.

Within 5 to 7 days after injection, rats developed redness, swelling, and tenderness of the affected joints. The degree varied from moderate to severe, and one to several joints showed involvement. Radio-carpal regions (wrists), tibio-tarsal regions (ankles), digits, and occasionally hips were affected, and some animals showed involvement of all four extremities at one time. Occasionally, the arthritis was migratory, disappearing from one joint only to appear in another. The incidence of clinically observed lesions in rats in five separate experiments is shown in Table 1. A total of 40 animals were injected intravenously, of which 37 animals developed gross joint changes. The clinically obvious joint involvement subsided within 10 to 30 days after the acute phase in most animals, although pathologic studies indicated that microscopic evidence of bone and joint involvement could persist long after grossly visible swelling and redness had disappeared. Microscopic study also revealed evidence of infection in and around joints which had never shown gross changes. Although the disease was reproduced consistently in a high proportion of animals injected intravenously, subcutaneous injection of the same doses produced no observable lesions.

Table 1. Joint lesions and cultures in rats infected with *Streptobacillus moniliformis.*

Experiment No.	No. arthritic/ No. injected	Percentage infection	No. positive/ No. cultured	Percentage recovery
28	5/5	100		
29A	5/5	100	3/3	100
29B	13/16	81.3	6/6	100
30	6/6	100	1/2	50
31	8/8	100	2/2	100
Total	37/40	92.5	12/13	92.3

Histologic study of a small number of infected animals showed an osteoarthritis, characterized by an acute inflammatory reaction at the sixth day, subacute to chronic at the 17th day, and chronic fibrotic and proliferative at the 33rd day. The component processes were an osteomyelitis, in most instances accompanied by periostitis, arthritis, and periarthritis. No lesions elsewhere than those described were found, either clinically or after detailed examination at autopsy.

Affected joints were ground and cultured at autopsy, and the infecting microorganism was recovered from the lesions in 12 of 13 animals cultured during the acute phase (5 to 11 days), and in one of two joints cultured after the acute phase had subsided (17 days). Of 16 heart blood cultures done at time of sacrifice, only one was positive. A degree of immunological response to the *Streptobacillus*, as evidenced by elevated plasma opsonin levels, has been demonstrated in infected animals.

This reproducible, high-incidence, infectious osteoarthritis of rats apparently specifically affects bones, joints, and periarticular tissues to the exclusion of all other tissues and organs, unlike the joint lesions found concomitantly with the generalized lesions of many systemic bacterial infections of man and animals. However, specific joint involvement has been shown with some other microorganisms. The production of an infectious osteoarthritis represents successful infection of the rat with *Streptobacillus moniliformis* at a site distant from injection. The natural occurrence of this microorganism in rats without evidence of systemic infection or joint involvement, and the characteristic joint involvement in human rat-bite fever caused by *S. moniliformis,* provoke interesting speculations about the role of infection in other arthritic conditions. The role of the filtrable "L" forms of this microorganism is at present under investigation.

EDWIN M. LERNER II
EMANUEL SILVERSTEIN

Laboratory of Pathology and Histochemistry, National Institutes of Health, Bethesda, Maryland

References and Notes

1. R. Tunnicliff, *J. Infectious Diseases* 19, 767 (1916).
2. W. L. Strangeways, *J. Pathol. Bacteriol.* 37, 45 (1933).
3. C. Levaditi, S. Nicolau, P. Poinclaux, *Compt. rend. acad. sci.* 180, 1188 (1925); *Presse méd.* 34, 340 (1926).
4. T. McP. Brown and J. C. Nunemaker, *Bull. Johns Hopkins Hosp.* 70, 201 (1942).
5. F. Parker, Jr., and N. P. Hudson, *Am. J. Pathol.* 2, 357 (1926); E. H. Place and L. E. Sutton, *Arch. Internal. Med.* 54, 659 (1934).
6. C. Levaditi, S. R. Selbie, R. Schoen, *Ann. Inst. Pasteur* 48, 308 (1932); T. Suzuki, *Japan. J. Bacteriol.* 8, 261 (1953).
7. G. J. Buddingh, *J. Exptl. Med.* 80, 59 (1944).
8. F. G. Blake, *ibid.* 23, 39 (1916); J. B. Nelson, *J. Bacteriol.* 21, 183 (1931).

21 March 1957

Atmospheric Oxidation of Homogentisic Acid: Spectrophotometric Studies

Since Boedeker (*1*) first fully described and named the metabolic disorder alcaptonuria, it has been recognized that the urine of patients with this disorder contains homogentisic acid (2,5-dihydroxyphenylacetic acid) and tends to darken on exposure to air or upon addition of alkali. It is also well known that affected individuals virtually always