Darwin[21])

$$\left.\begin{aligned}\sigma_1 &\cong 3.1\times10^{12}\ \text{sec}^{-1}\\ \alpha_1 &\cong 1.7\times10^{-2}\end{aligned}\right\}\text{for Ni,}$$

and

$$\left.\begin{aligned}\sigma_1 &\cong 30.0\times10^{12}\ \text{sec}^{-1}\\ \alpha_1 &\cong 4.3\times10^{-2}\end{aligned}\right\}\text{for Fe.}$$

Values for different wavelengths in the visible region are not very much different. In view of the approximations necessary to get the theoretical estimates, Eqs. (53) and (54), the comparison is satisfactory.

[21] C. G. Darwin, Proc. Roy. Soc. (London) **A151**, 512 (1935).

5. ACKNOWLEDGMENTS

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Neutron Damage to the Structure of Vitreous Silica

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The x-ray diffraction pattern of vitreous silica has been investigated before and after exposure to neutrons. Small, but significant, changes are observed. A relation between these changes and those caused by neutron damage to crystalline forms of silica is suggested.

INTRODUCTION

SEVERAL observations have been reported on radiation damage in silica, both crystalline and vitreous. Wittels and Sherrill,[1] for instance, found that fast-neutron irradiation of a total of $\sim2\times10^{20}$ neutrons/cm^2 caused all crystalline forms to become apparently glassy with a density of 2.26. Vitreous silica also reached this density. Primak, Fuchs, and Day[2] report an increase in density and refractive index of vitreous silica. No extended study of the x-ray diffraction pattern of irradiated vitreous silica has been published. In view of the tendency of all crystalline silica materials to become glassy with a common density and of vitreous silica to increase in density to the same value, it is of interest to examine the diffraction patterns of these damaged materials in some detail. In this report, the effect of neutron irradiation on the x-ray intensity curve of vitreous silica is discussed.

EXPERIMENTAL

Vitreous silica of high purity[3] was irradiated by neutrons at the Materials Testing Reactor to an exposure of $nvt\sim2\times10^{20}$, at a temperature of about 50°C. The specimen showed no visual change other than a slight violet discoloration. The density was found to have increased from 2.21 to 2.25, in excellent agreement with previous work. Also confirming earlier observations, the refractive index (sodium D line) increased from 1.45706 ± 0.00004 to 1.46687 ± 0.00010.

The x-ray intensity curves for both unirradiated and irradiated material were obtained using a spectrometer and Geiger counter. Values were measured at intervals of one degree 2θ or less, except at high angles where larger intervals were used. Readings were taken to $\sin\theta/\lambda=0.700$, above which point all scattering is essentially incoherent. Filtered copper and molybdenum radiations were used. Since the radiation was not strictly monochromatic, no Fourier analysis has been made of the data. It is hoped that in the future, data suitable for such analysis can be obtained. The intensities were measured by using the fixed count method in which the time to register a fixed number of counts is recorded. The probable error is less than one percent.

Spectroscopic analysis of the unirradiated material is given in Table I. The results may be in error by a factor of two or three.

TABLE I. Spectroscopic analysis of vitreous silica.

Constituent	Abundance[a] (parts per million)
V	400
Cr	200
Ti	120
Mn	60
Cu	16
Mg	6
Al	6
B	≪200

[a] May be in error by a factor of two or three.

* The Knolls Atomic Power Laboratory is operated by the General Electric Company under contract with the United States Atomic Energy Commission.

[1] M. Wittels and F. A. Sherrill, Phys. Rev. **93**, 1117 (1953).

[2] Primak, Fuchs, and Day, Phys. Rev. **92**, 1064 (1953).

[3] Provided by Corning Glass Works, Corning, New York.

RESULTS AND DISCUSSION

The results of the x-ray analysis are shown in Fig. 1. Open circles denote the unirradiated material, and the solid circles, the irradiated. Although the ordinate is arbitrary (being the recorded counts per second after correction for all factors except Compton scattering), the curves were adjusted to be equal at $\sin\theta/\lambda=0.700$. A rough quantitative relation can, thus, be assumed between the two. This relation must not be assumed too rigorously since there was a slight residual beta activity in the irradiated sample, which might tend to increase the apparent intensity measurements. However, the integrated intensities under the first peak of the two curves differ by only a few percent. The curves do not show data above $\sin\theta/\lambda=0.459$; at higher values, the intensity curves are essentially identical.

The intensity curve of unirradiated material agrees, except in minor detail, with that of Warren, Krutter, and Morningstar,[4] which was constructed from a microphotometer trace of a film made with crystal-monochromatized radiation. There is, for instance, a slight shoulder on the main peak which is not evident in their curve. However, considering the diversity of the techniques and the fact that the radiation used in the present work was not strictly monochromatic, the curves may be considered in good agreement.

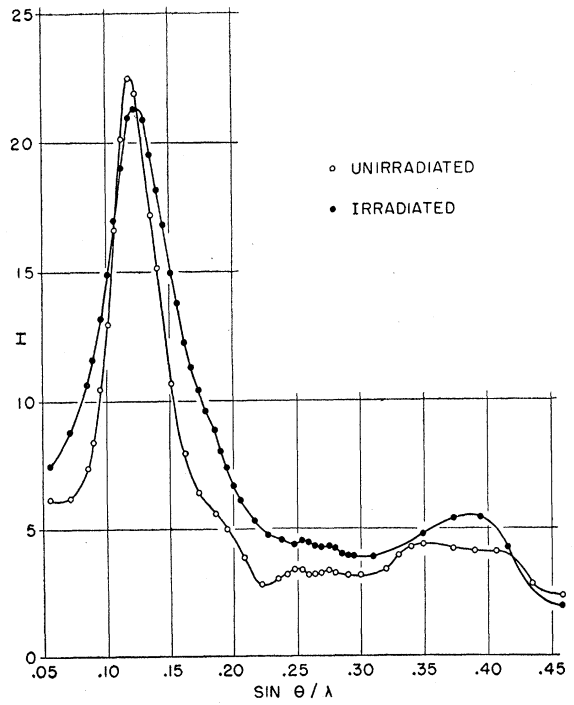

FIG. 1. X-ray intensity curves of irradiated and unirradiated vitreous silica. Vertical scale arbitrary.

The intensity curve of the irradiated vitreous silica is significantly different from that of the unirradiated. The main peak is broadened and shifted to a slightly higher value of $\sin\theta/\lambda$. The shift is from 0.120 to 0.124, and is considered to be real since every effort was made to eliminate such errors as might occur in specimen mounting, etc. The broad peak at $\sin\theta/\lambda=0.350$ to 0.400 is sharpened, and the integrated area appears to have increased. Detail in the region $\sin\theta/\lambda=0.230$ to 0.300 changed but slightly.

Although in visual appearance vitreous silica suffers little damage from neutron irradiation, there is a small but significant structural change. Wittels and Sherrill[1] suggest that all four solid forms of silica are reduced to a common vitreous phase and—more important—that there is a possibility of a solid state reaction from the vitreous phase to quartz. If such a reaction does prove to exist, particularly if it occurs from the vitreous phase regardless of the original state of the material, it would seem that the common vitreous phase must be one in which the silicon-oxygen tetrahedra have degrees of freedom not permitted in the original vitreous structure. Such freedom could be obtained by breaking of Si-O bonds, a condition that should be made evident by a decrease in the area under the first peak of the radial distribution curve of vitreous silica. If this is the case, neutron bombardment may be considered as playing a role similar to that of chemical fluxes in breaking down the tightly bonded silicon-oxygen framework.

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[4] Warren, Krutter, and Morningstar, J. Am. Ceram. Soc. **19**, 202 (1936).