

CONF-881031-64

ON THE SURVIVABILITY OF DIAGNOSTIC WINDOWS IN THE CIT REACTOR\*

A. Taylor  
Materials Science Division  
Argonne National Laboratory  
Argonne, Illinois 60439 USA

Received by 0571  
FEB 24 1989

NOVEMBER 1988

CONF-881031--64

DE89 007395

The submitted manuscript has been authored by a contractor of the U. S. Government under contract No. W-31-109-ENG-38. Accordingly, the U. S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or allow others to do so, for U. S. Government purposes.

\*Work supported by a subcontract with the Princeton Plasma Physics Laboratory, their prime DOE Contract #DE-AC02-76CH03073.

Presented at the Topical Meeting on Fusion Materials, Salt Lake City, Utah, October 9-13, 1988.

**DISCLAIMER**

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

ck

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

**MASTER**

ON THE SURVIVABILITY OF DIAGNOSTIC WINDOWS IN THE CIT REACTOR

A. Taylor,  
Materials Science Division,  
Argonne National Laboratory.  
Argonne, IL 60439 USA  
(312) 972-5005

ABSTRACT

The problem of radiation induced stresses in CIT diagnostic windows is discussed. Existing data indicate windows of existing design will probably survive if placed on the periphery of the reactor. There is a lack of adequate engineering data upon which the design and survivability of windows can be based.

INTRODUCTION

Two candidate window materials currently being considered for use in the diagnostic systems of the Compact Ignition Tokamak (CIT) are crystalline quartz for the far infrared, and fused vitreous silica for the visible and near UV (1).

CIT Radiation Field. K. Young and associates (2) have computed the neutron and gamma fields for 3000 full field 3 second shots in five different regions in the CIT reactor. The degraded 14 MeV neutron and gamma spectrum in region 2 of the reactor and typical end of life fluences in five different regions are given in Figure 1. Unlike TFTR window assemblies, no direct exposure of the windows to intense pulses of soft X-rays in the CIT system is expected. As it will not be possible to carry out extensive testing of prototype window assemblies in fusion neutron spectrum before the CIT is constructed, it will be necessary to carry out irradiations to simulate the CIT environment and derive the equivalent fusion neutron doses by calculation.

Although there is only a meager data base on the effects of fusion (14 MeV) neutrons on these window materials, an extensive literature exists for the effects of various photon and particle irradiations from which the general trends in their behavior in a CIT radiation field can be inferred. Changes in the optical absorption and radiation induced luminescence also have been investigated in considerable detail and have been reviewed for relevance to TFTR applications by Primak (3).

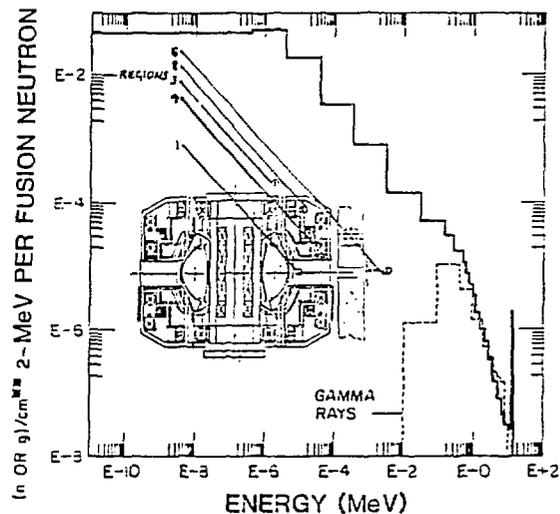


Fig. 1. Calculated neutron (solid line) and gamma (dotted line) spectra in region 2 after CIT reactor shown inset. Typical values for neutron fluences ( $\text{ncm}^{-2}$ ) and silicon kerma (MGY) at locations indicated are respectively: 1.  $3.3 \times 10^{19}$ , 94; 2.  $2.3 \times 10^{18}$ , 2.6; 3.  $2.5 \times 10^{17}$ , 3; 4.  $5.0 \times 10^{18}$ , 6.1; 5.  $7 \times 10^{16}$ , 0.6.

From the standpoint of operations of the CIT diagnostic systems, radiation induced changes in optical properties of windows may cause a degradation in a diagnostic system, whereas progressive changes in the dimensions of the window optical blanks will inevitably result in a failure of the window due to build-up of stresses at the glass to metal seals. It is probable that catastrophic implosion of the window will occur as a result of the release of accumulated stress in the brittle window materials.

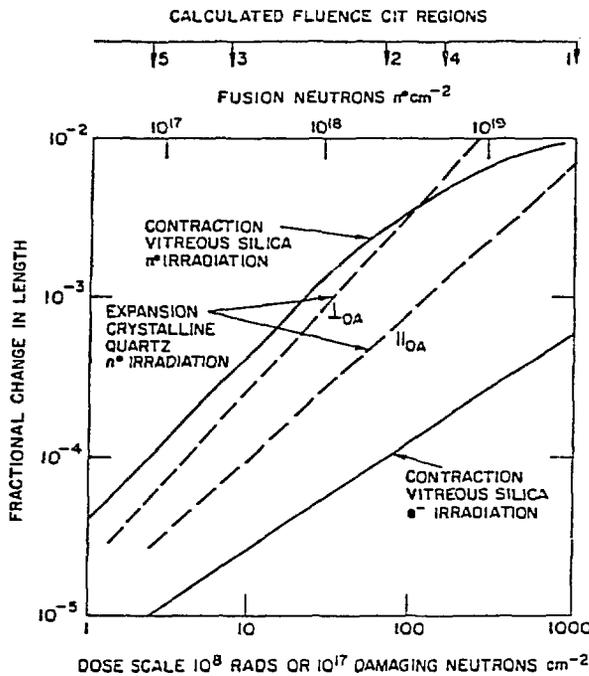


Fig. 2. Trend lines representing available data for the fraction contraction of vitreous silica and the fractional expansion of crystalline quartz versus the flux of damaging neutrons for fission reactor irradiations at ambient temperatures. Upper scales show the equivalent dosage of fusion neutrons and the end of life dosage in regions 1-5 of the CIT reactor, shown inset Fig. 1.

ON POSITIONING WINDOWS IN THE CIT REACTOR ON THE BASIS OF EXISTING DATA

To provide some guidance on positioning window in CIT, the differential strains corresponding to nominal working stresses in hard glass to metal seals of existing design are calculated. The CIT radiation doses at those critical strains must then be estimated. For this purpose, the author has represented the existing data on irradiation induced changes in linear dimensions in vitreous silica, and in quartz parallel and perpendicular to the optic axis, by the trend lines in Figure 2. The horizontal neutron dose axis represents the most effective component of the irradiation in the fission reactor experiments - the fluence of damaging neutrons.

In figures 3, 4, we illustrate the principal stresses and interference strains in window assemblies used in TFTR (4). Calculated interference strains corresponding to nominal working stresses for vitreous silica and quartz are listed in Table 1. Figure 3 shows the tensional principal stresses  $\sigma_z$ ,  $\sigma_\theta$  at the seal interface due to the radiation induced contraction in the

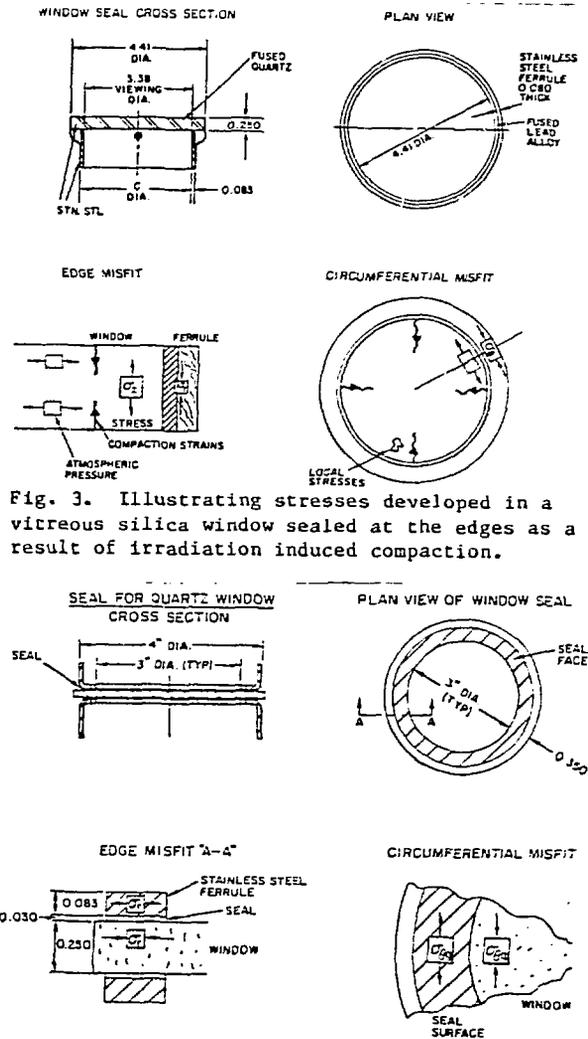


Fig. 3. Illustrating stresses developed in a vitreous silica window sealed at the edges as a result of irradiation induced compaction.

Fig. 4. Illustrating stresses developed in a crystalline quartz window as a result of radiation induced swelling.

vitreous silica. The strains are calculated on the basis of linear elasticity theory. For vitreous silica windows, a stainless steel ferrule is soft soldered to the edge of the optical blank, since there is no metal alloy with a thermal expansion coefficient matching that of silica. The ferrule and seal was treated as an elastic composite. Figure 4 illustrates the case for a C-cut quartz window which will undergo expansion in the irradiation field. Here, because the thermal expansion coefficient of quartz perpendicular to the optical axis closely matches that of stainless steel, the ferrule is assumed bonded to the face of the optical blank with a film of aluminum.

These computations show us that for the nominal working stresses listed in Table 1 and if the moduli listed for annealed silica and quartz are applicable to the irradiated state and that the ferrule is unaffected by the irradiation, then the upper limits for the radiation induced interference strain for the window assemblies are respectively  $5 \times 10^{-4}$  and  $2 \times 10^{-4}$ .

From Figure 2, this working strain corresponds to a fission neutron fluence of  $9 \times 10^{17} \text{ ncm}^{-2}$  for silica and for quartz. Because of the harder neutron spectrum within CIT and the corresponding higher damage production, we have estimated that CIT neutrons are up to three times more effective in causing dimensional changes than fission neutrons. In Figure 2, upper, end of life fluences for regions 1-5 are shown on the equivalent CIT dose. Thus on the basis of these estimates, windows of conventional design will probably only survive in regions 5 or 3 of CIT. It is possible that modification of the seals such as the use of thinner ferrules, could permit exposures to higher fluence levels. However, if the operating stress level is to be raised, or other sources of stress are identified, research must be carried out to establish the failure criteria of the window assemblies. The development of a stress relieving glass to metal seal would not only ensure greater safety against random failure, but would also give greater flexibility in positioning the diagnostic windows.

#### SUMMARY OF RADIATION INDUCED DIMENSIONAL CHANGES AND MECHANICAL PROPERTIES IN VITREOUS AND CRYSTALLINE QUARTZ

Several comprehensive reviews on the effects of irradiations on the structural changes in quartz and vitreous silica have been published (5,6,7). Wittels and Sherrill (8) showed that when virgin samples of these two materials are exposed at ambient temperatures to a fluence of reactor neutrons approaching  $2 \times 10^{20} \text{ ncm}^{-2}$  the densities are found to approach a common value characteristic of an amorphous state. Vitreous silica underwent a rapid compaction with an increase in density by 2.7%, while the density of quartz decreased by 14.3%. The origin of such density changes induced by both particle and by photon irradiations have been investigated extensively by Primak and coworkers (9,10,11). Irradiations that produce copious atomic displacement damage are the most effective in causing dimensional changes in these optical materials. Primak studied dimensional and density changes, stress birefringence and optical properties in a number of different silicas, shown in Table 2, Eernisse (12) carried out a systematic study of dimensional changes induced by charged particle bombardment and demonstrated the correlation to the displacement damage component in ions of

various masses. Primak also determined the dimensional changes parallel and perpendicular to the optic axis in specimens of optical grade quartz as a function of dosage of reactor damaging neutrons for dosages above  $5 \times 10^{19} \text{ ncm}^{-2}$  (13).

Primak concluded from observations of the internal stress distribution that the behavior of vitreous silica during reactor irradiations was complex. All reactor irradiated samples, although low in birefringence initially, developed strong stress birefringence patterns which depended upon their shape. The effect of X-irradiation on the internal stress distribution was studied at various neutron levels. Primak demonstrated that although the ionization caused a negative dilation initially, the dilation reversed in sign at higher doses. The internal stresses determined from the patterns reached about 3.5 MPa and were found to vary with dosage. He concluded that three concomitant effects were responsible for the dilations in reactor irradiated silica: a homogenization effect caused by fast neutrons, a compaction process, and a dilation caused by ionization. However, Eernisse (12) was able to analyze his data on the compaction of silica induced by  $\text{H}^+$  &  $\text{He}^+$  irradiations on the basis of the linear super-position of displacement and ionization damage. Primak and Kampwirth (10) found that the dosage dependence of compaction of vitreous silica could be well represented on a log vs log plot over a wide range of dosages for reactor neutrons, electron or X-rays. They reported that the exponent corresponding to reactor neutron induced density changes in silica was 1, while for absorbed electron irradiation and in some instances for gamma or X-rays the value was 2/3. Primak expressed the dosage dependence of the compaction caused by X and gamma rays as:

$$\delta = .762 \times 10^{-6} D^{.647}$$

where  $\delta$  is the linear contraction and  $D$  is the total ionization dosage in megarads. However,

Origin of Stress	Stress MPa	Interference Strain	Stress Type
Glass Compaction at: Seal Interference $\sigma_s$ Circumference $\sigma_c$	5 working	$5 \times 10^{-4}$	Tension
Atmospheric Pressure Working	5.6	-	Tension
Proof Test	16.8	-	Vacuum Side
Quartz Expansion at: Seal Interface $\sigma_r$ Circumference $\sigma_r, \sigma_z$	15.4 working	$2 \times 10^{-4}$ $1 \times 10^{-4}$	Compression
Atmospheric Pressure Working	4.6	-	Tension
Proof Test	13	-	Vacuum Side

Table 1. Summary of Stresses and Interference Strains

Type	Method of Manufacture	Manufacturing designations					
		General Electric	Thermal Syndicate	Heraeus	Corning	Quartz et Silice	Mitsubisi
I	Electrical fusion of quartz crystal	†101 204 125	IR Vitreosil	Infrasil	-	Pursil	-
II	Flame fusion of quartz crystal		OG Vitreosil	†Herosil †Homosil Vitreasil †+Optosil	-	-	-
III	Flame hydrolysis of SiCl <sub>4</sub>		Spectrosil	†Suprasil 1 +Suprasil 2 +Fluosil SSI.2	7943	Tetrasil	
IV	Vapor phase oxidation of SiCl <sub>4</sub>		Spectrosil WF	Suprasil W +Suprasil W2 (O <sub>2</sub> Plasma)	†7943		(Ar Plasma)
Other:				+Suprasil 200 +Suprasil 300 L +Suprasil 300 H			

†Silicas tested by Primak and Coworkers  
+Silicas tested by Friebele & Coworkers

Table 2. Some Different Types of Commercially Available Vitreous Silica

Higby et al. (14) recently reported an exponent of 1/3 for electron irradiated Heraeus Optosil. The data of reference 10 are plotted in Fig. 2 assuming the fractional change in length of silica is  $\frac{1}{3} \frac{\Delta V}{V}$ . The dosage dependence of dimensional changes in quartz at low dosages is uncertain. The data are represented by a broken line.

This survey of the literature uncovered no further data on the effect of neutron irradiation on the mechanical and fracture properties of vitreous silica. Primak's photoelastic observation indicate the importance of the development of additional internal stresses to those produced by differential strains in the window seals. The evolution of these stresses may be further complicated by the presence of structural defects such as variation of density in the vitreous silica and small twins and extrusions (11) in the quartz which may result in further local stress centers in the component.

The irradiation behavior of quartz crystals has been investigated although little work has been reported for neutron irradiations. Bahadur and Parshad (7) reported that quite low fluences of fast neutrons ( $5 \times 10^{12} \text{ ncm}^{-2}$ ) result in a measurable decrease in the sharpness of Laue diffraction spots, an increase in the resonance frequency, and an increase in hardness. Typical changes in hardness reported were from an initial unirradiated level of 400 to 468 Kg/cm<sup>2</sup>. Gamma irradiation following exposure to neutrons also resulted in a progressive increase in resonance frequency, with the increases being additive. There is very little data on dimensional changes of synthetic crystals.

In short, the existing data provides an excellent insight into the nature of the CIT window design problem, but gives us insufficient engineering data for assessing with any accuracy the survivability of window operated at differing stress levels in the CIT neutron field.

#### RESEARCH AND DEVELOPMENT NEEDED FOR CIT WINDOW ASSEMBLIES

##### Selection of Window Materials.

A wide range of optical grade vitreous silica and crystalline quartz blanks are now

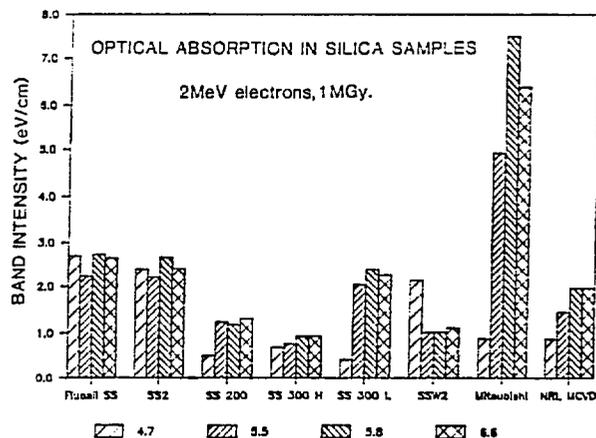


Fig. 5. Comparison of the Gaussian optical absorption bands resolved from spectra of peaks observed in various high purity silicas induced by electron irradiation.

available commercially. A list of commercial sources of vitreous silica categorized into Types I-VI by Bruckner(15) according to the manufacturing techniques is given in Table 2. Under the category of 'Others' are listed silicas made by processes that have been implemented since 1970. These methods include the modified chemical vapor deposition technique (16), the sol-gel process (17), and oxidation of pure silicon by thermal, electrochemical or ion implantation techniques. The majority of studies of radiation induced effects to date have been carried out on types I-IV (5) and only recently has work begun on the newer materials. For example, Friebel et al (18) have compared the optical absorption in various amorphous silicas induced by electron irradiation at ambient temperatures. A comparison of the intensities of the radiation induced optical bands at 4.7, 5.5, 5.8, and 6.6 eV in the high purity silicas, shown in Table (2), is given in Figure 5. The study shows there is significantly less absorption in samples of synthetic (and 'purer') silicas than in fused quartz, but there is no direct correlation with impurity content such as the OH<sup>-</sup> level. The infrared absorption of the high purity synthetic samples was insensitive to the electron radiation. Thus it will be interesting to see whether these newer materials will exhibit a slower rate of a degradation of their optical transmission when subjected to nuclear reactor irradiation. Neutron induced dimensional changes would not be expected to be impurity sensitive to first order. However, the interaction between impurities and neutron produced defects may have a significant effect on the production of color centers and also on the photo-luminescence behavior.

Therefore, in selecting the optical blanks for the CIT diagnostic windows, it appears worthwhile to screen the behavior of a representative sample of commercial materials for their optical behavior prior to investigating neutron induced dimensional changes.

#### Determination of Dimensional Changes in Terms of Equivalent Fusion Fluences

To determine the differential stresses developed in the glass to metal seal of the CIT windows, requires that the dimensional changes in the selected quartz and vitreous silica be redetermined. These dimensional changes must be related to calculated neutron fluences and the neutron spectra at the proposed locations of the window within the CIT structure. Since the larger contribution to the dimensional changes originate from the atomic displacement damage, see data Figs. 1 & 2, either ion or reactor irradiations can be employed to simulate the CIT case. In principle, the dimensional changes corresponding to CIT reactor can be calculated from the ion beam data using a calculation based on the computed primary knock-on spectra for

neutrons and ions and from the Lindhard et al. (19) treatment of the energy partition between nuclear and electronic processes for each primary event. The method has been applied to assess the dimensional changes in TFR window (1).

It is recommended by this author however, that for this program irradiations be carried out in fission reactors where the spectrum of the neutrons is known. Data from neutron irradiation is preferred from an engineering standpoint because reactor irradiation represents a closer simulation of the anticipated CIT service conditions. The dimensional changes can be readily observed in window test blanks, complexities such as the development of local stress can be studied, and the results compared with the behavior of window test assemblies. In contrast, ion damage studies require specialized techniques to observe a narrow region of non uniform damage, 1-10  $\mu$  deep. Further the technique cannot be readily employed for testing the glass to metal seals. The disadvantages associated with reactor irradiation, i.e., longer irradiation times and handling residual radioactivity, are outweighed by the more direct nature of the tests. Conversion from the measured fission fluence to the harder CIT fusion fluence can be accomplished using recently developed computer codes such as SPECOMP and SPECTER (20,21). These calculations lead to a simple multiplier by which the fluences measured in the reactor can be scaled to represent the CIT case. The concentration of isotope produced is also calculated. The scaling factor will depend on the neutron spectrum at the particular location in the CIT. Preliminary estimates by the author using existing data indicate that the multiplier is approximately 3 for the CP5 reactor irradiations used by Primak. An experimental verification of the calculated conversion factor using RTNSII or another source of high energy neutrons should be considered as part of this program.

#### Determination of Mechanical Behavior of Window Materials

In order to determine the mechanical behavior of the CIT windows, data for the strength, the moduli and Poissons ratio will be needed for the both vitreous silica and quartz in the irradiated state. To establish whether operating stress levels will result in propagation of crack nuclei, experimental parameters for a fracture mechanics analysis will also be required. Data will be needed for the stress intensity factor, the crack propagation rate, and parameters for a Weibull statistical analysis. A computer model using finite stress element analysis may then be constructed taking into account the measured distribution of stresses in each optical blank in order to determine the mechanical behavior of the window assembly. Varshneya (22) has recently reviewed the application of computer models in the design

of glass to metal seals. The operating stress level chosen for the window will determine the accuracy with which these parameter will be determined and also the number of samples to be tested. Some verification of the behavior of the window assemblies will be needed, and this will require irradiation of a complete assembly.

#### DISCUSSION OF PROPOSED PROGRAM

The first priorities for the CIT window program is to settle the question of the location of the windows in CIT and determine approximately the operating stress level expected. This will largely fix the size of the program. Operating stress levels close to currently used nominal values will reduce the extent of the mechanical testing program which for testing the irradiated state will be costly. A computer modelling effort is a desirable element of the program in general and should yield survival probabilities and criteria to evaluate destructive tests on window assemblies. The author believes that an evaluation of ways to construct a stress relaxing seal is worthwhile, whatever the operating stress level selected since all radiation induced stress build-up is potentially hazardous.

#### ACKNOWLEDGEMENTS

Work supported by a subcontract with the Princeton Plasma Physics Laboratory, their prime DOE Contract #DE-AC02-76CHO3073. The author acknowledges helpful discussions with M. Billone and S. Majumdar, MCT-ANL, D. Smith, FPP-ANL, and F. Clinard, LASL.

#### REFERENCES

- J. A. SCHMIDT and H. P. FURTH, "Compact Ignition Tokamak, Diagnostic Plan", PPPL/A-860606-P-03 June 6, 1986.
- K. M. YOUNG (1987) CIT Radiation Levels and TFTR Observations, Planning Meeting on Neutron Irradiation Effects and Materials Research and Development, Lawrence Livermore National Laboratory, Livermore, CA, April 7, 1987.
- W. PRIMAK, "Radiation Damage in Diagnostic Window Materials for TFTR", 1981 ANL/FPP/TH-146.
- M. A. MOZELESKI and G. LEWIN, "Bakeable Optical Windows for TFTR", (1981) IEEE Trans.
- E. J. FRIEBELE and D. L. GRISCOM, "Treatise on Materials Science and Technology", Radiation Effects in Glass, 17 Glass II. p. 257 (1979).
- E. LELL, N. J. KREIDL, and J. R., HENSLER, "Radiation Effects in Quartz, Silica & Glasses", Prog. in Ceramic Science, 4 Ed. Burke, (1966).
- H. S. BAHADUR and R. PARSHAD, "Effect of Irradiation on Crystal Defects in Quartz", IEEE Trans on Nuclear Science, NS 32 No.2, 1169 (1985).
- M. S. WITTELS and F. A. SHERRILL, "Radiation Damage in SiO<sub>2</sub> Structures", Phys. Rev. 93 No. 5, 1117 (1954).
- W. PRIMAK and E. EDWARDS "Radiation-Induced Dilatations in Vitreous Silica", Phys. Rev. 128 No. 6, 2580 (1962).
- W. PRIMAK and R. KAMPWIRTH, "The Radiation Compaction of Vitreous Silica", J. of Appl. Phys. 39, No. 12, 5651 (1968).
- W. PRIMAK, "Extrusion of Quartz on Ion Bombardment Further Evidence for Radiation Induced Stress Relaxation of the Silica Network", Phys. Rev. B 14 No. 10, 4679 (1976).
- E. P. EERNISSE, "Impaction of Ion-Implanted Fused Silica", J. Appl. Phys. 45 No. 1. 167, (1974).
- W. PRIMAIC, "Fast-Neutron-Induced Changes in Quartz & Vitreous Silica", Phys. Rev. 110, No.6 (1958).
- P. L. HIGBY, E. J. FRIEBELE, C. M. SHAW, M. RAJARAM, E. K. GRAHAM, D. L. KINSER, and E. A. WOLFF, "A Survey of Radiation Effects on the Physical Properties of Low Extension Coefficient Glasses and Ceramics", J. A. Ceram. Soc. 71 No. 9, 796-302 (1987).
- R. BRUCKNER, J. Non-Cryst. Solids 5, 123-1175 (1970).
- W. G. FRENCH, R. E. JAEGER, J. B. MACCHESNEY, S. R. NAGEL, K. NASSAU, and A. D. PEARSON, "Optical Fiber Telecommunications", S. E. Miller and A. G. Chynoweth Ed. Academic Press, NY 1979, p. 233-262.
- S. P. MUKHERJEE, J. Non-Cryst. Solids 42, 477-488 (1980).
- E. J. FRIEBELE, P. L. HIGBY, and T. E. TSAI, "Radiation-Induced Optical Absorption in Amorphous Silicas Prepared by Different Techniques", Diff. & Defect Data. 53-54, 203-212 (1987).
- J. LINDHARD, V. NIELSEN, S. M. SCHARFF, "Approximate Method in Classical Scattering by Screened Coulomb Fields", Kgl. Danske Videnskab. Selskab, Mat-FYS. Medd. 36 No. 10, 1968).
- L. R. GREENWOOD and R. K. SMITHERS, "Spector Neutron Damage Calculations for Materials Irradiations", ANL/FPP/TM 197 JAN 1985.
- L. R. GREENWOOD, "Radiation Damage Calculations for Compound Materials", Proceedings of the 14th International Symposium on Effects of Radiation on Materials, Andover, Mass, June 1988 (to be published ASTM/STP Series).
- A. K. VARSHNEYA, "Stresses in Glass to Metal Seals", Treatise on Materials Science and Technology, 22, 242 (1982), Glass III, edited by M. Tomozawa, R. H. Doremus, Academic Press, Inc., New York.