INVESTIGATION OF FIBEROPTICS DURING AND AFTER FISSION PRODUCT GAMMA IRRADIATION*

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SUMMARY

The transmission losses and possible annealing methods with various glasses and fiberoptics were studied after fission product gamma irradiation. Irradiation was performed in the thermal column of the 250 kW TRIGA Mark II reactor, Vienna, and transmission losses in visual range were investigated by optical spectrometry.

A total of 6 glass samples and 3 fiberoptic samples have been exposed to gamma doses varying from 8×10^4 R to 2.3×10^9 R. Thermal annealing parameters were studied at 300 C, 400 C and 500 C, optical annealing was performed by an arc lamp, by an UV-light source and by a pulsed UV-laser. In addition thermal annealing experiments were carried out simultaneously to gamma exposure resulting in shorter annealing periods and lower temperatures.

INTRODUCTION

Flexibility, large bandwidth, electromagnetic immunity and picture transmission are characteristics of optical fibers that make them particularly well suited to the requirements for endoscopes in nuclear power plants. The constraint on the use of fiberoptics in these applications is the sensitivity of fibermaterials to the nuclear radiation produced by the fuels in the core or by spent fuel elements in the spent fuel storage pit. Fortunately, the reduction in impurity levels of constituent glasses has also reduced their radiation sensitivity, increasing their potential application in nuclear power plants and in various experiments. In the present paper, the transient radiation response and thermal and optical annealing of a number of glasses and fiberoptics is examined. The effects of interest are

- the optical transmission loss which decreases the information of the transmitted picture
- the annealing temperature as a function of time which is responsible for the reapplication of the instrument after radiation exposure
- the possibility of optical bleaching.

Radiation effects in optical waveguides have been examined in step-index fibers by a number of investigators[1-5] with attention to absorption and luminescence. In the previously reported studies dealing with radiation effects in fiberoptics only Co-60 gamma sources[6,7], pulsed electron irradiation[8] and 14 MeV neutrons[5] were used. Many investigations were performed with fiberoptic plates[6] but thermal annealing processes are not reported except in[7]. The possibility of optical bleaching by UV-light for quartz glass is briefly reported by Schulman and Compton[9]. Technological requirements including the need for optical transmission of pictures during radiation exposure over the whole range of gamma energies and the use of fiberoptics as endoscopes in nuclear technology have motivated a more comprehensive study of these effects.

The investigations were performed in the thermal column of the TRIGA Mark II reactor, Vienna (250 kW). The gamma-dose rate was measured with TLD-100, shielded by cadmium foils for thermal neutron absorption, in various radial distances from the core center and at different reactor power levels (50 W - 250 kW). The TL-dosimeters were calibrated in a Co-60 radiation field with the secondary particle equilibrium conditions fulfilled.

EXPERIMENTAL PROCEDURES

In previous reports[1-8] special doped glasses and fiberoptics have been investigated for their transmission properties before and after irradiation, while the present work was concentrated on the behavior of commercially available glasses. The glasses however became opaque after being exposed to a given gamma dose. Therefore methods have been investigated to improve the transmission properties once the materials have been exposed and have partially lost their transmission in the visual range. The mentioned methods can be divided into two categories which are

- thermal annealing methods
- optical bleaching methods.

For the experiments the following glass materials from commercial suppliers were investigated: softglass, Duran 50, quartz glass, high synthetic silica samples, special cerium doped glasses. In addition fiberoptic grade rod samples were provided by various manufacturers including Schott-Mainz (Federal Republic of Germany FRG), American Optical Corporation (United States of America USA) and Barr & Stroud (Great Britain GB). Finally also plexiglass-rod samples were investigated.

All specimen had a cylindrical geometry with a length of 60 mm and a diameter of 5 mm, only one fiberoptic sample had a rectangular cross section (5 x 6 mm).

Optical transmission experiments were performed by using a standard double-beam spectrophotometric technique prior to exposure and one hour after gamma exposure or annealing process. The transmission spectra were recorded in the visible range from 400 to 700 nm.

Irradiations of the samples have been performed in the thermal column of the 250 kW TRIGA Mark II reactor, Vienna. The gamma-dose rate varied from 1.6×10^5 R/h to 5.4×10^6 R/h depending on the irradiation position. To avoid thermal neutron activation and to increase the gamma-dose rate all samples had been shielded by cadmium.

Annealing experiments were carried out at 300 C, 400 C and 500 C, except for the rectangular fiberoptic samples and the plexiglass sample which had upper temperature limits of 200 C and 130 C, respectively.

Optical bleaching was performed with an arc lamp and with a pulsed UV-laser.

RESULTS

Preliminary investigations on natural annealing of the samples by storing them at room temperature for more than six months did not show any significant effect on the transmission properties. Figure 1 shows the optical transmission loss in the visual range for Duran and softglass as a function of the gamma dose and of the annealing temperatures.

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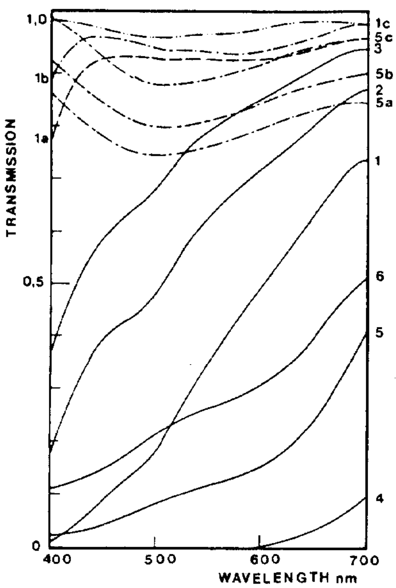

Figure 1. Softglass and Duran

DURAN 50

(1) 6.4×10^5 R (2) 3.2×10^5 R (3) 1.6×10^5 R

(1a) 6.4×10^5 R, 1 h 300 C
(1b) 1 h 400 C
(1c) 1 h 500 C

Softglass

(4) 6.4×10^5 R (5) 3.2×10^5 R (6) 1.6×10^5 R

(5a) 3.2×10^5 R, 1 h 300 C
(5b) 2 h 300 C
(5c) 3 h 300 C

The spectral characteristics of the transmission in Duran 50 and softglass measured 1 hour following exposure to various gamma doses in the thermal column. Dose rate 1.6×10^5 R/h.

The small absorption band of Duran around 470 nm is associated with a 12.8 to 13 weight% B_2O_3 impurity. Nevertheless, this band is annealed after 1 hour at 300 C. The dominant absorption band in the near UV however is not annealed by temperatures less than 400 C. The transparency of softglass is very small even after a moderate gamma dose of 8×10^4 R. The transmission in this case is less than 25% for blue light and about 50% for red light. Two absorption bands can be observed around 450 nm and near 620 nm which can be removed completely by annealing at 300 C.

Figure 2 shows the optical transmission loss induced in quartz glass rods in the range from 400 to 700 nm after an one-hour gamma exposure to doses ranging from 8×10^4 R to 6.4×10^5 R. The spectra show the typical A-absorption band at 540 nm which can be related to the presence of impurities within the material thus creating trapping centers (electron traps) and recombination centers (hole traps). The smoky color induced by gamma exposure (above 10^5 R) is intimately related to the presence of aluminum (1 - 10 ppm) substitutionally incorporated into the quartz lattice in place of silicon.

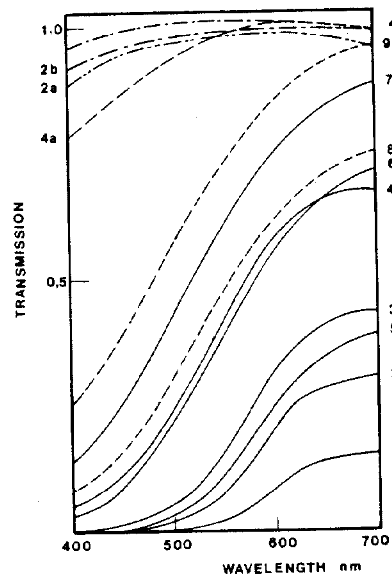

Figure 2. Quartz

(1) 6.4×10^5 R (2) 3.2×10^5 R (3) 1.6×10^5 R

(4) 6.4×10^5 R LASER, λ = 193 nm, 760 µs

(5) 1.6×10^5 R LASER, λ = 193 nm, 190 µs repetition rate 3.7 Hz

(6) 6.4×10^5 R arc lamp 1 h

(7) 6.4×10^5 R arc lamp 2.5 h

(1a) 6.4×10^5 R, 1 h 300 C
(1b) 1 h 400 C
(1c) 1 h 500 C
(1d) 6 h 300 C
(1e) 12 h 300 C

The optical characteristics of the transmission in quartz glass measured 1 hour following the exposure to various gamma doses in the thermal column. Dose rate 1.6×10^5 R/h.

This aluminum absorption band cannot be annealed by temperature treatment at 300 C even after 12 hours. However, exposure to 350 C or more destroys the centers and the sample turns colorless. For example at 500 C the annealing period is less than 1 hour. A simultaneous gamma and temperature exposure, however, protects the quartz glass from absorption bands even at a temperature below 300 C.

Optical bleaching was performed with a commercial 1000 W projection lamp. It took about 13 hours to increase the transmission from 17% to about 60% after a gamma exposure of 6.4×10^5 R. Better results were obtained by an arc lamp which has a sun-like spectrum with relative maxima at 380 nm and 750 to 800 nm. The emission intensity of an arc lamp near the C-center of quartz ($\sim$ 215 nm) is strong enough to bleach the A-band of a 6.4×10^5 R exposed sample within 10 hours. At a lower gamma dose ($\sim 1.6 \times 10^5$ R) optical bleaching with this method took about 2 hours. Optical bleaching experiments were also performed with special ArF (λ = 193 nm), KrCl (λ = 222 nm) and KrF (λ = 249 nm) UV-laser light. The pumped laser bleached the 6.4×10^5 R exposed samples after 760 µs effective time (real time 30 min) from 17% to about 65% transmission.

Suprasil I (Figure 3) being the most radiation resistant glass investigated[2] was exposed to a maximum gamma dose of 10^9 R.

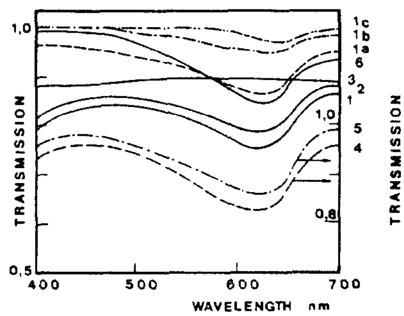

Figure 3. Suprasil I
(1) 2.3×10^9 R (2) 1×10^9 R (3) 6.3×10^7 R
(4) 2.3×10^9 R, arc lamp 30 min
(5) arc lamp 5 h
(6) 1.0×10^9 R LASER, λ = 193 nm, 190 μs
(1a) 2.3×10^9 R, 1 h 300 C
(1b) 1 h 400 C
(1c) 1 h 500 C
The spectral characteristics of the transmission in Suprasil I measured 1 hour following exposure to various gamma doses in the thermal column. Dose rate 1.6×10^5 R/h and 5.4×10^6 R/h.

Up to 6.3×10^7 R, no transmission loss can be observed. A gamma dose above 10^8 R induces an absorption band near 620 nm. Impurity analysis showed that this material contains only very small amounts (less than 0.1 ppm) of Al. A transparency improvement can be obtained by annealing above 250 C. Samples with high dose exposure (2.3×10^9 R) are annealed in the visible range within 1 hour when exposed to 500 C, except around 620 nm where a small transmission loss exists. However, the transmission after gamma exposure is about 98% of the original transmission. Therefore, at an elevated temperature level (> 450 C) fused silica can be used with gamma doses greater than 10^9 R.

Both the UV-laser light and the arc lamp shift the whole visual transmission spectrum towards better transparency but the relative depth of the absorption band at 620 nm is still the same. It seems that this absorption band cannot be bleached optically.

A commercially available radiation resistant glass is manufactured by Schott, Mainz, FRG (Figure 4) which is a cerium doped bulk glass containing 2.5% CeO_2 (type BK7G25). After being exposed to 10^6 R a coloration can be observed which increases slightly after exposure to 10^8 R. In the UV range the edge of the UV Ce^{4+} band near 240 nm causes a yellow appearance in this glass[10].

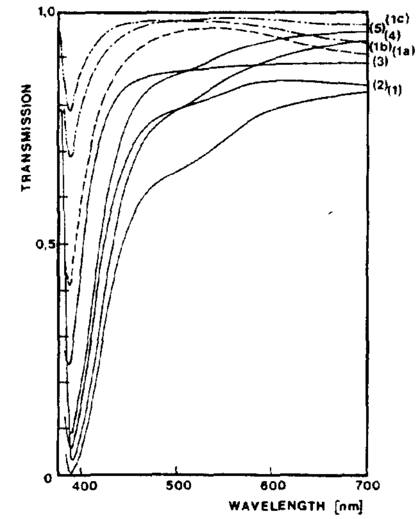

Figure 4. Cerium-doped glass BK7G25
(1) 2.3×10^9 R (2) 1.0×10^9 R (3) 6.3×10^7 R
(4) 2.3×10^9 R LASER, λ = 193 nm, 1.5 h real time, effective time 360 μs, repetition rate 5 Hz
(5) 1.0×10^9 R LASER, λ = 193 nm, 30 min real time, effective time 760 μs, repetition rate 30 Hz
(1a) 2.3×10^9 R, 1 h 300 C
(1b) 1 h 400 C
(1c) 1 h 500 C
The spectral characteristics of the transmission in cerium-doped glass (BK7G25) measured 1 hour following exposure to various gamma doses in the thermal column. Dose rate 1.6×10^5 R/h and 5.4×10^6 R/h.

The transmission spectra of glass rods exposed to doses greater than 3.6×10^7 R show a second absorption band near 510 nm, which can be annealed above 300 C. Thermal annealing of 2.3×10^9 R exposed samples at 400 C and 500 C improves the transmission for wave lengths above 450 nm. Below this wave length the transparency is about 70% to 80%. For lower annealing temperatures the annealing time increases rapidly (i.e. 13 h at 300 C for samples exposed to 2.3×10^9 R). It has also been found that the annealing time increases with the exposure dose. With UV-laser application the slight absorption band near 510 nm can be bleached, but the stronger band near 380 nm cannot be removed. Experiments with the arc lamp showed negligible transmission improvements.

A sample of bulk plexiglas rod (Figure 5) was exposed to 2.7×10^6 R. Below 10^6 R a small transmission loss was observed in the visual range, at larger gamma doses an absorption band between 400 nm and 450 nm appeared. Due to the temperature stability of the sample, annealing experiments had to be performed below 120 C. After 1 hour at 100 C the transmission for λ > 450 nm was equal to the original transmission prior to exposure. Optical bleaching even for longer periods showed no transmission improvement.

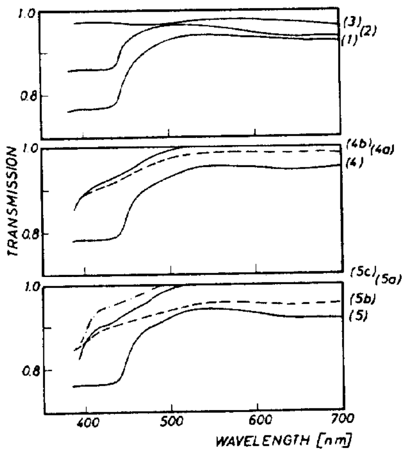

Figure 5. Plexiglass
(1) 2.7×10^6 R (2) 1.6×10^6 R (3) 6.4×10^5 R
(4) 2.5×10^6 R (4a) 2 h 80 C (4b) 2 h 90 C
(5) 2.7×10^6 R (5a) 2 h 100 C (5b) 1.75 h optical annealing (5c) 1 h 110 C

The spectral characteristics of the transmission in plexiglass measured 1 hour following exposure to various gamma doses in the thermal column. Dose rate 1.6×10^5 R/h and 5.4×10^6 R/h.

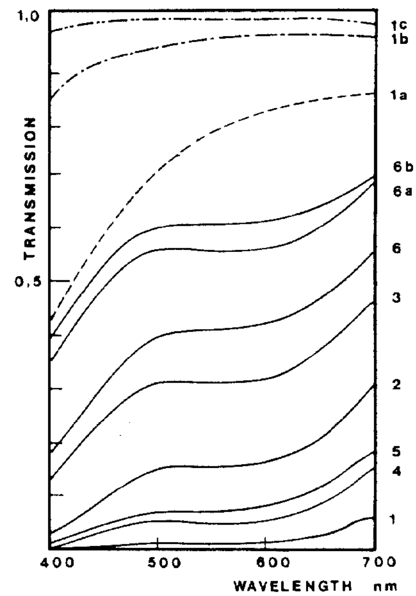

Figure 6. Schott-Mainz fiberoptic
(1) 6.4×10^5 R (2) 3.2×10^5 R
(3) 3.2×10^5 R LASER, λ = 193 nm, 300 μs, repetition rate 1 Hz
(4) 6.4×10^5 R, projection lamp 1 h
(5) projection lamp 4 h
(6) 1.6×10^5 R
(1a) 6.4×10^5 R, 1 h 300 C
(1b) 1 h 400 C
(1c) 1 h 500 C
(6a) 1.6×10^5 R, arc lamp 1 h
(6b) arc lamp 1.5 h

The spectral characteristics of the transmission in Schott-Mainz fiberoptics measured 1 hour following exposure to various gamma doses in the thermal column. Dose rate 1.6×10^5 R/h and 5.4×10^6 R/h.

Fiberoptics

All commercial fiberoptics investigated had a step refraction index profile. The core of the Schott Mainz fiberoptic is optical glass with a refraction index n_1 = 1.63, while the index of the cladding glass is n_2 = 1.52. These values give a numerical aperture of 0.589 which is equal to the angle of incidence of = 36°.

Detailed data of the fiberoptics supplied by American Optical Corp. were not available. Barr & Stroud fiberoptics have a working temperature range from -200 C to 450 C, with a maximum permitted temperature of 500 C. Below 350 nm no transmission is observed even in the unexposed state. The numerical aperture is 0.43 and the maximum angle of acceptance is 50°.

When exposing the Schott fiberoptic to 3.2×10^5 R an absorption band near 540 nm can be observed (Figure 6), similar to quartz glass with its A-absorption band, as reported in[11]. This is an indication that the core material of the fibers contains traces of aluminum, which had been verified by neutron activation analysis resulting in 7 ppm of Al. A second strong absorption band exists in the near UV.

Annealing the sample at 300 C for one hour removes the absorption band at 540 nm, but not the strong absorption in the near UV. Thermal treatment at 400 C for 1 hour increases the transmission to 90%, but a small absorption in the UV remains, which can only be annealed completely by 500 C. An extended annealing period of 300 C for 8 hours leads to a saturation value in transmission which cannot be improved by increasing the time.

Therefore, it can be concluded that two different color centers at different energetic levels are created by ionizing radiation. More stable centers need higher annealing temperatures. Nevertheless, the Schott fiberoptic has also a good optical bleaching behavior by laser pulses or by arc lamp treatment for samples exposed to a gamma dose below 1.6×10^5 R. The recovery speed of the 540 nm absorption band depends on the annealing methods with the thermal treatment being the slowest method, followed by optical arc lamp bleaching and being fast with pulsed laser light (Figure 7).

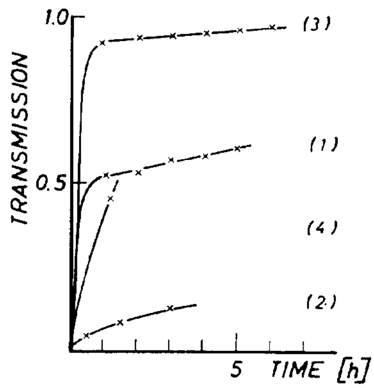

Figure 7. Annealing methods

Competition of recovery rate of a 6.4×10^5 R irradiated fiberoptic at 540 nm. The time for the Laser (4) is indicated as real time, the effective time is 300 μs. Thermal annealing is better than the method with the arc lamp (1). For the thermal annealing the large increase within the first hour is obvious afterwards the transmission improvement is negligible. The transmission improvement by an arc lamp (1) exposure within the first hour is smaller than (3) but afterwards the slope of (1) is slightly steeper than (3).

(1) arc lamp
(2) projection lamp
(3) thermal 300 C
(4) LASER, λ = 193 nm, real time 70 min, effective time 300 μs

The spectral behavior of exposed American Optical fiberoptics is completely different as shown in Figure 8. After exposure to 3.2×10^5 R, strong absorption can be observed in the visual range; it is greater at shorter wavelengths. This absorption cannot be bleached with a temperature treatment below 160 C. Beyond this temperature the fiberoptic ends become dark brown due to excess temperatures. By neutron activation analysis a 24 ppm Li- and a 0.2 ppm B-content was detected. Further traces of germanium are suspected in the material, because its transmission properties are similar to the investigations reported by[8]. Optical bleaching of this fiberoptic by arc lamp light shows a quick recovery within the first hour but afterwards the transmission remains constant.

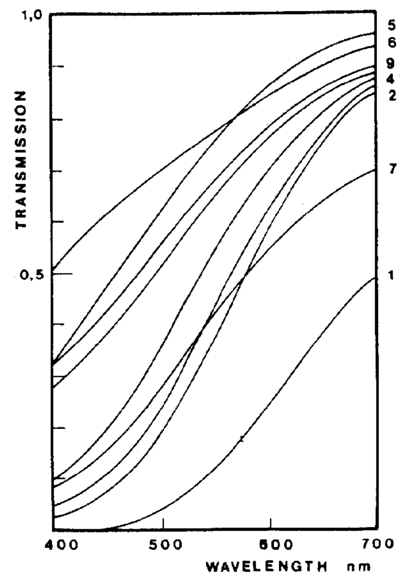

Figure 8. American Optical Corporation fiberoptic

(1) 3.2×10^5 R
(2) annealing of (1) 1 h 100 C
(3) (1) 1 h 130 C
(4) (1) 1 h 160 C
(5) (1) 1 h 200 C
(6) (1) 1 h 250 C
(7) 1.6×10^5 R, arc lamp 1 h
(8) arc lamp 2.75 h
(9) arc lamp 4.5 h

The spectral characteristics of the transmission in American Optical Corp. fiberoptics measured 1 hour following exposure to various gamma doses in the thermal column. Dose rate 1.6×10^5 R/h and 5.4×10^6 R/h.

The spectral behavior of exposed Barr & Stroud fiberoptics is characterized by a strong transmission loss at short wavelength in the visual range (Figure 9). Exposing this material to 3.2×10^5 R, the fiberoptic becomes opaque for a wavelength below 400 nm. This limit increases to 500 nm after an exposure to 6.4×10^5 R. Nevertheless, this fiberoptic has excellent thermal annealing properties even at a temperature below 300 C. Annealing the 6.4×10^5 R exposed sample for 1 hour at 300 C the original transparency is obtained. At a lower temperature (200 C) only a small absorption band around 400 nm remains in the 1.6×10^5 R sample. Simultaneous thermal annealing and gamma exposure to 1.1×10^6 R reduces the annealing temperature and increases the transmission. While a non-annealed sample becomes totally opaque, a simultaneously annealed sample has a transmission of 95% to 99%. With optical bleaching by arc lamp light the transmission improvement was better in the red range than in the blue. A linear relationship between transmission increase and time has been found.

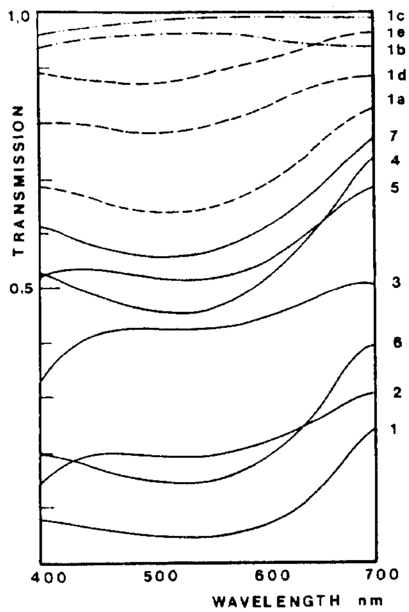

Figure 9. Barr & Stroud fiberoptic

(1) 6.4×10^5 R (2) 4.8×10^5 R (3) 3.2×10^5 R

(4) 1.6×10^5 R

(5) 6.4×10^5 R, arc lamp 30 min
(6) arc lamp 2.25 h
(7) arc lamp 5.75 h

(8) 4.8×10^5 R, arc lamp 2.25 h
(9) arc lamp 5.75 h

(2a) annealing of (2) 1 h 250 C
(2b) (2) 1 h 175 C
(4a) (4) 1 h 200 C
(4b) (4) 1 h 275 C

The spectral characteristics of the transmission in Barr & Stroud fiberoptics measured 1 hour following exposure to various gamma doses in the thermal column. Dose rate 1.6×10^5 R/h and 5.4×10^6 R/h.

CONCLUSIONS

The experiments show that commercially available glasses and fiberoptics may be exposed to high gamma doses. The radiation damage in the material resulting in a transmission loss can be annealed either by thermal or by optical treatment within a reasonable range of parameters.

Quartz glass and all fiberoptics with a SiO_2 core can be treated successfully by optical bleaching. Practically all transmission losses in the visual range can be annealed at 500 C. If the annealing process is performed simultaneously to the gamma exposure, shorter annealing periods or lower temperature are possible for a given transmission improvement. With radiation resistant fiberoptics visual inspections of reactor components can be extended to such areas which are inaccessible by present visual inspection methods (i.e. control rod guide tubes, single fuel rods, reactor internals). Conventional visual inspection equipment like binoculars, TV cameras, periscopes etc. continue to be valuable tools in present reactor technology. Fiberoptics increases the range of applications if lack of space, inconvenient environmental conditions or inaccessability prevents other visual methods.

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Helmuth Boeck, born December 12, 1942 in Vienna, Austria, received an engineering degree from the Technical University Vienna in 1966 and the PhD. in reactor technology in 1969. Since 1967 he works at the Atominstitut of the Austrian Universities directing the TRIGA reactor operation group. He is specialized in the fields of reactor safety and reactor technology where he published more than 60 papers and research reports. In addition he lectures at the Technical University Vienna and at the University of Linz in the above mentioned fields. Dr. Böck is member of the German Nuclear Society (KTG), of the Austrian Nuclear Society (OKTG), of the Austrian Atomforum (OAF) and of the Austrian Association of Radiation Protection (OVS).

Josef Siehs, born August 25, 1954 in Vienna, Austria, finished his thesis in November 1980 about the behavior of fiber optics under gamma irradiation. He will receive the PhD. from the University Vienna in spring 1981. J. Siehs is a member of the Austrian Nuclear Society (OKTG), the Austrian Atomforum (OAF) and the Austrian Physical (OPG) and Chemical Society (CPG).