Optical absorption of neutron-irradiated silica fibers

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Abstract
Silica-based optical fibers capable of transmitting light in the ultraviolet, visible and infrared regions of the electromagnetic spectrum are gaining widespread use in fusion reactor diagnostic systems. To assess radiation damage in these optical materials, the optical absorption has been measured at room temperature in the interval 250–2000 nm of neutron-irradiated silica fibers containing low or high hydroxyl concentrations. Fiber irradiations were done in either low (1.4 × 10^{21} n/m^2) or high (1.1 × 10^{23} n/m^2) neutron fluence (E > 0.1 MeV) regions of the Los Alamos Spallation Radiation Effects Facility. Attenuation in high-fluence irradiated fibers exceeds 10^4 dB/km for wavelengths less than about 700 nm, presumably due to radiation-induced absorption of peroxy radical, non-bridging oxygen hole and E'-type centers. Even the low fluence exposures induced considerable fiber absorption in this spectral region. High-OH content fibers exhibit significant radiation-induced absorption above 1500 nm, in addition to the intrinsic absorption due to OH stretching modes.

1. Introduction
Utilization of optical fibers for transmitting diagnostic information on fusion reactor plasma performance has increased in recent years primarily due to improved fibers that offer enhanced radiation resistance [1]. And although the general consensus is that pure silica-core/F-doped silica-clad fibers are currently the best available materials for this application [2], they are not ideal because they suffer from radiation-induced optical absorption and luminescence [3]. Furthermore, during reactor operation scattered electrons produce Cherenkov radiation in the silica fibers, thus contributing additional unwanted emission that complicates transmission of the diagnostic signal [4]. These problems notwithstanding, silica-based fibers currently offer the best properties for diagnostic use in the harsh environment of fusion reactors [5].

Much research has been devoted to understanding radiation-induced defects in silica and to methods for mitigating the damage (for an excellent review see [3] and references therein). High-quality unirradiated silica fibers generally exhibit weak optical attenuation (20–40 dB/km) in the visible portion of the spectrum, which is usually attributed to intrinsic defects (vacancies, etc.) [6], impurities (OH, Cl, for example) [7], and processing methods [8]. Of course radiation produces defects in silica by trapping electrons and/or holes at intrinsic precursor sites [9], and atomic displacements occur via either knock-on [10] or radiolytic processes [11].

Three types of defect centers in irradiated silica have been identified: E' [12], peroxy radical [13], and non-bridging oxygen hole center (NBOHC) [14]. Usually denoted (=Si··), the E' center is an unpaired electron (·) in a dangling sp^3 orbital of the silicon atom bonded to three oxygen atoms (=). There are several variants of this type of defect, all characterized by optical absorption in the ~ 250 nm spectral region. The peroxy radical (=Si–O–O·) is a hole-center defect that is thought to exhibit optical absorption in the region near 600 nm, although some researchers have suggested that it may absorb near 250 nm [3]. The NBOHC (=Si–O·) exhibits optical absorption near 630 nm and is described as a hole trapped in a 2π orbital of a single oxygen bonded to a single silicon, which is then bonded to three oxygen atoms of the SiO_2 structure.

Although much work has been devoted to understanding radiation damage in silica fibers [3], it is not known if...
the presently available material will be suitable for use in future fusion devices, such as the International Thermonuclear Experimental Reactor (ITER), with their anticipated high neutron fluences. Thus, the objective of the present work is to measure radiation-induced optical attenuation in high-quality silica fibers following exposure to neutron fluences typical of those expected from ITER.

2. Experimental aspects

2.1. Test fibers and irradiation facility

Two types of optical fibers consisting of a pure fused-silica core with fluorine-doped (~4 mol%) cladding were obtained from Fiberguide Industries, Inc., [15] and tested in the as-received condition. Commercially referred to as anhydroguide™ (A) and superguide™ (S), these fibers contained <1 ppm and 600–800 ppm of OH, respectively, as stated by the manufacturer. The fibers were supplied with either nylon (used for low fluence exposures) or aluminum (used for high fluence exposures) jackets with typical core/cladding/jacket diameters of 200/220/280 μm. The low-OH fiber A was fabricated from Heraeus Suprasil F300 core material, and the high-OH fiber (S) was drawn from Heraeus F100 material. In addition to these commercially available fibers, we also investigated unirradiated fibers obtained from the Fiber Optics Research Center, General Physics Institute, Russian Academy of Sciences, Moscow, Russia. These fibers, labeled ‘low OH’ (<200 ppb) and ‘high OH’ (800–900 ppm) were obtained from D. Griscom, Naval Research Laboratory, and were investigated only in the unirradiated condition. Our intent was to compare the optical attenuation of these fibers with that of the commercially available specimens.

Several meters of fibers A and S were coiled and exposed to neutrons in high and low fluence regions of the Los Alamos Spallation Radiation Effects Facility (LASREF), which is located at the beam stop of the Los Alamos Meson Physics Facility. Spallation neutrons produced by interaction of incident 800 MeV protons and Cu nuclei yield a spectrum similar to that of a fast fission reactor but with the addition of a high energy tail [16]. The pulsed proton beam at LASREF is characterized by a 500 μs width and 6% duty factor. Fibers located in the ‘high fluence’ region were exposed to a neutron flux of $2 \times 10^{16}$ n/m²/s and γ flux of 200 Gy/s. The accumulated neutron fluence in this region was $1.1 \times 10^{23}$ n/m² and the γ fluence was $1.1 \times 10^9$ Gy. Nuclear heating caused the fiber temperature to increase to 270°C–300°C during irradiation. Fibers located in the ‘low fluence’ region at LASREF were subjected to a total neutron fluence of $1.4 \times 10^{21}$ n/m², and, due to extensive shielding, were not subjected to any significant γ fluence. No appreciable heating occurred during irradiation of these fibers. Following their irradiation in 1992, they were stored in ambient at room temperature until the optical measurements reported here were made in 1995.

2.2. Spectrophotometer

Optical absorption measurements were made at room temperature utilizing a Cary 5E spectrophotometer equipped with a fiber optic multiplexer. Attenuation was measured in the interval 250 to 2000 nm by exciting 0.5 m sections from the irradiated fibers, mounting fiber subminiature (FSMA) connectors to each end, polishing the
fiber using standard techniques [17], and connecting the two prepared fiber ends to the multiplexer. Because of the very low attenuation of the unirradiated fibers, we used the standard ‘cut-back method’ [18] with 1 and 50 m lengths for these measurements.

3. Results

3.1. Commercial fibers

Shown in Fig. 1 are the optical attenuation measurements for the low-OH fiber A following exposure to low- and high-fluence neutrons, along with attenuation of the unirradiated fiber for comparative purposes. In the pristine state we see that overall attenuation is less than 100 dB/km between 400 and 2000 nm. There is a relatively large attenuation (1000 dB/km) peak of unknown origin near 330 nm. Awazu and Kawazoe [19] have measured a similar band in silica at 3.8 eV (330 nm) and assigned it to the presence of Cl₂ molecules dissolved in the glass; by comparison we assume that our peak at 330 nm is also attributable to the presence of Cl₂ gas.

Exposure of fiber A to low-fluence neutrons induces broad attenuation below 400 nm (actual attenuation exceeds 10⁴ dB/km) and additional peaks at 630 and 1400 nm. The 630 nm peak (2 eV) is the well-known NBOHC [14] and the latter peak is attributable to an overtone mode of the OH molecule fundamental vibration [20]. High fluence exposure of fiber A induces attenuation exceeding 10⁴ dB/km for wavelengths below 700 nm, with additional band-like absorption above ~1200 nm.

Fig. 2 shows attenuation data for the high-OH content fiber S when exposed to both low- and high-fluence neutrons, and in the unirradiated state. The effect of the OH impurity is evident by the presence of peaks near 950, 1250 and 1400 nm, which represent the overtone and combination frequencies of the fundamental OH vibrational mode [20]. Notice that these peaks are essentially unaffected by neutron exposure, thus confirming their assignment to the OH molecule. Origin of a peak near 1900 nm is unknown, although possibly ascribable to the OH molecule. Radiation-induced band-like attenuation in this region overshadows the growth of the 1900 nm peak, thus making it difficult to definitively assign it to the OH molecule.

Interestingly, the magnitude of attenuation (~800 dB/km) for the NBOHC-peak in fiber S (Fig. 2) following low fluence irradiation is significantly less than that observed for fiber A (4000 dB/km) under similar conditions (Fig. 1). In fact the overall attenuation below 500 nm is less in the high-OH content fiber than in the low content one, suggesting that the OH impurity may be beneficial in radiation hardening in this spectral region.

3.2. Russian fibers

Optical attenuation of the unirradiated Russian fibers is shown in Fig. 3 and may be compared to the commercially-available pristine fiber data of Figs. 1 and 2. The low-OH fiber exhibits attenuation similar to that observed in fiber A, except for the absence of the strong peak near 350 nm. There is, however, an indication of enhanced
absorption near 400 nm, which may be related to the Cl₂ impurity previously discussed.

A comparison of attenuation data for the Russian high-OH fiber (Fig. 3) with commercially-available fiber S (Fig. 2) shows much similarity, the major exception being the presence of a peak near 700 nm in the former specimen. Although somewhat higher in wavelength than expected for NBOHC absorption (630 nm), it is likely that the 700 nm peak is associated with this particular defect.

Unfortunately, neither of the Russian fibers were available when fibers A and S were irradiated at LASREF, and, consequently, we have no comparison of the radiation sensitivity of these particular fibers. However, Griscom has exposed similar fibers to 60Co γ radiation (~ 10 Mgy) and found the low-OH content specimen to have a rather flat induced loss in the visible region at a level of 2000–2500 dB/km [21].

4. Discussion

Commercially-available fibers containing low OH impurity concentration exhibit good transmission in the region 250 to 2000 nm with typical attenuation being less than 100 dB/km. However, when subjected to high neutron fluences (1.1 × 10²³ n/m²), with accompanying γ component, these fibers are characterized by attenuation exceeding 10⁴ dB/km for wavelengths less than about 700 nm. For wavelengths above ~1200 nm there is band-like attenuation that increases with increasing wavelength, exceeding 5000 dB/km at 2000 nm. As shown in Fig. 1, the only region of relatively low optical attenuation (≤ 1000 dB/km) is from about 800 to 1400 nm. These results suggest that low-OH silica fibers are not presently suited for visible-spectrum diagnostic use in close proximity to a fusion reactor plasma such as ITER. Of course some amelioration of the radiation-induced damage might be achieved through thermal or optical annealing, but progress to date appears to have only minimal effects [22]. Recent experiments by Tighe et al. [1] show that silica fibers heated to 400°C during exposure show 20% less attenuation in the visible region than unheated fibers.

High-OH content fibers exposed to high neutron fluences are less suitable for close-proximity fusion plasma applications than their low OH counterparts because of the intrinsic OH absorption. Peaks at 950, 1250, 1400, and possibly 1900 nm (Fig. 2) are combination and overtone modes of the OH fundamental vibration and are not radiation induced. Nevertheless, they contribute appreciable attenuation that is not observed in the low-OH specimens. Both types of fibers exhibit radiation-induced absorption for wavelengths greater than ~1500 nm that is evidently not associated with OH content (see Figs. 1 and 2) and whose origin is presently unknown. A review of optical absorption data on fused silica by Griscom [3] also shows increased absorption in this spectral region.

Turning now to the optical attenuation data of both low- and high-OH content fibers that have been subjected to low fluence neutrons, we conclude that the magnitude of attenuation is sufficiently low to allow their use in reactor regions that are shielded from the plasma. An interesting feature of the data shown in Figs. 1 and 2 is the greater attenuation of the NBOHC (630 nm) in the low-OH fiber compared to the high content specimen. The radiation-induced attenuation is 4000 dB/km in fiber A at 630 nm, but only about 800 dB/km in fiber S. It is known [23] that OH-containing silica undergoes a radiolytic reaction

≡Si-OH → ≡Si-O - + H⁺  

producing NBOHCs. The incorporation of OH produces a
more radiation resistant bond than Si–O alone; therefore we expect more radiation-induced NBOHCs in fiber A than in fiber S, in agreement with experimental observation. These results suggest that in a low neutron fluence environment, low-OH content fibers are best for diagnostic use in the spectral region ~ 800 to 2000 nm, but high-OH content fibers are better suited for use in the visible portion of the spectrum.

The low-OH content Russian fibers exhibit relatively low attenuation over the spectral region 250 to 2000 nm, and may indeed represent an improvement over the currently-available fibers. Knowledge of their resistance to neutron damage at fluences expected from fusion plasmas is presently unavailable.

5. Conclusions

We have measured the optical attenuation of pristine and neutron-irradiated, low- and high-OH content silica fibers in the wavelength interval 250 to 2000 nm. The low-OH content fibers are devoid of the intrinsic absorption associated with combination and overtone modes of the OH molecule and exhibit minimal attenuation over the entire region investigated. Exposure to high fluence neutrons \((1.1 \times 10^{23} \text{ n/m}^2)\) induces strong absorption of the well-known defects (E'-type, peroxy radical and NBOHC), in addition to band-like absorption above ~ 1400 nm. Overall, the useful optical window is ~ 800 to 1400 nm and exists only in the low-OH specimen, making these fibers not particularly well suited for diagnostic use near the fusion plasma, and certainly not useable in the visible region.

Fibers exposed to low-fluence neutrons \((1.4 \times 10^{21} \text{ n/m}^2)\) suffer radiation-induced attenuation similar to that observed from high fluence exposure, but with the attenuation reduced in magnitude. For diagnostic use in the visible portion of the spectrum, the high-OH content fibers exhibit less attenuation than the low content counterpart and are preferred. Their intrinsic OH absorption at longer wavelengths, however, makes them less attractive than the low-OH fibers for diagnostic use above ~ 800 nm.

The data suggest that presently-available silica fibers can be used in plasma diagnostics, but the choice and suitability depends upon the spectral region of interest. Low-OH content fibers can be used for diagnostic purposes in the interval ~ 800 to 1400 nm if the exposure is to high-fluence neutrons. For low-fluence neutron exposures, the low-OH content fibers are best suited for use in the interval ~ 800 to 2000 nm, and the high-content fibers are the choice for the interval ~ 400 to 800 nm. Strong absorption of all fibers below ~ 400 nm will likely preclude their use in this spectral region unless other mechanisms (heating during irradiation, thermal or optical bleaching, etc.) can be found to mitigate the radiation-induced damage.

References

[15] Fiberguide Industries, Inc., 1 Bay Street, Stirling, NJ 07980, USA.