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Growth of optical transmission loss at 850 nm in silica core optical fibers during fission reactor irradiation

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Abstract

Pure, OH-doped and F-doped silica core optical fibers were irradiated in a fission reactor at 400 ± 10 K using an electric heater at a reactor power greater than 10 MW (20% of the full power). The temperature was not controlled well at the early stage of the reactor startup, when the temperature was about 320–340 K. The optical fibers were irradiated with a fast neutron ($E > 1$ MeV) flux of 3.2×10^{17} n/cm^{2}s and a gamma dose rate of 3×10^3 Gy/s for 527 h. Optical transmission loss at 850 nm was measured in situ during irradiation. A prompt increase in optical transmission loss was observed as irradiation started, which was probably due to dynamic irradiation effects caused by short-lived and transient defects and is probably recoverable when irradiation ceases. After the prompt increase in optical transmission loss, a so-called radiation hardening was observed in fibers containing OH. Radiation hardening was also observed in 900 ppm OH-doped fiber at the second startup. The optical transmission loss increased linearly with irradiation dose, denoted as the accumulated loss, which we believe is due to irradiation-induced long-lived defects. Accumulated loss dominates radiation-induced optical transmission loss in a fission reactor irradiation. © 1998 Elsevier Science B.V.

1. Introduction

Optical fibers are thought to be vulnerable to heavy irradiation, but only a few attempts [1–3] have been made to explore their usage in environments such as in a nuclear fission reactor core. Recent advances in nuclear fusion reactor development have highlighted the importance of optical measurements in heavy irradiation environments near a burning plasma [4]. Optical fibers can play important roles if they can be used under heavy irradiation. Optical fibers can also be used as signal transportation media in a high magnetic field in fusion reactors, where conventional electrical signal transportation systems will have strong electromagnetic disturbance.

Up to now, radiation effects in optical fibers have been studied mainly using X-ray and gamma-ray sources with concern only for electronic excitation effects. Maximum dose rates for these studies are in the range of 100 Gy/s and total maximum doses are limited to about 10^8 Gy. Under these conditions, extensive studies have been carried out to identify which defects, such as the so-called E′-center, non-bridging oxygen hole center and peroxy radicals, are responsible for optical absorption [5–7].

Increased interest in fusion research is resulting in more studies of heavy radiation effects in optical fibers that consider not only electronic excitation but also atomic displacement. In these studies, dose rates are up to 10^4 Gy/s for electronic excitation and 10^{19} n/m^{2}s ($\sim 10^{-6}$ displacements per atom) for atomic displacement. Expected total doses are as high as 10^{25} n/m^2 (a few dpa) for fast neutron fluence and 10^{10} Gy for electronic excitation after a few years operation of an experimental fusion reactor [4]. Such conditions are likely in a high-flux fission reactor [1,3,6]. In a high-flux fission reactor, a large number of lattice defects are introduced by displacive irradiation and through radiolysis. Interaction among defects will be strong and complicated. Mechanisms of introduction of

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optical absorption will be different from those in the low dose rate, purely electronic excitation irradiation.

The development of radiation-resistant optical fibers has depended mainly on testing them in pure electronic excitation irradiation fields [9,10]. Some optical fibers show better radiation resistance and some of them show radiation hardening [8]. Radiation hardening is the phenomenon where radiation-induced optical transmission loss recovers in the course of subsequent irradiation. Irradiation studies during ^{60}Co gamma irradiation showed that fused silica core optical fibers with appropriate concentrations of OH (oxyhydrate) have a better radiation resistance after radiation hardening [9]. Also, there are reports that F-doped silica core optical fibers have good radiation resistance under gamma ray irradiation [9,10].

Radiation hardening is an interesting phenomenon from scientific and engineering points of view. One popular theory on how this process works is that radiation breaks bonds to produce free radicals, such as Si and O and that these radicals result in increased optical absorption. Impurities within the material then cure these broken bonds. For example, H atoms may act as curing impurities by bonding with O and Si radicals to form OH and SiH. The optical absorption of these resulting compounds will be localized in very narrow wavelength regions. Kakuta et al. [9] showed that a hydrogen treatment, which apparently dopes hydrogen into fused silica, improved the radiation resistance of fused silica core optical fibers. Griscom [8,10,11] proposed that hydrogen atoms or radicals, which are generated during radiation-induced decomposition of plastic jackets surrounding the fibers, is responsible for the observed radiation hardening during gamma-ray irradiation. Griscom also reported that radiation hardening was not observed during a fission reactor irradiation [10]. In general, radiation effects in heavy irradiation fields, where both electronic excitation and atomic displacement effects exist, will be quali tatively and quantitatively different from radiation effects during purely electronic excitation, such as in ^{60}Co gamma-ray irradiation.

In this study, radiation-resistant silica core optical fibers were irradiated and radiation-induced optical transmission loss was measured at 850 nm in a Japan Materials Testing Reactor (JMTR) in the Oarai Research Establishment of the Japan Atomic Energy Research Institute. Previous studies [1,5] showed that fission reactor irradiation introduced large optical transmission loss in the wavelengths shorter than 700 nm at an early stage of irradiation. Some strong optical absorption peaks, such as a peak at 600–650 nm whose mechanism of optical absorption is not clearly understood yet, were reported in this wavelength range. Also, there are several strong radioluminescence peaks in the wavelength range shorter than 700 nm, such as at 350 and 450 nm [1,12–14] and there is a so-called Cerenkov radiation whose strength is proportional to wavelength^{-3} [1,13,14]. This radiation-induced luminescence will disturb measurements of optical transmission loss. Thus, optical transmission behavior during irradiation is complicated in fused silica core optical fibers at wavelengths below 700 nm. Radiation-induced optical transmission loss is small and nearly wavelength-independent between 700–1800 nm [1]. The dominant optical absorption peaks in this range are those of OH absorption and they will not have a strong influence on optical transmission at 850 nm. Thus, radiation-induced defects, which absorb or scatter photons nearly uniformly in the wavelength range longer than 700 nm, can be studied by measuring radiation-induced optical loss at 850 nm. Infrared spectroscopy and signal transportation can also be carried out in this wavelength region.

2. Experimental procedures

Sample names and their compositions are listed in Table 1. The core and clad diameters of these fibers are 200 and 250 μm, respectively. These samples were placed into an instrumented irradiation rig and were inserted in a JMTR core (Fig. 1). The total length of the fibers was about 46 m, with about 0.5 m exposed to the reactor core radiation. The measuring system is shown schematically in Fig. 2. The optical transmission loss of the fibers is in the range of 10–50 dB/km before irradiation, which corresponds to about 0.5–2.5 dB in this measuring system. The numerous optical connections shown in Fig. 2 resulted in an additional loss of about 10–20 dB.

The standard fiber (H3 fiber in Fig. 2) was found to have good resistance to gamma-ray irradiation at room temperature [9]. A hydrogen treatment was carried out to

Table 1
Characteristics of fused silica core optical fibers

Fiber name	Treatment	Dopant	OH content (ppm)	Jacket
F-doped, F		florine, 0.35%	120	plastics
Standard, H3			900	plastics
H1	hydrogen treated, 24 h		900	plastics
H2	hydrogen treated, 120 h		900	plastics
OH-doped, C1, C2			18000	carbon

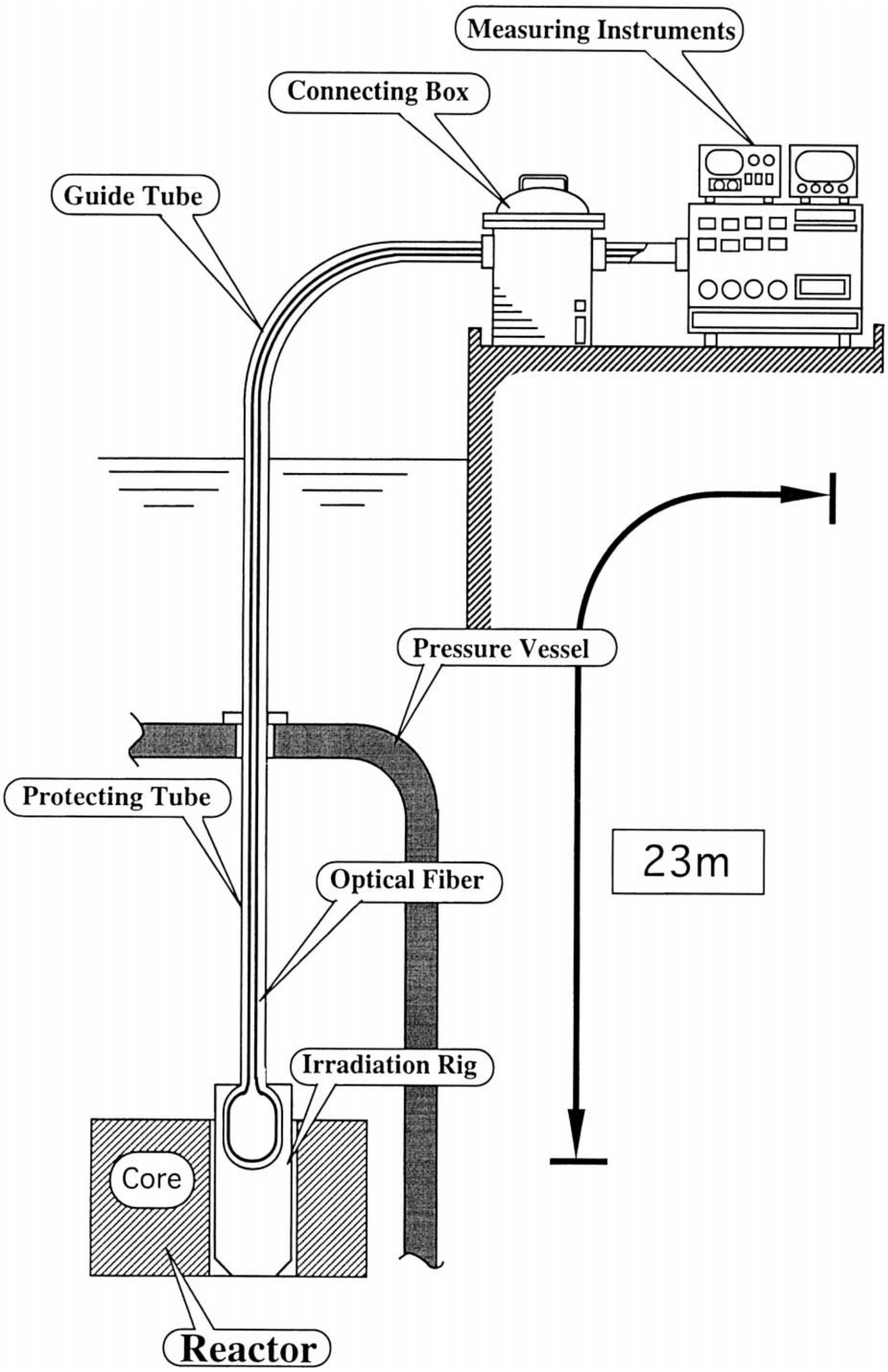

Fig. 1. Schematic view of reactor setup.

improve the radiation resistance of the standard fiber [9]. The fibers were heat treated at 430 K in a 1 atm hydrogen environment for 24 h (H1 fiber) and for 120 h (H2 fiber). A ^{60}Co gamma-ray irradiation at room temperature and at 470 K showed that the hydrogen treatment improved radiation resistance substantially [9]. H1, H2 and H3 fibers had a plastic jacket. Two identical fibers doped with 18000 ppm OH and covered with a carbon jacket (C1 and C2 fibers in Fig. 2) and a F-doped fiber with a plastic jacket (F in Fig. 2) were also irradiated. The OH-doped fiber showed very low optical transmission loss (9.6 dB/km) at 850 nm before irradiation. Carbon-jacketed samples were included in the irradiation to test the theory that H generated by the decomposition of plastic jackets may effect radiation response in fibers. The F-doped optical fibers were reported to perform well under gamma-ray irradiation [9,10].

The irradiation was carried out for 527 h with an intermediate stop for 14 days. The neutron fluxes and gamma-dose rate were 3.2×10^{17} n/cm^{2}s for fast neutrons ($E > 1$ MeV), 1.3×10^{18} n/cm^{2}s for thermal neutrons ($E < 0.683$ eV) and 3×10^3 Gy/s, respectively. During the reactor stop, the fibers were exposed to a residual gamma ray of a few to a few tens of Gy/s at 290 K. Irradiation temperature was controlled at 400 ± 10 K using an electric heater at a reactor power greater than 10 MW (20% of the full power). The temperature was not controlled well at the early stage of the reactor startup, when the temperature was about 320–340 K. The irradiation environment was 1.3 atm helium. Total fluence was 6.0×10^{23} and 2.5×10^{24} n/m^2 for fast and thermal neutrons, respectively. The total electronic excitation dose was 5.7×10^9 Gy.

The optical transmission loss of the C1 fiber was measured continuously, and the optical transmission loss of other fibers was measured occasionally with manual connections at couplers (Fig. 2). The optical transmission spectrum was also measured occasionally in the range of 350–1850 nm. Further details of the measuring setup is reported elsewhere [1,9].

3. Results and discussion

As previously reported [1,3], strong absorption grew at the beginning of the irradiations at wavelengths shorter than 700 nm. Above 700 nm, radiation-induced optical absorption is weak and nearly independent of the wavelength [1,3]. Some results indicate an increase in OH absorption during irradiation, whereas others do not. During the irradiation, systematic trends in the strength of OH absorption were not observed.

Fig. 3 shows data for optical transmission loss during an entire irradiation period as a function of 50 MW equivalent irradiation time for the standard fiber, 18 000 ppm OH-doped fiber and a F-doped fiber. Two 18 000 ppm OH-doped fibers with carbon jackets behaved the same. Also, the hydrogen-treated standard fibers (H1 and H2) behaved nearly identically to the standard fiber (H3). The hydrogen treatment [9], which was found to be effective in improving gamma-ray radiation resistance up to 470 K, did not improve radiation response in a fission reactor irradiation. Because of the similarities between the results of the standard fibers, hereafter only the results of the standard fiber H3 will be discussed.

The OH-doped fiber C1, which had very low optical transmission loss before irradiation, showed a large increase in loss at the beginning of the irradiation. It then showed a sharp decrease in optical transmission loss, corresponding with radiation hardening. Changes in the

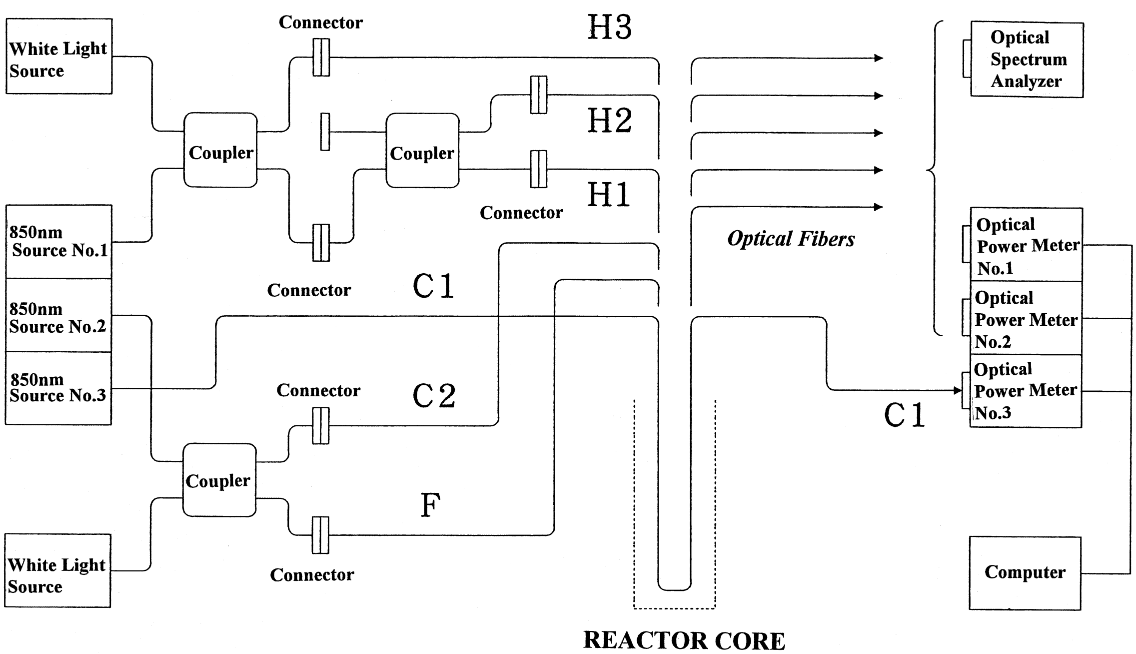

Fig. 2. Schematic diagram of optical measuring system.

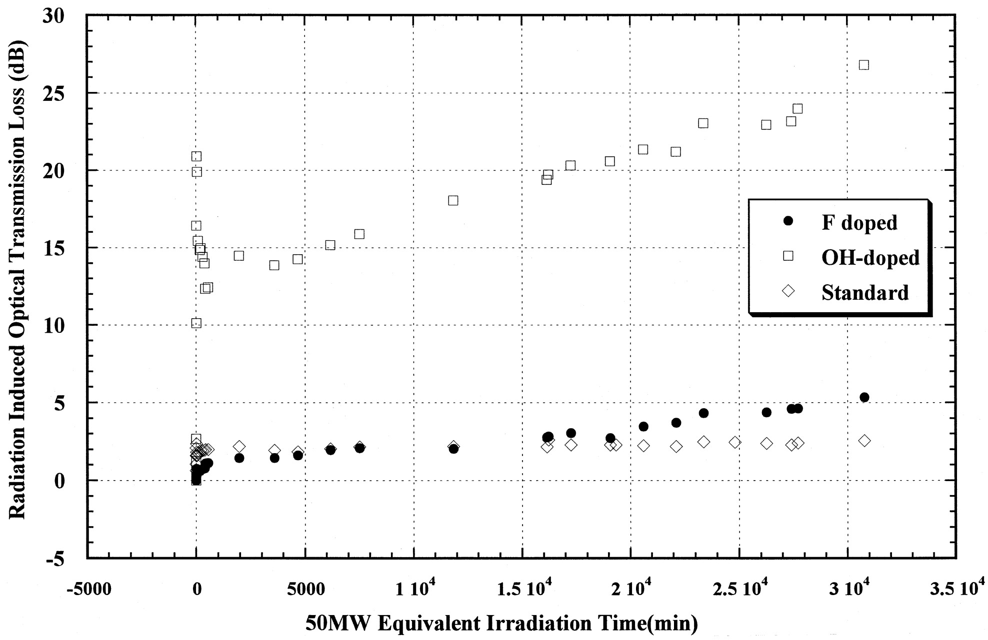

Fig. 3. Radiation-induced optical transmission loss as a function of 50 MW equivalent irradiation time.

optical transmission loss of the OH-doped fiber in the initial irradiation period are shown in Fig. 4. A large increase in loss (22 dB) was followed by sharp decrease in loss (6 dB). Optical transmission continued to decrease up to about 1000 min, or a fast neutron fluence of 1.9×10^{22} n/m^2 and an electronic excitation dose of 1.8×10^8 Gy. The standard fiber H3, which contains 900 ppm OH, showed less radiation hardening, with a decrease of about 0.8 dB (Fig. 5). The F-doped fiber F, whose OH content is low, did not show radiation hardening (Fig. 6). We note

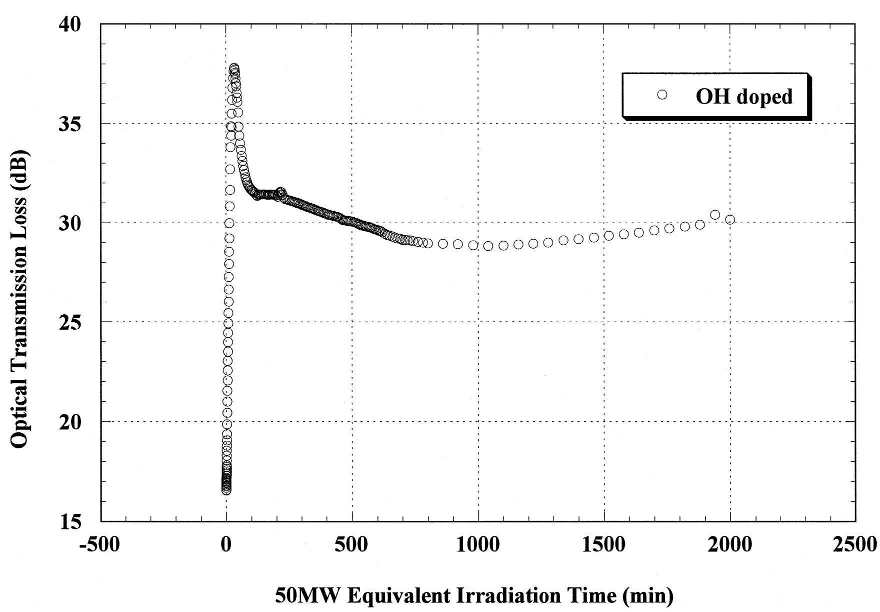

Fig. 4. Optical transmission loss in the OH-doped fiber at the beginning of the first irradiation.

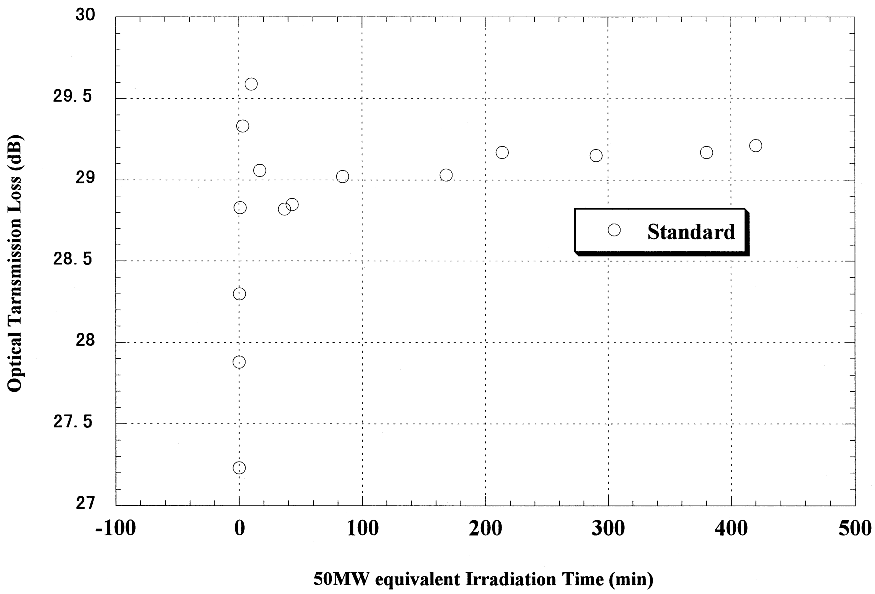

Fig. 5. Optical transmission loss in the standard fiber at beginning of the first irradiation.

that all of the fibers in this study, including the F-doped fiber, showed radiation hardening during ^{60}Co gamma-ray irradiation [9,10].

Figs. 4–6 show that the optical transmission loss had an abrupt increase at the start of irradiation. After the initial changes, every fiber showed a steady increase in the optical transmission loss (Fig. 3). The loss increase is linearly dependent on the irradiation time, as shown for OH-doped and F-doped fibers in Fig. 3. The standard fiber, which showed the smallest rate of increase, also showed a linear dependence on irradiation time (Fig. 7). Although the F-doped fiber had the smallest initial loss increase, its optical transmission loss exceeded that of the standard fiber due to a larger linear increase rate (Fig. 3).

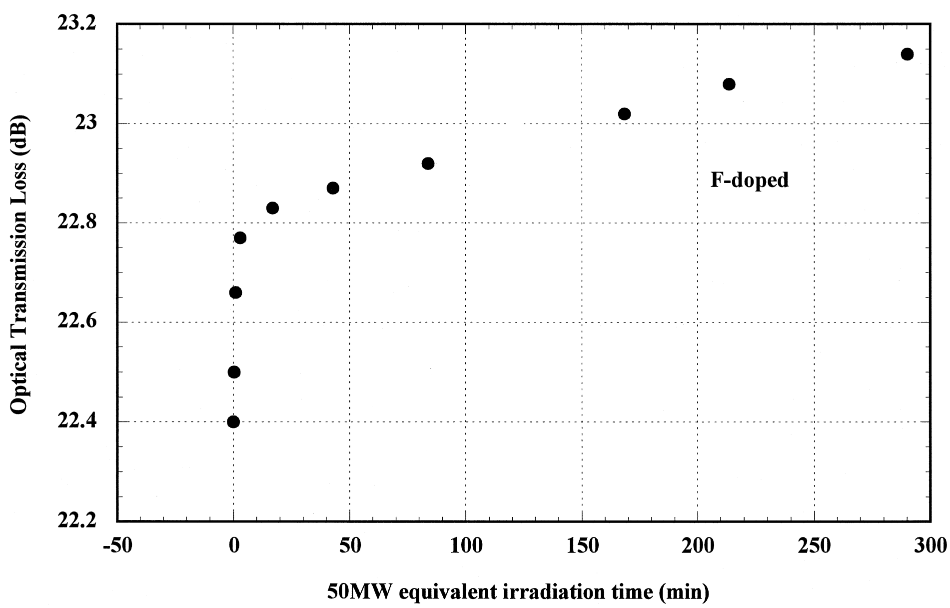

Fig. 6. Optical transmission loss in the F-doped fiber at the beginning of first irradiation.

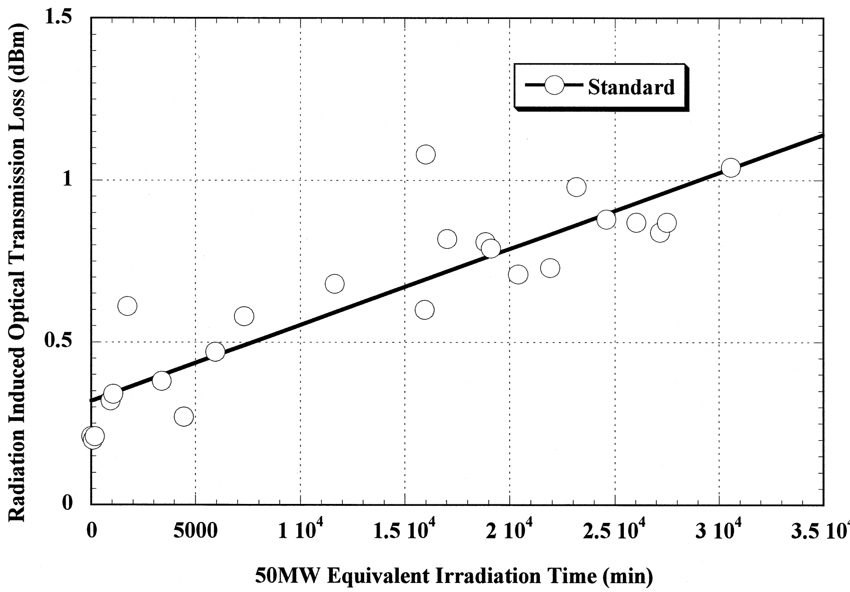

Fig. 7. Linear increase in the optical transmission loss in the standard fiber as a function of irradiation time.

Fig. 8 shows the change in the optical transmission loss for the standard fiber when the reactor had an intermediate stop. The sample recovered slightly, by about 1.6 dB, the same as the initial 1.6 dB increase shown in Fig. 5 and neglecting radiation hardening. The standard fiber also showed radiation hardening during the second startup (Fig. 8), though the extent of the hardening (0.3 dB) is smaller than that in the first startup (0.8 dB). The OH-doped fiber, which showed significant radiation hardening, showed no radiation hardening at the second startup (Fig. 9). The recovery of the OH-doped fiber during the reactor stop was about 2.6 dB, far smaller than the initial increase of

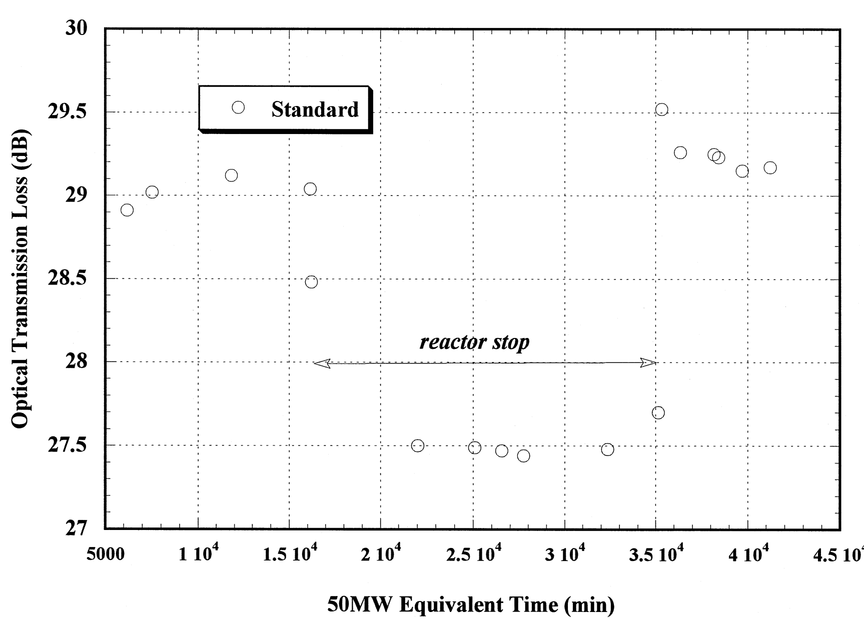

Fig. 8. Change in the optical transmission loss in the standard fiber measured during the intermediate reactor stop.

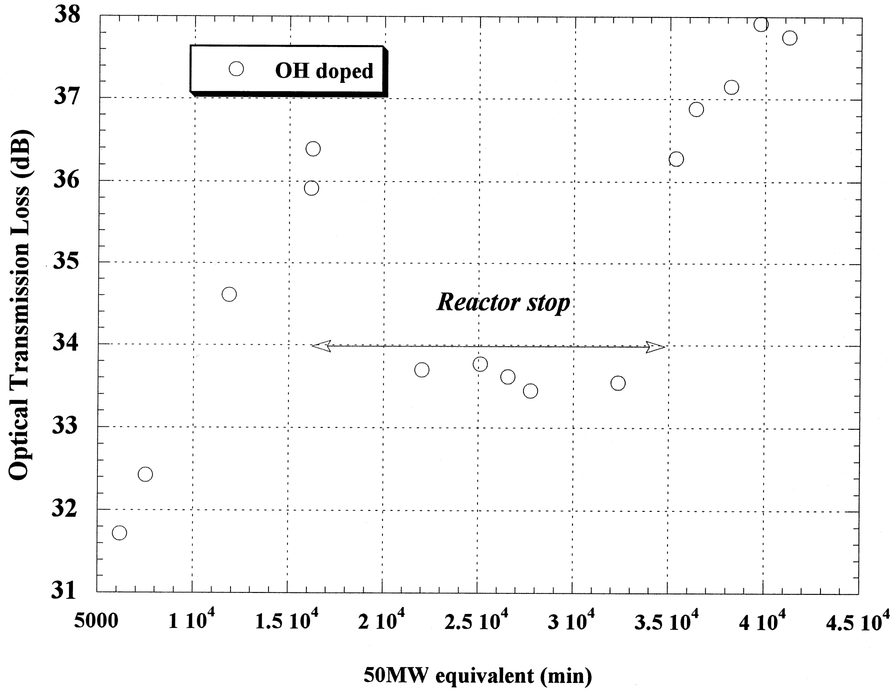

Fig. 9. Change in the optical transmission loss in the OH-doped fiber during the intermediate reactor stop.

about 10 dB. The F-doped fiber did not harden at the second startup nor in the first startup (Fig. 10). Its recovery during the reactor stop was about 0.4 dB, comparable to the initial increase of 0.4 dB.

Fig. 11 shows schematically the radiation-induced optical transmission loss as a function of irradiation time. Loss can be described as a combination of the initial loss, $L_{initial}$, the radiation hardening loss, $L_{hardening}$, the loss that recovers when the irradiation stops, $L_{dynamic}$, and the accumulated loss, L_{accm}. Our results show that $L_{initial}$ and $L_{dynamic}$ have the same magnitude in the standard and the F-doped fibers and that $L_{initial}$ and the $L_{dynamic}$ are optical

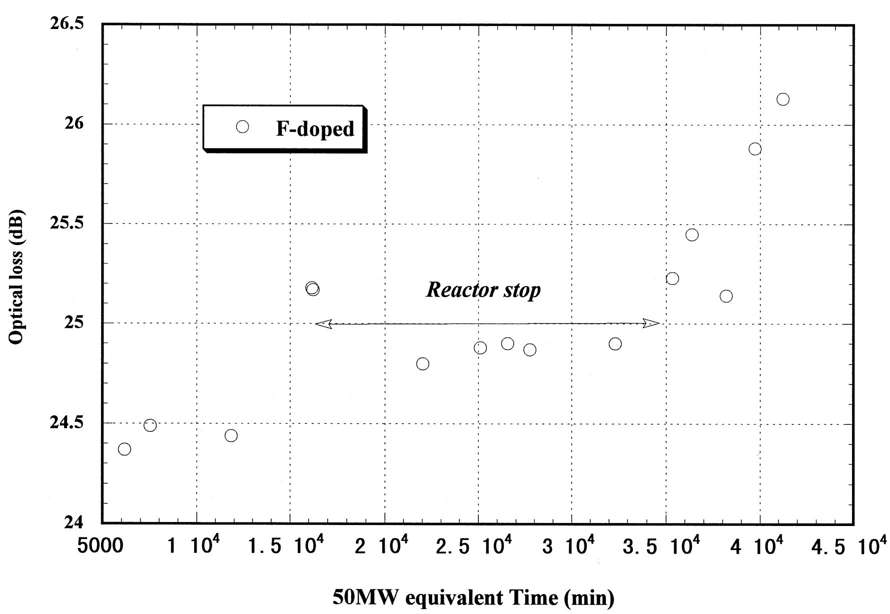

Fig. 10. Change in the optical transmission loss in the F-doped fiber during the intermediate reactor stop.

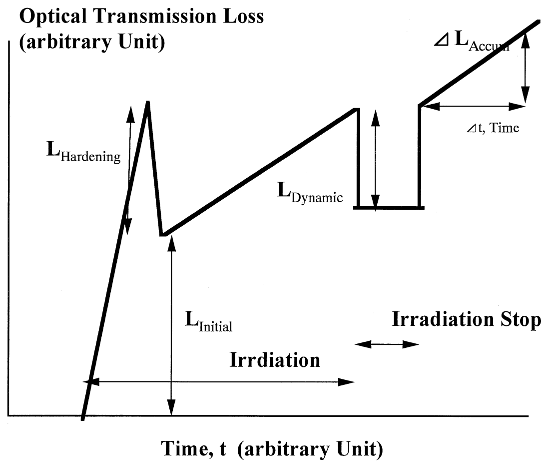

Fig. 11. Schematic diagram of radiation-induced optical transmission loss during fission reactor irradiation.

transmission losses caused by short-lived and transient defects. In the meantime, some permanent or long-lived defects result in L_{accm}. The concentration of the defects responsible for L_{accm} will increase linearly with the dose.

Our results imply that fluorine doping decreases the concentration of transient defects, which are responsible for $L_{initial}$ and the $L_{dynamic}$. The presence of OH may increase the concentration of transient defects, and radiation hardening implies an abrupt decrease in the concentration of these defects. As described above, hydrogen atoms introduced by radiation-induced decomposition of plastic jackets may help the transient defects to vanish. The equilibrium concentration of the transient defects will then decrease with an increase in the hydrogen concentration. However, the OH-doped fiber with carbon jacket showed radiation hardening and the F-doped fiber with plastic jacket did not.

The hydrogen treatment [9], which was found to effectively improve radiation resistance and enhance radiation hardening under gamma-ray irradiation, was found ineffective during fission reactor irradiation. We interpret that this treatment introduces hydrogen atoms into the fused silica matrix. The hydrogen treatment was confirmed to be effective up to 470 K during ^{60}Co gamma-ray irradiation [9,10]. These results imply that the hydrogen may not play a role in radiation hardening effects during fission reactor irradiation.

Permanent defects, which are introduced directly by atomic displacement in a fission reactor irradiation, may annihilate the transient defects responsible for the dynamic loss. Introduction of the permanent defects will then decrease the dynamic loss, resulting in radiation hardening in the early stages of irradiation. The permanent defects are responsible for the accumulated optical transmission loss. In the second startup, a substantial amount of the permanent defects would already exist in the OH-doped fiber as it has a large accumulated loss. Thus, radiation hardening would not occur in the OH-doped fiber in the second startup. The concentration of permanent defects would be very low in the standard fiber as it had a very small accumulated loss. Thus, radiation hardening was observed in the standard fiber even in the second startup. After heavy reactor irradiation with a fast neutron fluence higher than 10^{24} n/m^2, the recovery of radiation-induced optical transmission loss is negligible [15]. These results may also support the speculation that the increase in concentration of the permanent defects decreases the concentration of the transient defects.

Radiation hardening was more clearly observed during gamma-ray irradiation, where the atomic displacement due to Compton scattering is negligible and the accumulated loss is very small. Therefore, radiation hardening is not due to the introduction of permanent defects by the irradiation in this case. A sharp peak in the optical transmission loss due to radiation hardening took place at a small irradiation dose, in the range of 10^4 Gy in the gamma-ray irradiation [9] and the radiation hardening observed in the present fission reactor took place at about 10^6 Gy. Mechanisms responsible for radiation hardening are probably different in the electronic excitation-dominant irradiation and in the irradiation associated with atomic displacement.

An optimum doping concentration of OH may decrease the concentration of the permanent defects responsible for the accumulated loss. Our results show that a large concentration of OH increases the amount of permanent defects. The doping level of 900 ppm was selected as it performed

Table 2
Summary of radiation-induced optical transmission loss in fibers

Fiber	Linear loss (dB/m)		Initial loss (dB/m)	Hardening (dB/m)	Dynamic loss (dB/m)
	per fast neutron fluence (10^{24} n/m^2)	per electronic excitation (10^{10} Gy)			
F-doped	11.7	12.3	0.8		0.8
Standard	2.33	2.46	3.2	1.8	3.2
OH-doped	43.33	45.62	26	18	6

best during gamma-ray irradiation [9]. Therefore, the optimum concentration may be around 900 ppm even for a reactor irradiation. However, as described above, there is a distinct difference between radiation effects during gamma ray and fission reactor irradiation. Systematic studies are needed to understand the mechanisms responsible for radiation-induced optical transmission loss and to optimize the properties of fused silica core optical fibers for irradiations associated with atomic displacement.

Table 2 summarizes the magnitude of radiation-induced optical transmission loss in the various samples. It is clear that accumulated loss is dominant for fission reactor irradiations. In this case, fluorine doping will not be effective for improving radiation resistance. However, a promising fiber for fission reactor irradiations is the standard fiber, whose radiation-induced optical transmission loss is on the order of 10 dB/m for a fast neutron irradiation up to 10^{25} n/m^2 and for an electronic excitation dose of 10^{11} Gy.

4. Conclusion

Fused silica core optical fibers were irradiated at about 400 K in a fission reactor core and radiation-induced optical transmission loss was measured in situ at 850 nm. Radiation-induced optical transmission loss is composed of the dynamic loss, $L_{dynamic}$, and the accumulated loss, L_{accum}. Fibers doped with OH showed radiation hardening at the beginning of the irradiation. A mechanism for the observed radiation hardening was proposed and is different from the mechanism for radiation hardening during irradiation dominated by electronic excitation, such as gamma-ray irradiation. The accumulated loss is the dominant optical transmission loss in a fission reactor irradiation and doping with 900 ppm OH suppresses the growth of the accumulated loss.

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