Irradiation Behavior of developed radiaiton resistance optical-fibers and observed optical radiation from their Si0_2 cores under reactor irradiation

著者	SHIKAMA Tatsuo, NARUI Minoru, KAKUTA Tsunemi, SAGAWA Tsutomu, KAYANO Hideo, SANADA Kazuo, SHAMOTO Naoki, URAMOTO Toshimasa
journal or publication title	Science reports of the Research Institutes, Tohoku University. Ser. A, Physics, chemistry and metallurgy
volume	40
number	1
page range	147-152
year	1994-09-16
URL	http://hdl.handle.net/10097/28515

Sci. Rep. RITU A40 (1994) pp. 147-152
No. 1, July 1994

Irradiation Behavior of developed radiaiton resistance optical-fibers and observed optical radiation from their SiO_2 cores under reactor irradiation*

Tatsuo SHIKAMA[1)], Minoru NARUI[1)], Tsunemi KAKUTA[2)], Tsutomu SAGAWA[3)], Hideo KAYANO[1)], Kazuo SANADA[4)], Naoki SHAMOTO[4)], and Toshimasa URAMOTO[3)]
[1)] *the Oarai Branch, Institute for Materials Research, Tohoku University, Oarai, Ibarakiken, 311-13 Japan*
[2)] *Tokai Research Establishment, Japan Atomic Energy Research Institute, Tokai, Ibarakiken, 319-11 Japan*
[3)] *Oarai Research Establishment, Japan Atomic Energy Research Institute, Oarai, Ibarakiken, 311-13 Japan*
[4)] *The Fujikura Co. Ltd., Sakura, Chibaken, 285 Japan*
(Received February 28, 1994)

Two kinds of optical fibers were irradiated in a fission reactor, JMTR(Japan Materials Testing Reactor), up to a $1.55x10^{19}n/cm^2$ fast neutron fluence and a $3.3x10^9Gy$ ionizing dose at 370K. Optical transmission spectra were measured in the wavelength range of 450-1750nm, in-situ. Growth of strong optical absorption bands were observed in the range of wavelength shorter than 750nm. In the meantime, the fibers showed good radiation-resistance in the range of wavelength longer than 750nm. Optical radiations were observed from SiO_2 optical fibers under irradiation. A major part of the observed optical radiations is thought to be composed of broad optical radiation in the whole wavelength range studied in the present experiment. This broad optical radiation will be generated by the process of so-called Cerenkov radiation. Also, a sharp optical radiation peak was found at 1270nm on a F-doped fiber. This peak is thought to relate with doped Fluorine ions and ionizing gamma-ray irradiation.
KEY WORDS: optical fiber, SiO_2, reactor-irradiation, in-situ measurements, optical radiation.

I. Introduction

Ceramic materials are anticipated to play crucial roles in developing nuclear fusion reactors, where ceramic materials will be used under heavy irradiation environments for substantial periods for the first time[1,2,3)] in a nuclear eingineering. Extensive studies have been carried out for more than several decades on radiation effects of ceramics concerning fundamental aspects[4)], however, we seriously need understandings of effects of neutron-associated high flux and high fluence irradiations on ceramics for their fusion-reactor applications[5,6)]. Up to now, heavy irradiation effects on ceramics have been studied mainly by the post irradiation examinations(PIE), although it is understood that transient and dynamic irradiation effects will be important in ceramics.

Dynamic irradiation effects on ceramics in irradiation environments relevant to fusion reactor environments have been studied recently but mainly on electrical properties such as conductivity and dielectric loss[6)]. Recent experimental results strongly indicate that studies of optical properties will be very useful even for complicated irradiation phenomena observed in fusion-relevant environments[7)]. In general, the optical signals evoked by an irradiation is weak, and a highly intense irradiation source and a high-sensitive and sophisticated instruments are needed. So, the optical measurements have been carried out mainly using electron and ion beams. The optical measurements under irradiation in a fission reactor were scarce[8,9)] except for in a pulse-mode small experimental reactor[10)] and a beam neutron source[11)]. Griscom[11)] tried to observe the optical radiation from SiO_2 under the irradiation in a beam neutron source but he could not do it. He reported that the intensity of optical radiation would be less than -70dBm under the fast neutron irradiation of 10^{12} n/cm^2s[11)].

A high-power nuclear fission reactor is a powerful irradiation tool. It has a very high ionizing dose rate and a relatively high and spatially uniform displacement rate with bulk specimens. It is expected that optical radiation from insulators could be detected under such high intensity irradiation. No one has ever carried out in-situ type optical experiments successfuly in a high power reactor, although the fission reactor irradiaiton is crucial for appropriate studies of radiation damage in ceramics. In this study, we are trying to develop a technique which will enable us to see directly specimens irradiated in a reactor core. As the first step, we tested the durability of optical fibers in a reactor core.

This paper will describe measured performance of optical fibers in a reactor core, especially optical absorption generated by the reactor irradiation. In the course of irradiation experiment, we attempted to observe optical radiation from SiO_2 of optical fiber cores being irradiated in a fission reactor core. The experimental configurations were arranged to minimize a background optical noise and two kinds of weak optical radiations could be detected under the in-reactor irradiation. This paper will also describe the preliminarily observed optical radiations and will discuss their origins.

II. Experimental Procedures

Two kinds of SiO_2-base optical fibers, details of which are tabulated in Table 1, are irradiated in JMTR(Japan Materials Testing Reactor) fission reactor of Oarai Research Establishment of Japan Atomic Energy Research Institute. Cores of the OH-fiber is made of OH(Hydroxyl radical)-doped SIO_2 and that of F-fiber is made of F(Fluorine) doped SiO_2. Clads of the both fibers are made of F-doped

*IMR, Report No. 1977

Table 1 Details of optical fibers.

	OH-fiber		F-fiber	
	core	clad	core	clad
diameter (μm)	100	125	200	250
material	SiO_2 (OH 800ppm)	SiO_2 (F-doped; Δ^-=1.0%)	SiO_2 (F-doped; Δ^-=0.35%)	SiO_2 (F-doped; Δ^-=1.0%)

Δ^- is a decrease of reflecting coeficient due to doping

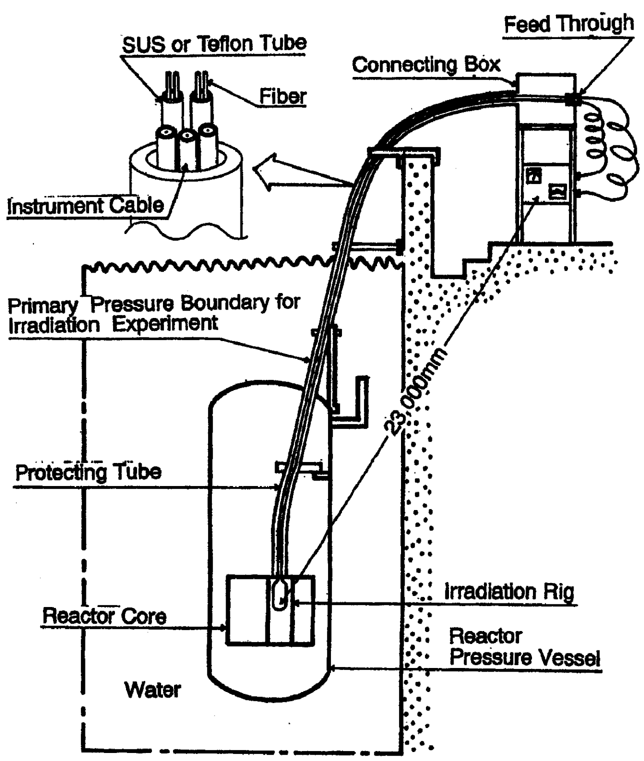

Figure1 Schematic configuration of insertion of optical fibers into a reactor core.

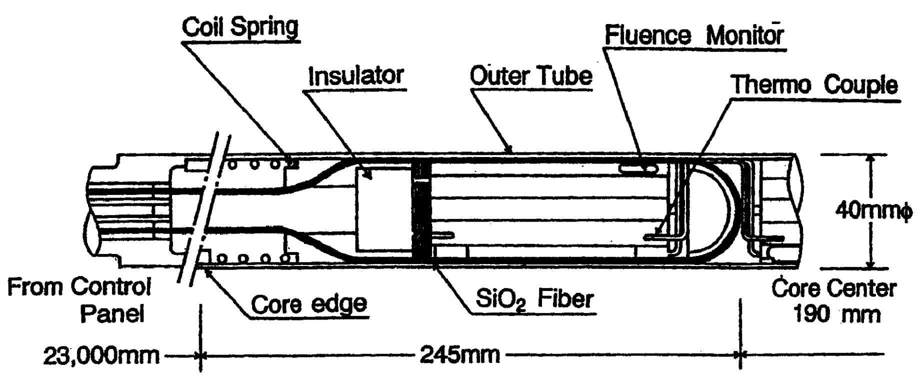

Figure 2 Structure of temperature controlled rig and accommodation of optical fibers in it

SiO_2. The optical fibsers were developed to be radiation-resistant to Co^{60} gamma-ray irradiation at room temperature.

Total length of the fiber was about 50m, and about 1m was exposed to a heavy irradiation in the reactor core. The F-doped fiber(F-fiber) was irradiated, because recent experimental results[11,12] strongly suggested that F-doping would improve the radiation-resistance of optical fibers in the ultra-violet wavelength region. However, we obtained similar results on the OH- and F- optical fibers. Namely, the present experiments did not give us results which showed improving effects of F-doping. Hereafter, we will mainly describe results obtained on the OH-fiber. When we mention results obtained on the F-fiber, we will notice it.

Accommodation of the optical fibers in JMTR is schematically depicted in Fig.1. No optical connections were used at any pressure boundaries and safety boundaries of the reactor. The fibers were sheathed with SUS- or teflon-made fine tubes along their whole length. Measured optical loss through the fibers before insertion into the reactor core was evaluated to be 0.1-0.2dB, which confirmed that there was little unexpected optical loss along the 50m long optical fibers. The irradiation environment was 1.3Pa helium.

One end of each fiber was connected to a xenon-lamp of white optical source and a 850nm light source. The other end was connected to the optical spectrum analyzer, Ando Electric Co. Ltd made AQ-6312A and an optical power meter. The measuring wavelength range is 450-1750nm. The magnitude of optical leakage in the present experimental system was measured to be less than -90dBm in the whole wavelength range and below the sensitivity of AQ-6312A.

Temperature of the optical fibers was controlled by the specially designed irradiation rig[13] shown in Fig. 2 to be 370K during the experiment, regardless of change of the reactor power. Neutron fluxes were $8.82x10^{12}$ and $9.85x10^{12}$ n/cm^2s for the fast(E>1MeV) and the thermal(E<0.682eV) neutrons, respectively, and a gamma-ray dose rate is $1.9x10^3$Gy/s, at the reactor full power of 50MWth. Concerning the neutron fluxes, they change with the reactor power linearly. In the meantime, there was a residual gamma-ray dose rate of about a few Gy/s before the reactor-start-up. At the reactor-shut down, the gamma-ray dose rate decreased linearly with the reactor power down to about a few tens Gy/s at the complete reactor-shut-down. Then, the gamma-ray dose rate decreased further exponentially to several Gy/s for 24-36 hours and then decreased slowly down to a few Gy/s for several days.[14]

During the course of 22 days reactor irradiation, we noticed some evidence of detection of optical radiations from the SiO2 fibers from the beginning. However, sensitive detection of optical radiations from the irradiated optical fibers were attempted only after the fibers were irradiated for about 16.7rfpd(reactor full power days).

III. Experimental Results and Discussions

When the optical fibers were inserted into the shut-down reactor core, we did not observe any increase of optical transmission loss, although there was

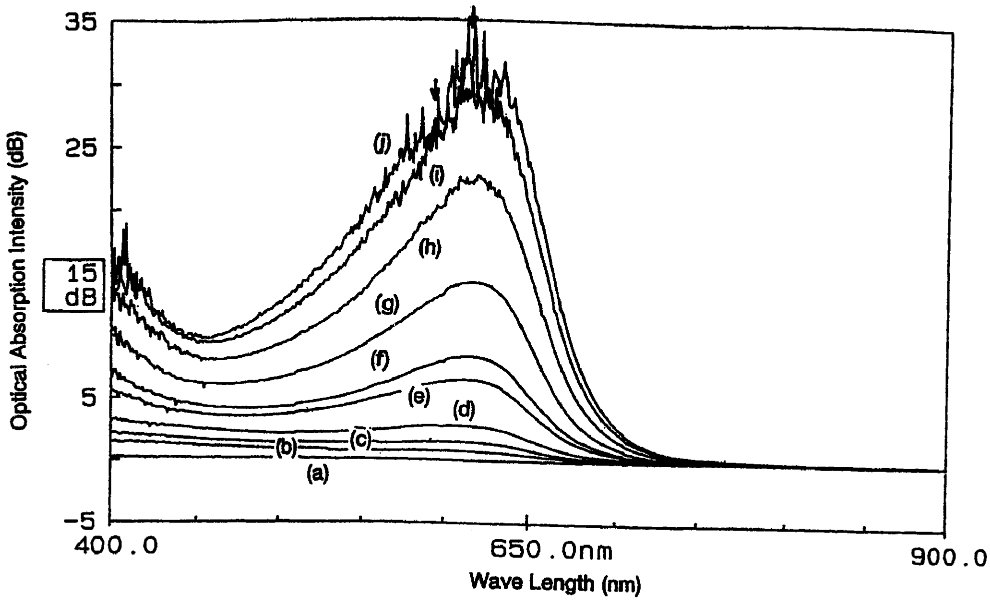

Figure 3 Transmission optical loss through OH-fiber caused by the irradiation during start-up of the reactor.

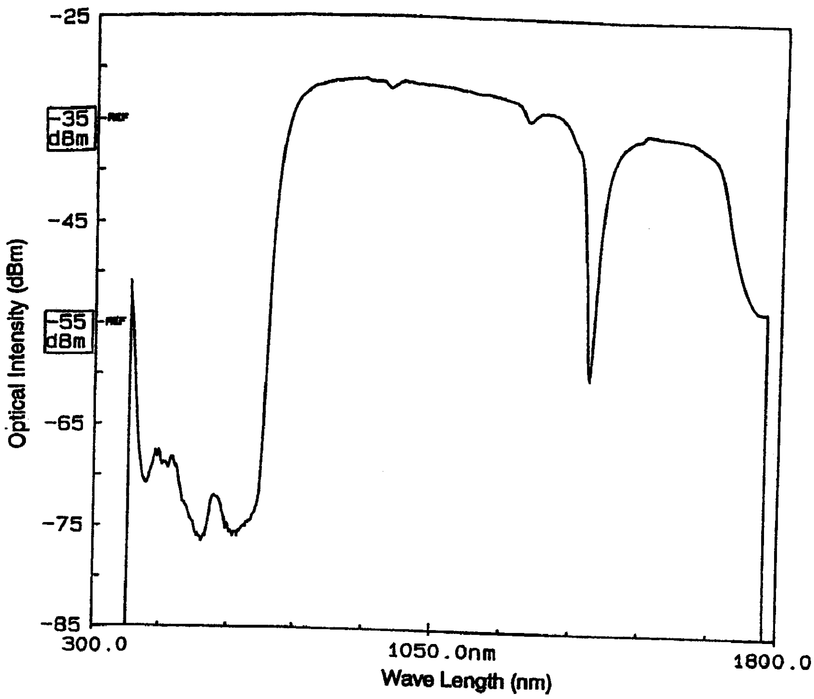

Figure 4 Transmission optical spectrum through the fiber at the end of 20.2rfpd irradiation. The reactor power was 50MW.

a ionizing radiation field of a few Gy/s in the shut-down reactor core. At the initial start-up of the reactor, the transmission optical loss increased with the reactor power as shown in Fig. 3. However, it should be pointed out that the optical transmission loss was not affected substantially in the wavelength range larger than 750nm. Even after 488h of irradiation, optical transmission was not affected substantially at wavelengths less than 750nm as shown in Fig. 4.

In the range of wavelength shorter than 650nm, the absorption exceeded the dynamic range of the present analyzer. Previous gamma-ray irradiation experiments at room temperature[15] strongly indicated that radiation induced OH-absorption bands are caused by disbonding and resultant formation of OH radicals. This was not the case in the present experiment as it can be seen in Fig 4, where strength of the OH-absorption peaks did not change with irradiation. This difference may come from the different irradiation conditions such as temperature and irradiating environment. The previous results showed that a helium environment will improve the radiation-resistance of the present optical fiber.[15] However, the observed difference may also reflect the difference between reactor irradiation and pure ionizing irradiation.

The strong optical absorption in the wavelength range shorter than 750nm could be separated into three absorption bands. The first is the strongest absorption peak, centered at 620-630nm, which can be observed from the very early stage of the irradiation as shown in Fig. 3. The second absorption band is centered at about 570nm and could be observed after the irradiation proceeded and the absorption peak at 620nm grew substantially, as shown in Fig. 3. This second absorption band developed earlier and more clearly on the F-fiber. The last is the band whose absorption is inversely proportional to the wavelength shorter than 750nm as shown in Fig. 5. In Fig. 5, ridges could be seen at about 500-550nm. This ridge was thought to be generated by the change of sensitivity of the instrument.

The first absorption peak has already been observed in many experiments[16] and was attributed to the optical absorption by so-called non-bridging oxygen hole centers(NBOHC). Previous results[16], mainly with gamma-ray irradiation, indicated that this NBOHC absorption peak would shift its position to shorter wavelengths(to about 600nm) when optical fibers had a substantial amount of OH. However, we did not observe any peak-shift due to change of the OH content, and the peak position of the OH-fiber was found to be the same as that of the F-fiber. This difference between the present reactor irradiation and the previous gamma-ray irradiations will also indicate different effects from neutron irradiation.

The amount of irradiation-induced optical absorption was very large and exceeded the dynamic range of the present analyzer during the very early stage of the irradiation as shown in Fig. 3. An optical loss of $3x10^4$dB/km in the wavelength range shorter than 650nm was induced by a total ionizing dose of $1.4x10^7$Gy and a total displacement damage of $1x10^{-4}$dpa, before the reactor power attained its

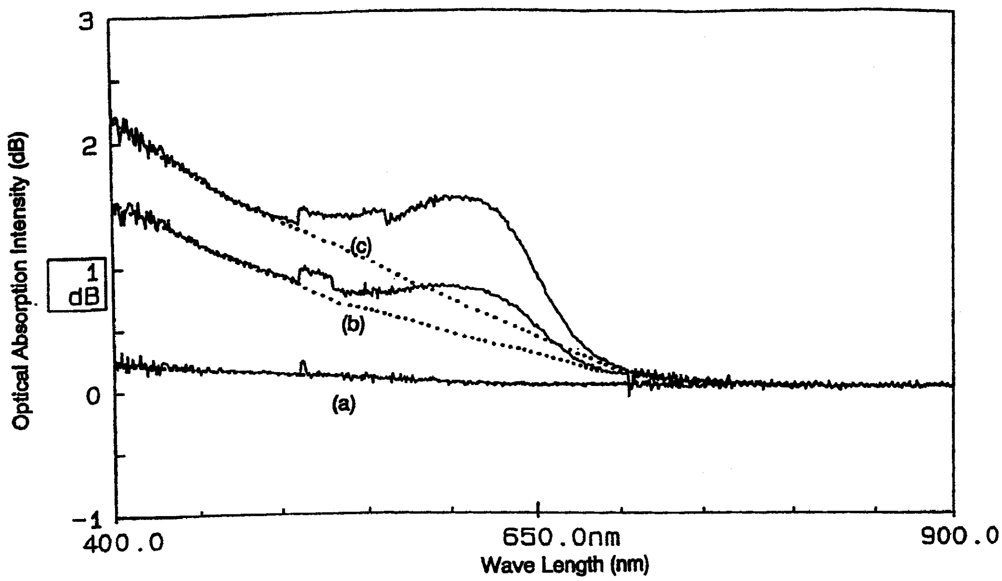

Figure 5 Optical absorption loss which has inverse linear dependence on wavelength. Rector power; (a)20kW, (b)500kW, (c)1MW.

maximum power of 50MWth. No improvement of the optical transmissivity in this wavelength range was observed on the F-fiber, during the course of irradiation. This is contradictory with the previous results[11,12]. Further examination will be needed.

In the course of the experiment, we observed that the fibers yielded some optical fluorescence radiation. Figure6 shows observed optical radiations from the F-fibers at the reactor power of 50MW, its full power. The similar optical radiations could be also observed on the OH-fiber, except for absence of a sharp peak at 1270nm. It could be definitely deduced that the observed optical signals are originated from the irradiated optical fibers, standing on the following experimental results.

1)There are distinct OH-absorption bands in the observed optical spectra(one example is shown in Figure 7), which indicated that the observed optical radiations went through the optical fibers.

2)Intensity of the observed optical radiation changed with the reactor power.(Figure 8) And the optical radiation could not be detected several days after the reactor shut down, when the gamma-ray dose rate is small and there is little neutron fluxes.

3)There was no optical leakage greater than that could be detected by the present optical spectrum analyzer, through the experiment.

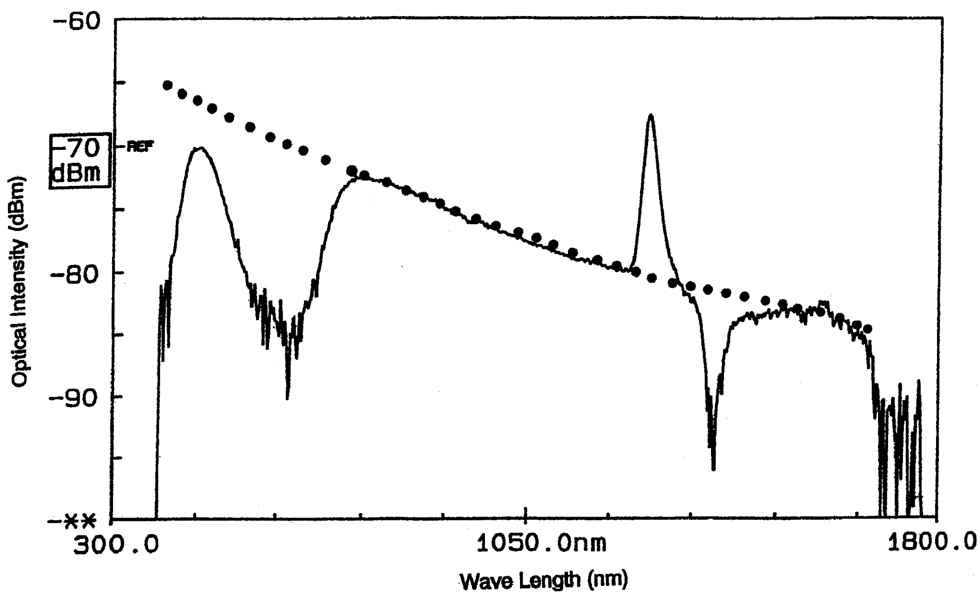

Figure 6 Observed optical radiation from F-fiber during irradiation. Dotted curve is optical radiation strength proportional to the third power of inverse of wave-length(strength distribution of Cerenkov radiation).

Superficially, the observed optical radiation from the F-fiber could be interpreted to be composed of three parts as can be seen in Fig. 6. The first is a sharp peak at 1270nm. The second is a broad band in the wavelength range of 600-1750nm. The last is a broad peak at about 500nm. Among these three optical radiations, the peak-like optical radiation at 1270nm has not been reported yet. Also, the present experiment has not yielded substantial results on behavior of this peak to deduce its origin. However it may be thought to relate with Fluorine ions doped in the SiO_2, because this peak could not be observed on the OH-fiber. Change of the strength of this peak due to change of the reactor power is shown in Fig. 7. With decrease of the reactor power, namely with decrease of the neutron flux and the gamma-ray dose rate, the peak strength decreased. However, even after the reactor power became zero, the peak could be observed as shown in Fig. 7.

The peak could be detected until 48h after the reactor shut-down. This outlived behavior may suggest that the peak is a fluorescent-natured one. However, we should notice that there is a substantial dose rate of the gamma-ray even after the reactor-shut-down. The duration of 48h is just the period during which the gamma-ray dose rate decreases rapidly to the level of several Gy/s as described in the previous section.

Then, the peak may be interpreted to be generated by the ionizing gamma-ray. The experiment with a pure gamma-ray irradiation will reveal the

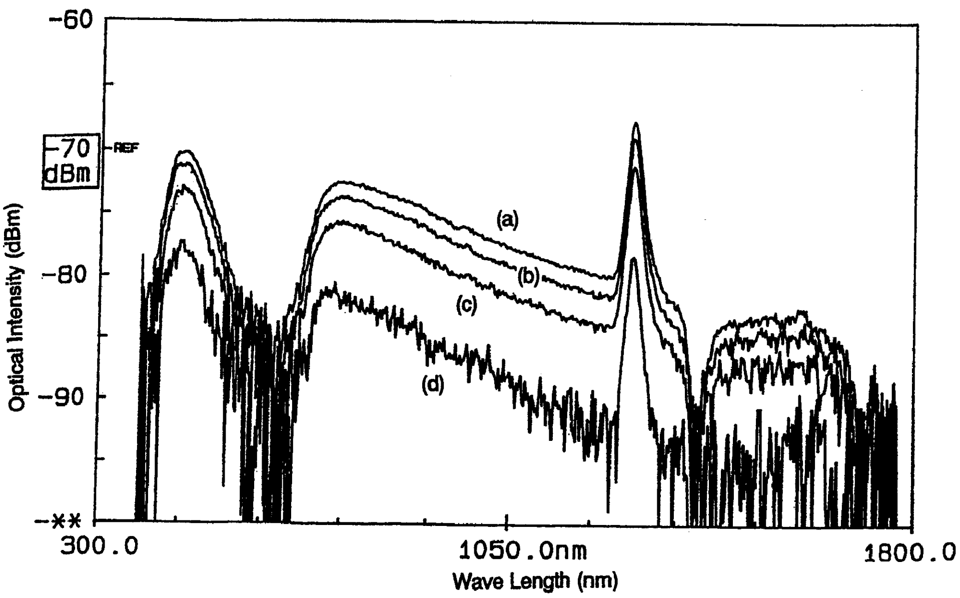

Figure 7 Change of strength of optical radiation during decrease of reactor power. (a) 50MW, (b)37MW, (c)25MW, (d)8MW.

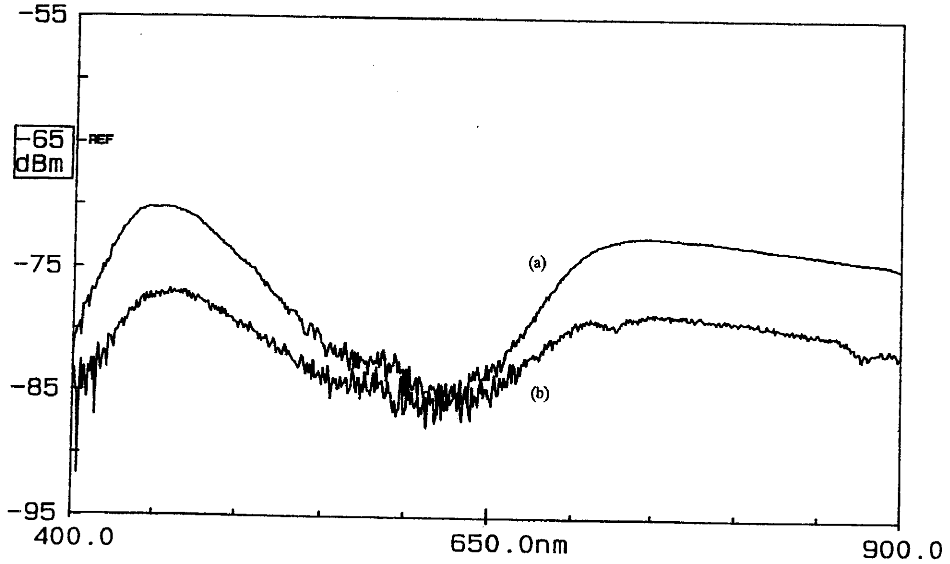

Figure 8 Comparison of optical radiation between F-fiber and OH-fiber. (a)F-fiber, (b)OH-fiber

optical luminescent peaks in this wavelength region, using electrons, ions and neutron irradiations. Griscom et al.[11] indicated that the optical luminescent peak at about 400-600nm was due to decay of transient defects. Tanabe[9] speculated that the peak, which was observed at about 400-500nm under the neutron irradiation, was a luminescent peak related with F or F^+ centers. Extensive studies have been accumulated on radioluminescence peaks in the wavelength range of 200-500nm on SiO_2 and Al_2O_3. The peaks could be identified as F and F^+ peaks in general.[18] The observed third peak may be the same peak as those observed previously and may be a luminescent peak related with some transient defects.

However, reported optical spectra in 200-500nm have structures and peaks whose half-width would be less than 50-70nm. The spectra observed in this experiment is different from these reported spectra. Another interpretation might be considered. In this experiment, a distinct optical absorption band grew in the wavelength range of 500-650nm as described above. When the white light goes through the irradiated optical fiber, we observe a distinct dip in the optical spectrum at about 500-650nm. The dip at 500-650nm in Fig. 6 corresponds to this absorption band. So, the third peak may not be an independent peak but may belong to the continuous broad optical radiation band and may be separated by the absorption band. The continuous broad optical band is the Cerenkov radiation described above.

Compared with the expected strength, the observed strength of the third peak is a little smaller as shown in Fig. 6. However, in the wavelength range shorter than 650nm, there is a optical absorption band whose absorbing strength is proportional to inverse of the wavelength. This absorption band will weaken the strength of the observed optical radiation in this wavelength range. Also, the third peak strength of the F-fiber is about 4 times as that of OH-fiber as shown in Fig. 8. This strength ratio is just the same as the second optical radiation discussed above. Furthermore, the third peak changed its strength with the reactor power in a

nature of this peak.The broad optical radiation in the wavelength range of 600-1750nm has a strength which depends on the third power of inverse of the wavelength as shown in Fig.6 . Golob et al.[17] reported the optical radiation from the SiO^2 optical fibers which was caused by the so-called Cerenkov radiation, using a pulsed electron irradiation. They showed that the strength of the observed optical radiation was proportional to the third power of inverse of the wavelength, which could be deduced from the theory of Cerenkov radiation. Also, the intensity of this optical radiation from the F-fiber is just 4 times larger than that from OH-fiber in the whole wavelength range. This means that the intensity is proportional to the cross section of the fiber, regardless of their impurity contents. These results strongly indicate that this optical radiation was caused by the so-called Cerenkov radiation.

Concerning the third optical radiation peak at about 500nm, there are some reports which have already showed similar manner as the second optical radiation band as shown in Fig. 7.

These correlations also support that the third peak belongs to the same-origin optical radiation as the second optical radiation. In the present experiment, we did not measure change of the strength of the observed optical radiations as a function of a irradiation duration. So, we have little information concerning a behavior of the growth of these optical radiations with the progress of the reactor irradiation. However, from the initial stage of the reactor irradiation, we happened to observe the third peak at 450-550nm, and the obtained results suggested that the peak existed from the very beginning of the reactor irradiation and its strength did not changed substantially through the reactor irradiation. This observation may also support that the third peak is not originated from the decay of transient defects.

The present experiment will be the first to report the strong and continuous optical radiation from SiO_2 fiber under a reactor irradiation. If the observed optical radiation is really Cerenkov radiation, it will cause a serious problem for the optical measurements in a reactor irradiation. Strength of the observed optical radiation is about -65dBm for 1m long SiO_2 fiber under the present irradiation. Also, the strong optical radiation will interfere planned optical measurements using SiO_2 fibers in developing fusion reactors. However, on the other hand, there will be possibility that the observed optical radiations could be used for new techniques diagnosing irradiation fields.

IV. Conclusion

We examined the feasibility of using optical fibers for insitu optical measure ments in a heavy irradiation environment. We irradiated two kinds, OH-doped and F-doped, optical fibers in a JMTR core up to $1.55x10^{19}n/cm^2$ fast neutron fluence and $3.3x10^9$Gy ionizing dose at 370K. The optical fibers kept their optical transparency very well in the range of wavelength longer than 750nm. The results indicate that the optical fiber can be used in this wavelength range in heavy irradiation environments. However, deterioration of the optical transparency is serious at wavelengths shorter than 700nm. The present results did not indicate any improvement by F-doping in the ultra-violet wavelength range. Fluorescence of about -60dBm/m was observed, which may disturb optical measurements in a heavy irradiation environment.

We observed optical radiations emitted from SiO_2 optical fibers irradiated in the fission reactor, JMTR. A sharp peak at 1270nm will be thought to relate with Fluorine-ions doped in the fiber core. The other optical radiations at 450-550nm and 650-1750nm would be due to the Cerenkov radiation. The observed optical radiation will be hazardous for optical measurements using optical fibers in heavy irradiation environments, but can be used for new techniques diagnosing irradiation fields.

.1) S. Yamamoto compiled, "ITER Workshop on Radiation Effects on Diagnostic Components," held at The Dvefremov Scientific Research Institute of Electrophysical Apparatus, St. Petersburg, USSR, Oct., 1991, compiled by Naka Research Establishment, Japan Atomic Energy Research Institute, Japan, (1992)
2) G.P.Pells, presented at Int. Symp. on Sci.of Al2O3, Schloss Ringberg, March, 1993, to be published in J. Amer. Ceram. Soc., (1994).
3) A.Ibarra, R.Heidinger and J.Molla, J. Nucl. Mater., 191-194 (1992) 530.
4) A.A.Stoneham, presented in this conference, to be published in Nucl. Instr. Methods.
5) F.W.Clinard, G.F.Hurley, L.W.Hobbs, D.L.Rohr, and R.A.Youngman, J. Nucl. Mater., 122-123 (1984) 1386.
6) T.Shikama and G.P.Pells, presented at 7th Int. Conf. on Fusion Reactor Materials, Stresa, Oct., 1993, to be published in J. Nucl. Mater..
7) A.Morono and E.R.Hodgson, presented at 7th Int. Conf. on Fusion Reactor Materials, to be published in J. Nucl. Mater..
8) H.Ohno, N.Igawa, Y.Ishii, N.Umesaki, and M.Tokita, J.Nucl. Mater., 191-194 (1992) 525.
9) T.Tanabe, in E. H. Farnum edited, "A US/Japan Workshop on Dynamic Effects of Irradiation in Ceramics," Los Alamos, New Mexico, (1992) pp.319, Los Alamos National Laboratory, New Mexico.
10) W.Williams, in E.H.Farnum edited, "Radiation Effects in Materials for Fusion Diagnostic Systems, Planning Workshop," Princeton, March, 1991, pp. 292, (1991) Los Alamos National Laboratory, New Mexico.
11) G. Griscom, in ref.9) pp.319.
12)T. Kakuta, et al., to be presented at Proc. Annual Meeting of Jpn. Electr. Infor. Soc., Fall Meeting of 1993.
13) M. Narui, et al., to be present at the 7th Inter. Conf. on Fusion Reactor Materials, to be published in J. Nucl. Mater..
14) R. Oyamada, edited, "JMTR Irradiation Handbook," Division of JMTR Project, Oarai Research Establishment, Japan Atomic Energy Research Institute, JAERI-M-83-053, Oarai, Japan, (1983).
15)T. Kakuta, et al., Reports on Insulating Materials of Jpn. Electr. Soc., EIM-89-133 (1989), Jpn. Electr. Soc., Tokyo.
16)K. Nagasawa, M. Tanabe, and K. Yahagi, Jpn. J. Appl. Phys., 23 (1984) 1608.
17) J.E. Gobol, et al., IEEE Trans. Nucl. Sci., NS-24 (1977) 2164.
18)K.H.Lee and J.H.Crawford, Phys. Rev., B19 (1979) 3217.