Radiation Hardening of Pure Silica Optical Fibers by High-Pressure Hydrogen Treatment

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***Abstract*—It was proved that hydrogen (H_2) treatment, consisting of loading with H_2 under high pressure and immediate pre-irradiation up to dose values $\geq 10^4$ Gy considerably increases the radiation hardness of optical fibers with a pure SiO_2 core, especially of those with a low OH content. These fibers even show a reduction of their initial attenuation at wavelengths <700 nm, in contrast to a significant increase between 1200 and 1600 nm (growth of OH absorption bands). Therefore, H_2 treatment should be restricted to fiber applications in the visible wavelength range, up to about 850 nm. A treatment with highly enriched deuterium should extend the usefulness to wavelengths around 1300 nm.**

***Index Terms*—Hydrogen treatment, optical fibres, radiation, radiation hardening.**

I. Introduction

FIRST successful attempts in reducing the radiation sensitivity of optical fibers with pure silica core by a hydrogen treatment were reported by Lyons and Looney [1], [2]. In principle, these authors applied the same procedure as we do in our present investigation: fibers that are saturated with hydrogen of a certain pressure are irradiated. They measured the radiation-induced loss increase during this first irradiation (=pre-irradiation) and got only about 1/5 of the values of fibers without hydrogen (= untreated fibers) at 850 nm. The fiber samples then rested at room temperature up to 80 days and were irradiated again. Despite the fact that the hydrogen should have diffused out meanwhile, the loss increase still was lower than with untreated fibers, in some cases about 25%. The authors explained this permanent improvement by a radiation-induced dissociation of H_2 molecules into H atoms, which bind to defects that otherwise would act as precursors of color centers during subsequent irradiations. The authors gave a comprehensive overview of all earlier activities in the field of hydrogen treatment of optical fibers with and without radiation exposure, so that this was not repeated in the present paper.

For their efforts Lyons and Looney selected a fiber type (pure SiO_2 with high OH content) that is known to be radiation hard anyhow, hoping to achieve further improvement. The hydrogen pressure was only 4.7 bar, and the ^{60}Co gamma dose for pre-irradiations was only 10, 100, and 500 Gy, respectively. These might be the reasons for their relatively minor and short-lasting reduction of the radiation sensitivity.

Tomashuk *et al.* [3] reported on a similar procedure to improve the radiation hardness of two Russian fibers with the core material KU (pure silica, 800 ppm OH, 80 ppm Cl) and KS-4V (pure silica, 0.25 ppm OH, 20 ppm Cl), respectively. The authors said that especially the latter material shows the lowest radiation-induced loss (at high dose values) in the visible spectral range of all commercial core materials, so that they restricted their investigations to this wavelength region. Their fiber samples were exposed to a H_2 pressure of 24 bar during 11 days at room temperature. Two irradiations were performed within "several days" after the H_2 loading and a third one three months later. They report that "the stockpile of molecular hydrogen in the fiber was exhausted during irradiation run #2." For comparison, one H_2 loaded sample was also exposed to a heat treatment (200 min at 200 °C). After having reached a few dose levels, they only performed off-line spectral loss measurements (cut-back method). They observed distinct improvement of the radiation hardness of H_2 treated fiber samples, also three months later during the third irradiation. Successive H_2 loading, followed each time by an irradiation, would further increase the radiation hardness.

Our present approach was as follows: exposing the fibers to the highest available H_2 pressure ($\lesssim$150 bar with our autoclave), (pre-)irradiation up to the highest allowable dose immediately after removal from the autoclave, and waiting until the residual H_2 is diffused out of the fibers ($\geq$4 weeks). That is what *we* call "hydrogen treatment." After that we perform spectral attenuation measurements in order to compare the "initial" attenuation of H_2 treated and untreated fibers. The treatment should not lead to an unacceptably high attenuation increase. And only now we perform the irradiation tests when the result is no longer influenced by residual (unknown and changing) amounts of H_2. The test conditions should be as well defined and as reproducible as possible.

The pre-irradiation dose might influence the final dose up to which an improvement of the radiation hardness would hold. We varied this dose, but also the dose rate in order to identify optimal parameters.

Furthermore, we wanted to find out whether an additional "temperature treatment" of the hydrogen treated fibers which first of all could accelerate the out diffusion of the residual hydrogen would have positive or negative influence on the observed reduction of the radiation sensitivity. Finally, it will be compared with our most promising fiber whether a deuterium (D_2) treatment would have the same result as the H_2 treatment. This could become necessary in case the H_2 treatment would lead to an unacceptably high attenuation increase between about 1200 nm and 1600 nm (OH absorption bands around 1380 nm). The OD bands in this region are orders of magnitude lower than the OH bands.

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To sum up it can be said that investigation of the influence of H_2 treatment on the radiation hardness of pure SiO_2 fibers was done for the first time under optimized, unambiguous, and reproducible conditions and with a number of different fiber types.

II. Experimental

In order to find out the dependence of the effectiveness of the H_2 treatment on fiber type, we choose five distinctly different, but commercially available pure SiO_2 core fibers: one Sumitomo single-mode (SM) fiber optimized for 1550 nm, one "radiation hard" Mitsubishi multimode step-index (MMSI) fiber and three different MMSI fibers from Heraeus Quarzglas (now Heraeus Tenevo). Two of them are made of the well-known core materials F 100 (high OH, low Cl; "Fluosil SSU" fiber) and F 300 (low OH, high Cl; "Fluosil SWU" fiber), respectively, and one of their new STU core material (low OH, reduced Cl; "Fluosil STU" fiber). All MMSI fibers had a core diameter of about 100 μm. The Mitsubishi fiber and the Heraeus STU and SSU fibers are described in more detail in [4].

For H_2 loading, the fibers remained in the autoclave for 14 days at room temperature, exposed to a pressure of about 125 bar. Because of the limited time, D_2 loading took place only during six days, but at a mean temperature of about 125 °C. The mean pressure was about 120 bar. Nearly all fiber samples were pre-irradiated (some with varying dose rate up to different dose values) immediately after removal from the autoclave. For comparison, one Sumitomo SM fiber sample and one STU fiber sample were not pre-irradiated. One of the pre-irradiated STU samples was exposed to a temperature of 200 °C for 200 minutes, immediately after pre-irradiation. After pre-irradiation, the H_2 and D_2 loaded fibers are called "H_2 (or D_2) treated." The two fiber samples that were not pre-irradiated are called "H_2 loaded."

We made spectral attenuation measurements as well as measurements at fixed wavelengths with the equipment described in [4]. With some fiber samples we measured the change of attenuation also during pre-irradiation at 829 nm or 1310 nm wavelength. With some other samples we made spectral attenuation measurements four weeks after H_2 or D_2 treatment (or H_2 loading) and compared the results with those of untreated or unloaded (="virgin") fibers. All irradiations that should show the influence of H_2 or D_2 treatment (or loading) on the radiation hardness were made at least four weeks after removal from the autoclave and pre-irradiation. With most of the fiber types we performed a second irradiation between six and 16 months after the first one in order to demonstrate the still present improvement of their radiation hardness. All measurements were made at room temperature, those at fixed wavelengths with the recommended [5] light power of 1 μW in order to reduce the influence of "photo bleaching."

III. Results

A. Attenuation Behavior During Pre-Irradiation

Mostly, we made no measurements during the pre-irradiation of H_2 or D_2 loaded fibers. Fig. 1 illustrates the reason why. Here we see the change of the light intensity at the output of 223 m long H_2 loaded samples of the STU and the SWU fiber during and after pre-irradiation with a dose rate of about 0.27 Gy/s up to a dose of $2 \cdot 10^4$ Gy. Irradiation time was 74 350 s. At the beginning we really see a radiation-induced increase of attenuation with both fibers. After about 30 s (8 Gy) the STU fiber reaches its well-known intermediate maximum [4]. But only about 10 s later we see a steady decrease of attenuation, and after an irradiation time of $\geq 10^4$ s the attenuation of both fiber samples is lower than before irradiation. This decrease still continues after the end of irradiation, with the same slope than before. During irradiation of an untreated STU fiber the maximum begins after about the same time (about 30 s), but then the attenuation begins to decrease only after times >1000 s and does not in the least reach the zero level again. Only after the end of irradiation we see a sharp decrease, but to values still far below zero. The latter is also valid for untreated SWU fibers. The pre-irradiation behavior can be explained by two processes: transformation of absorbing color centers by binding of H atoms, and outdiffusion of H_2 molecules dominating at later times. These molecules cause a wavelength dependent attenuation increase compared with unloaded fibers that continuously decreases during the outdiffusion with unchanged rate after the end of irradiation (since the radiation-induced temperature increase is negligible, as well as transformation of H_2 into H during irradiation). The slope is the same with both fibers since they have the same dimensions and were H_2 loaded in exactly the same way.

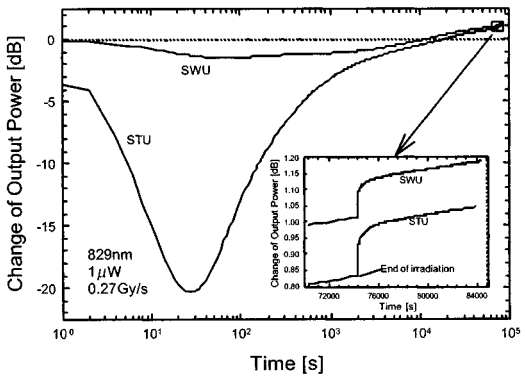


Fig. 1. Attenuation behavior of STU and SWU fibers during "pre-irradiation" (see text). Irradiated fiber length: 223 m each.

These two processes prevent performing expressive and reproducible measurements of the radiation-induced attenuation during pre-irradiation, when the fibers are still saturated with H_2. We only can determine its amount at the end of irradiation from the height of the annealing step (insert of Fig. 1). The height of this step leads to values of about 0.5 dB/km for the SWU fiber and 0.7 dB/km for the STU fiber, compared with about 75 dB/km of an untreated STU fiber and about 5 dB/km of a H_2 treated STU fiber. Hence, the best method of radiation-hardening would be to confine a high amount of H_2 in a fiber—if possible! But this would only be useful for wavelength regions with negligible H and H_2 absorption bands.

B. Influence of the Pre-Irradiation Parameters

The pre-irradiation parameters were only varied with the Sumitomo SM and the Heraeus STU fibers. In Fig. 2, we see the influence of the pre-irradiation dose for the Sumitomo fiber.

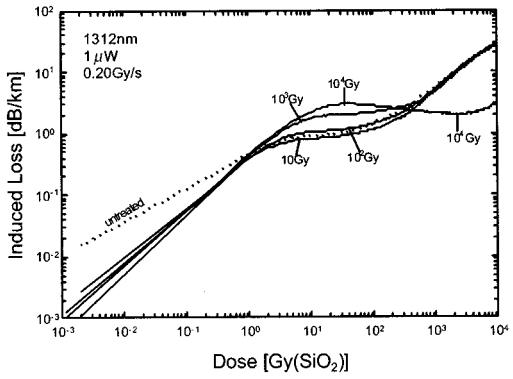


Fig. 2. Influence of the "pre-irradiation" dose (10–10^4 Gy) on the increase of attenuation during irradiation of H_2 treated Sumitomo SM fiber samples. For comparison attenuation increase of an untreated fiber sample.

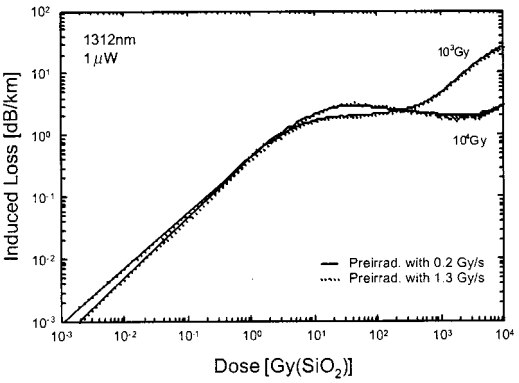


Fig. 3. Influence of the pre-irradiation dose rate on the increase of attenuation of H_2 treated Sumitomo SM fiber samples.

The pre-irradiations were made with the same dose rate of about 0.20 Gy/s up to dose values of 10, 10^2, 10^3, and 10^4 Gy, respectively. About one month later, these four H_2-treated fiber samples were irradiated with about the same dose rate up to 10^4 Gy each (="first irradiation"). Only after a pre-irradiation dose of 10^4 Gy we see a reduction of the radiation-induced attenuation at dose values $\gtrsim 10^3$ Gy. Between 1 Gy and 100 Gy we even see an increase [see also Fig. 9(a) and (b)].

The pre-irradiations up to 10^3 Gy and 10^4 Gy were made with a dose rate of about 0.20 Gy/s (Fig. 2) as well as with about 1.3 Gy/s. It could perhaps be that pre-irradiation with only 10^3 Gy nevertheless would become effective, if only the dose rate would be increased. This dose rate increase should, e.g., lead to a higher concentration of H atoms (radiation-induced dissociation of H_2 molecules). However, Fig. 3 shows that the dose rate has no influence, for both dose values.

With the Heraeus STU fiber we pre-irradiated two H_2 loaded samples up to $2 \cdot 10^4$ Gy and one up to $2 \cdot 10^5$ Gy. Each time the dose rate was about 1.1 Gy/s. Immediately after pre-irradiation one of the $2 \cdot 10^4$ Gy samples was heated up to 200 °C for 200 min. If this would have no negative (or even positive) influence on the attenuation increase during subsequent irradiations, one could use this procedure for accelerating the H_2 outdiffusion. In order to check the effectiveness of the heating procedure, we made a spectral attenuation measurement that would have shown noticeable H_2 concentrations. One month later we performed such measurements with all three samples. It became clear that the heating had removed the hydrogen completely. Further details of these spectral measurements are treated in Section III-C. In Fig. 4, we see the influence of these measures on the attenuation increase during irradiations up to 10^5 Gy. These tests began after the spectral measurements. The additional heat treatment caused a slightly higher attenuation increase, whereas pre-irradiation up to $2 \cdot 10^5$ Gy reduced the radiation response considerably. Untreated fibers under the same irradiation conditions show a loss increase of about 275 dB/km [4]! The question whether such a high pre-irradiation dose may

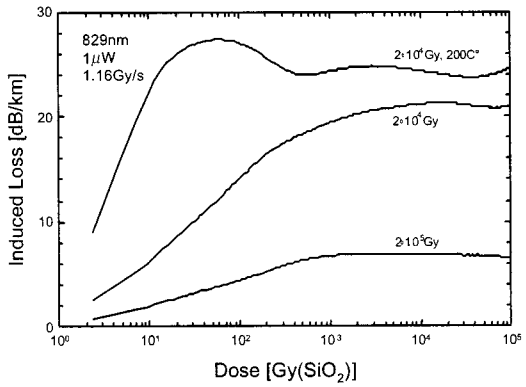
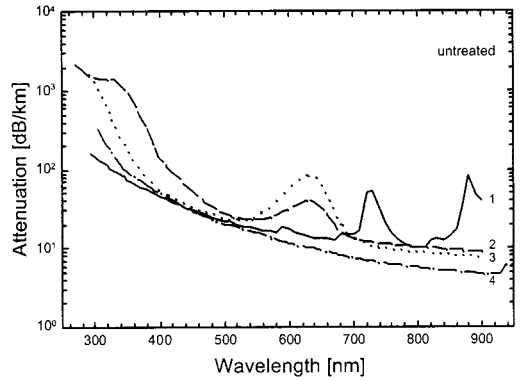
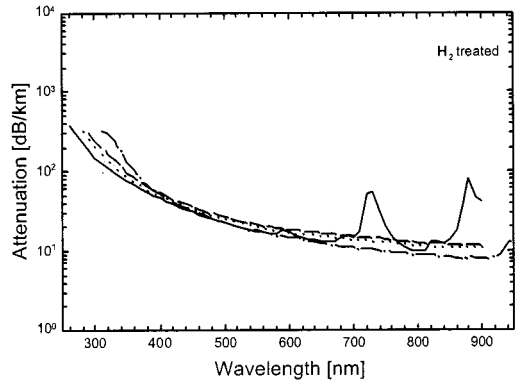


Fig. 4. Influence of pre-irradiation dose and subsequent heating (200 min at 200 °C) on the increase of attenuation during irradiation of H_2 treated Heraeus STU fiber samples.



(a)



(b)

Fig. 5. (a) Spectral attenuation of four MMSI fibers before and (b) after H_2 treatment. Pre-irradiation dose was 10^4 Gy (curve 1) and $2 \cdot 10^4$ Gy (curves 2–4), respectively. 1: Heraeus SSU1.2107/00 (F-100), 2: Heraeus SWU1.2 087/2000 (F-300) 3: Heraeus STU1.2 237/2000, 4: Mitsubishi STR1000-SY.

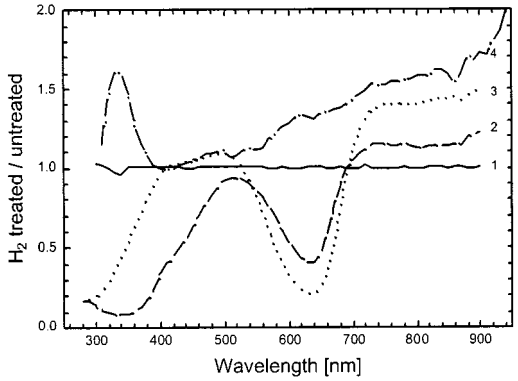


Fig. 6. Change of fiber attenuation caused by Hz treatment (=attenuation H_2-treated/attenuation untreated). 1: Herueus SSU1.2107/00 (F-100), 2: Heraeus SWU1.2 087/2000 (F-300), 3: Heraeus STU1.2 237/2000, 4: Mitsubishi STRI OOC-SY.

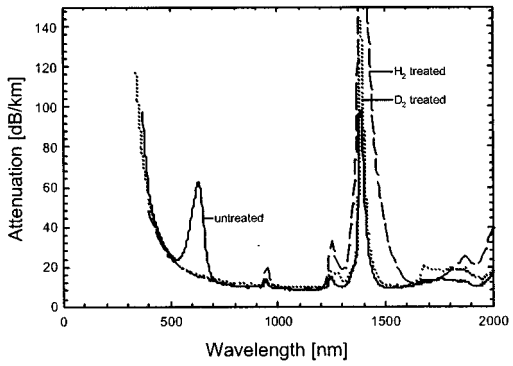


Fig. 7. Influence of H_2 and D_2 treatment on the initial attenuation of a Heraeus STU fiber.

have negative influence on the "initial" fiber attenuation will be treated in Section III-C.

C. Influence of H_2 and D_2 Treatment on Fiber Attenuation

An essential precondition for reducing the radiation-induced attenuation of optical fibers by H_2 treatment is that this procedure will not increase their initial attenuation ($\geq$one month after the treatment) distinctly above that of untreated fibers. Otherwise, the reduction of the radiation-induced attenuation might even be overcompensated by the increase of the initial attenuation. Fig. 5(a) and (b) show the spectral attenuation of the four investigated MMSI fibers between about 300 nm and 900 nm for untreated Fig. 5(a) and H_2 treated samples Fig. 5(b). Especially the Heraeus SWU and STU fibers experience a significant attenuation decrease by the H_2 treatment so that they become comparable with the high OH SSU fiber or the “radiation hard” Mitsubishi fiber. The H_2 treatment removes the “drawing-induced peak” at about 630 nm [6] of the SWU and STU fiber as well as the Cl peak at about 330 nm [7] of the SWU fiber! Fig. 6 shows the influence of the H_2 treatment more quantitative, i.e., we divided the attenuation curve of the H_2 treated fibers by the respective curve of the untreated fibers. This is a well suited method to compare the influence of the H_2 treatment on the different fiber types, so that we will also apply it to demonstrate the different amounts of radiation hardening. Fig. 6 clearly shows that the attenuation of the SSU fiber with its high OH content remains completely uninfluenced by the H_2 treatment and that the attenuation of the low OH fibers increased in the IR.

The influence of a H_2 and D_2 treatment at wavelengths up to 2000 nm is shown for the STU fiber in Fig. 7. Only that fiber was also treated with D_2. We can see that the “drawing-induced peak” at about 630 nm is also removed by the D_2 treatment. However, the nearly identical H_2 treatment (same pressure and pre-irradiation dose of $2 \cdot 10^4$ Gy) caused a significant increase of the OH absorption peak at 1380 nm. The flanks of this peak increased the attenuation around 1300 nm and 1550 nm considerably. H_2 treatment with following heat treatment caused exactly the same increase of the 1380 nm peak, whereas the pre-irradiation dose of $2 \cdot 10^5$ Gy (see Section III-B) caused still higher attenuations around 1300 nm and 1550 nm. From the measured curves we roughly determined the following attenuation increase: 23 dB/km at 1300 nm and 28 dB/km at 1550 nm for $2 \cdot 10^4$ Gy and 47 dB/km at 1300 nm and 60 dB/km at 1550 nm for $2 \cdot 10^5$ Gy.

These values are distinctly higher than the radiation-induced loss of untreated fibers up to a dose of 10^6 Gy, at 1310 nm as well as at 1514 nm [4], so that H_2 treatment is unsuited for the reduction of the anyhow moderate radiation-induced loss of STU fibers in the infrared wavelength range (IR). A change to D_2 treatment improves the situation. That also this treatment caused an increase of the 1380 nm peak (and, thus, of the attenuation around 1300 and 1550 nm) might come from the H_2 contamination of the D_2 gas in the order of 0.2%.

With the Mitsubishi fiber, we also saw a distinct increase of the 1380 nm-peak after H_2 treatment with a pre-irradiation dose of $2 \cdot 10^4$ Gy. With the sensitivity of 62.7 dB/km per ppm OH published in [8], we calculated an increase of the OH content of nearly 3 ppm, compared with only about 1 ppm of the untreated fiber.

D. Radiation Hardening: Measurements at Fixed IR Wavelengths

In the IR optical fibers usually are used for data transfer at fixed wavelengths around 840, 1300, and 1550 nm, respectively. Because of the OH absorption bands the Heraeus SSU fiber (F 100 material) with its high OH content can only be used up to about 860 nm. Our numerous comparisons of the radiation-induced loss increase at 1300 and 1550 nm showed that during long-term fiber use in a radiation environment ($>$1 d) the loss increase at 1550 nm will be higher than at 1300 nm. Some early results are published in [9]. We therefore restricted our tests to the two shorter wavelengths.

In order to determine the influence of a H_2 or D_2 treatment on the radiation sensitivity of a given fiber type, we performed comparable irradiation tests with untreated and H_2 or D_2 treated fiber samples. The time between treatment (pre-irradiation included) and first irradiation test was at least one month. With most of the fibers we made a second irradiation with untreated and treated sample between six and 16 months later in order to see whether the improvement of the radiation hardness would be “permanent.” Most of the irradiations were only made up to 10^4 Gy. Only in some cases we tried to find out whether the improvement would also hold up to higher dose values.

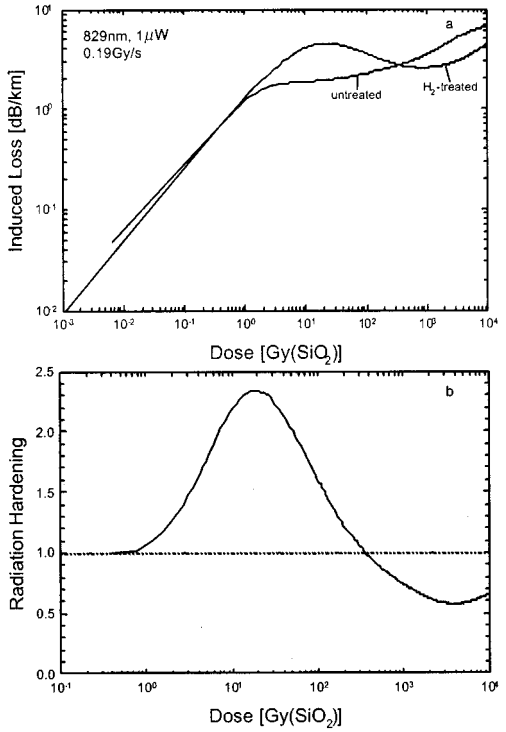


Fig. 8. (a) Increase of attenuation during irradiation of untreated and H_2 treated samples of the Heraeus SSU fiber and (b) "radiation hardening" = ratio of H_2-treated and untreated curve from (a).

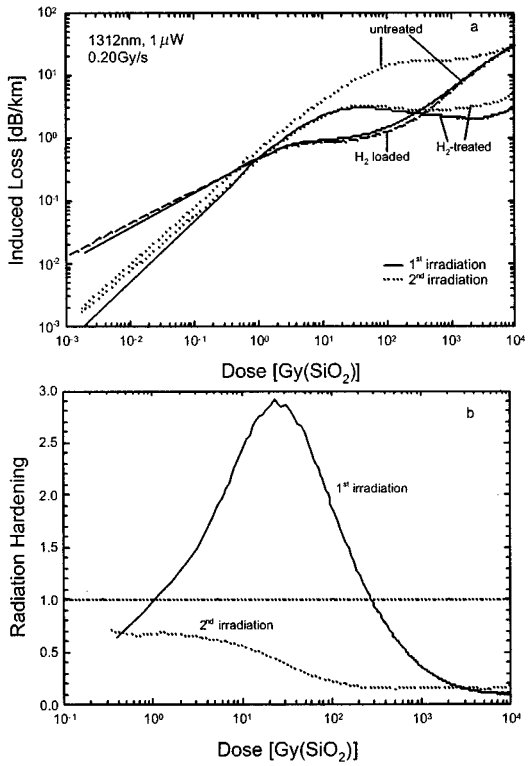


Fig. 9. (a) Increase of attenuation during first and second irradiation of untreated and H_2 treated samples as well as of a "H_2 loaded" sample of the Sumitomo SM fiber. (b) "Radiation hardening" as a function of dose.

Fig. 8(a) and (b) show the influence of H_2 treatment (pre-irradiation dose 10^4 Gy) on the Heraeus SSU fiber (high OH). Especially Fig. 8(b) (ratio of H_2 treated and untreated fiber measurement = "radiation hardening") shows that at the beginning the H_2 treatment even causes an increase of attenuation and only above 300 Gy a reduction of about 50%, finally. This low reduction is not surprising, considering the high OH content of this fiber that is known to be radiation hard. Therefore, Lyons and Looney [1], [2] could only find minor improvements, especially with their low pre-irradiation dose values (see Section III-B).

The Heraeus SWU fiber (low OH) is intended for the far IR, but we could not make quantitative and clear measurements of a radiation-induced loss increase of H_2 treated samples at 1310 and 1550 nm. We saw an increase of attenuation only at the beginning, followed by a more (1514 nm) or less (1310 nm) pronounced decrease, so that after the end of irradiation we usually had lower fiber attenuation than at the beginning. The same could be observed with the Heraeus STU fiber. The reason might be the high increase of attenuation around the 1380 nm absorption band by the H_2 treatment (Fig. 7) that seems to decrease during subsequent irradiations. This behavior suggests that the improvement of the radiation hardness by H_2 treatment might become exhausted during longer irradiation periods—if there is a connection between improvement of the radiation hardness and increase of the OH content (see Section III-C).

In contrast to the Heraeus SWU and STU fibers, we were able to measure radiation-induced attenuations at 1300 nm with H_2 treated samples of the two low OH fibers made by Sumitomo (SM) and Mitsubishi (MMSI), respectively, i.e., we did not observe a decrease of attenuation in the course of an irradiation. Fig. 9(a) illustrates the increase of attenuation of untreated and H_2 treated Sumitomo SM fiber samples, Fig. 9(b) their "Radiation Hardening" by H_2 treatment. Again, we observe a higher attenuation of the H_2 treated fiber at the beginning. After the same dose of about 300 Gy as with the Heraeus SSU fiber [Fig. 8(b)] the H_2 treatment leads to an attenuation reduction to about 1/10 of the attenuation increase of untreated fiber samples, finally. A second irradiation of the untreated and H_2 treated samples 16 months later showed that the attenuation increase of the H_2 treated sample was still less than 1/5 of the untreated sample. Now we did not observe the higher initial attenuation of the H_2 treated sample. This indicates that the improvement of the radiation hardness by H_2 treatment is really permanent. For comparison, we added the attenuation increase of an only "H_2 loaded" fiber sample, i.e., we made no pre-irradiation immediately after the end of the H_2 loading. Here we have almost exactly the same attenuation increase as during the first irradiation of the untreated fiber samples. This indicates that after one month the H_2 has left the fiber completely, and that without pre-irradiation the "H_2 loading" has negligible influence on the radiation hardness.

The same irradiation tests were made with the Mitsubishi MMSI fiber, apart from the investigation of a "H_2 loaded" fiber sample. The second irradiation was made seven months after the first one. The results for measurements at 829 and 1310 nm are shown in Figs. 10(a), (b) and 11(a), (b). Here we observe a final reduction of the attenuation increase of the H_2 treated sample to about 30% of that of an untreated sample at both wavelengths, during first and second irradiation. The tendency toward higher dose values is not quite clear. The second irradiation at 829 nm indicates an increase of the ratio above 10^3 Gy, i.e., decrease of

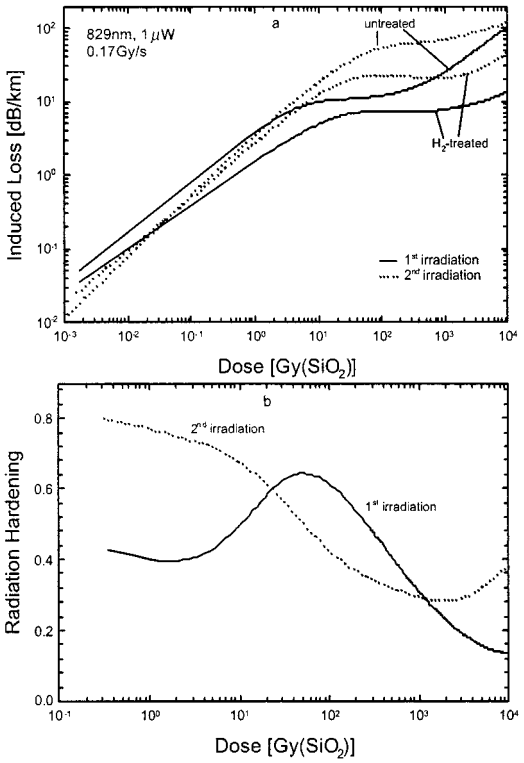


Fig. l0. (a) Increase of attenuation during first and second irradiation of untreated and H_2 treated samples of the Mitsubishi fiber and (b) "radiation hardening" as a function of dose.

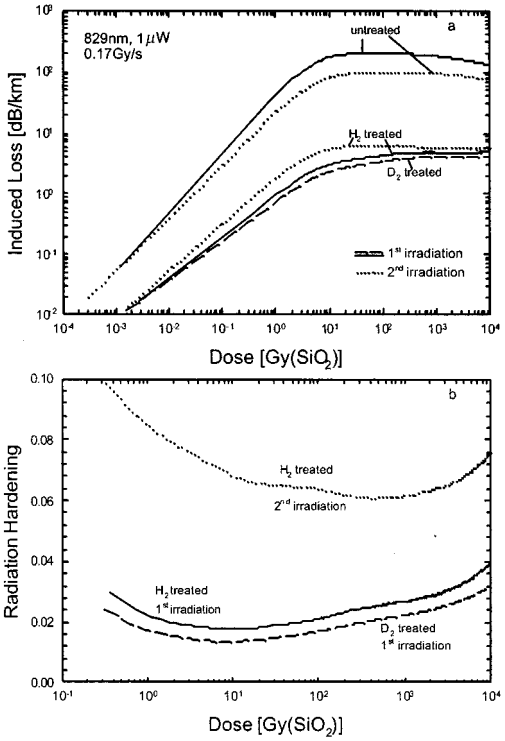


Fig. 12. (a) Increase of attenuation during first and second irradiation of untreated and H_2 treated samples of the Heraeus STU fiber and during first irradiation of a D_2 treated fiber sample, as well as (b) "radiation hardening" as a function of dose.

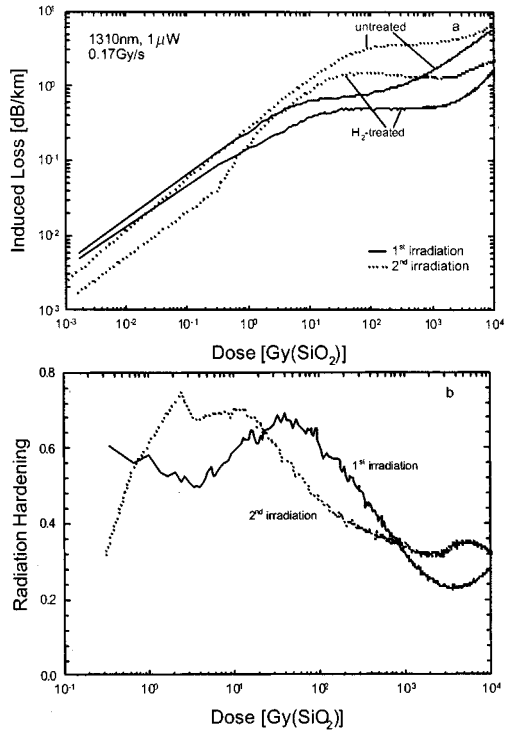


Fig. 11. (a) Increase of attenuation during first and second irradiation of untreated and H_2 treated samples of the Mitsubishi fiber and (b) "radiation hardening" as a function of dose.

the radiation hardening by H_2 treatment, whereas the (less accurate) measurements at 1310 nm do not show this tendency.

The highest reduction of the radiation sensitivity of H_2 treated fibers was observed with the STU fiber. Fig. 12(a) shows the increase of attenuation during first and second (nine months later) irradiation of untreated as well as H_2 treated fiber samples and during a first irradiation of a D_2 treated sample. During the first irradiation H_2 or D_2 treated fibers show only about 2%–3% of the attenuation increase of untreated fibers [Fig. 12(b)]. The reason for the still higher improvement by the D_2 treatment could be explained, e.g., by slower outdiffusion of D_2 than of H_2. As a consequence, the D_2 concentration was higher during the about 22 h lasting pre-irradiation than the H_2 concentration.

The reason for the smaller attenuation reduction (only to about 7%) during the second irradiation mostly arises from the lower attenuation increase during the second irradiation of the untreated fiber sample. A "H_2 loaded" sample showed approximately the same attenuation increase as untreated STU fiber samples, i.e., no improvement as observed with the Sumitomo SM fiber.

H_2 and D_2 treatment of STU fibers reduces their attenuation increase at 829 nm to values comparable with that of SSU fibers (Fig. 13). At dose values above 10^4 Gy especially a D_2 treated STU fiber will even show lower attenuation than the SSU fiber. Its attenuation at about 1310 nm is less than 3.6 dB/km up to 10^4 Gy.

An irradiation with 1.11 Gy/s up to 10^6 Gy of a STU fiber sample that was pre-irradiated with $2 \cdot 10^5$ Gy (Fig. 4) should clarify whether the improvement of the radiation hardness by H_2 treatment would also last up to distinctly higher dose values. The result (Fig. 14) is affected by a decrease of attenuation, parallel to the radiation-induced increase that after the end of irradiation

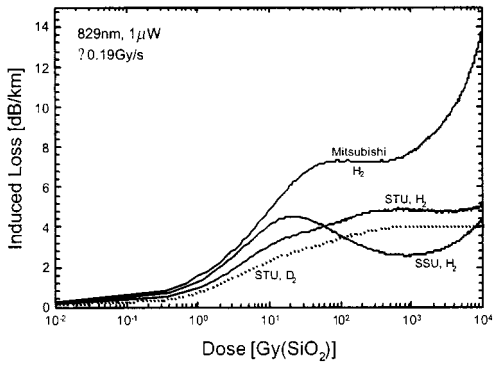


Fig. 13. Comparison of the radiation sensitivity of H_2 and D_2 treated samples of the Heraeus SSU and STU fibers and of the Mitsubishi fiber.

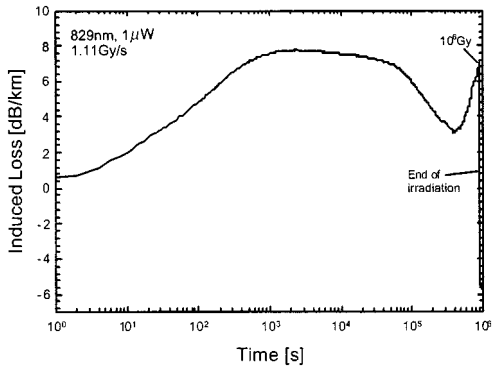


Fig. 14. Attenuation behavior of a H_2 treated Heraeus STU fiber (pre-irradiation dose $2 \cdot 10^5$ Gy) during irradiation with 1.11 Gy/s up to 10^6 Gy (see text).

led to a lower loss than before irradiation. This decrease might be caused by a decrease of the far-reaching high OH absorption bands of H_2 treated fiber samples (Fig. 7). Nevertheless, the measurement seems to indicate a new increase of attenuation above about $4 \cdot 10^5$ s ($>4.4 \cdot 10^5$ Gy), perhaps caused by a weakening of the improvement by the H_2 treatment. Untreated STU fiber samples show no new increase of attenuation below 10^6 Gy [4].

E. Radiation Hardening: Spectral Measurements in the "Visible"

With the four MMSI fibers we measured the loss increase from 300 to 850 nm with untreated and H_2 treated samples online at dose values of 30 Gy, 100 Gy, 300 Gy, 1 kGy, and 10 kGy (=1 Mrad), as well as 10 h 45 min after the end of irradiation. The loss increase was calculated from the light intensity measured immediately before irradiation and at the abovementioned dose values or time after irradiation, respectively. The pre-irradiation dose values were those given below Fig. 5(a) and (b) and the dose rate was about 0.18 Gy/s.

Fig. 15(a) shows the results obtained with untreated and H_2 treated samples of the Heraeus STU fiber that was most improved by the H_2 treatment. At higher dose values the attenuation increase was reduced to $<1/20$ of that of the untreated fiber sample at wavelengths >350 nm. This is emphasized by Fig. 15(b) showing the ratio of the respective curves of H_2 treated and untreated fibers from Fig. 15(a). Most striking is

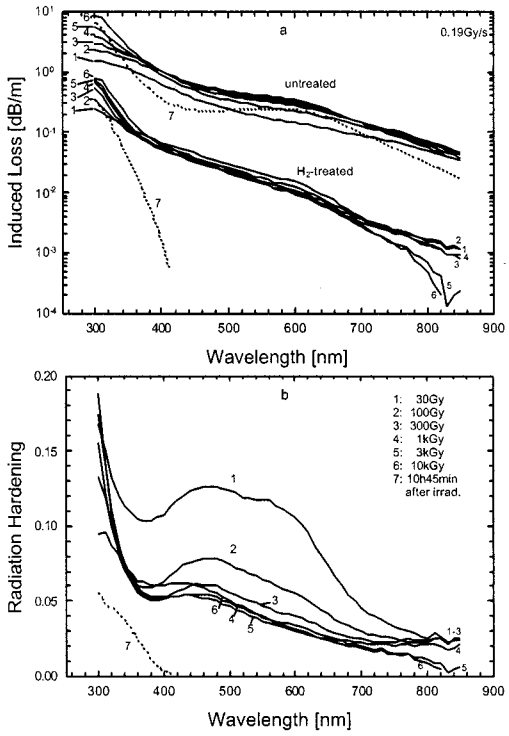


Fig. 15. (a) Radiation-induced loss increase of untreated and H_2 treated samples of the STU fiber at different dose values and 10 h 45 min after the end of irradiation and (b) the ratio of the respective H_2 treated and untreated fiber attenuation increase that is defined as "radiation hardening."

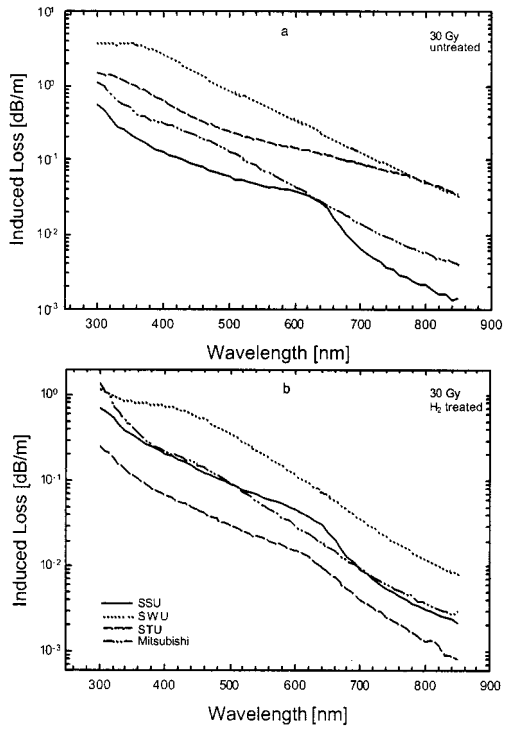


Fig. 16. Attenuation increase as a function of wavelength of (a) untreated and (b) H_2 treated samples of the four MMSI fibers at a dose of 30 Gy.

the acceleration of the "annealing," i.e., the loss reduction 10 h 45 min after the end of irradiation.

Figs. 16(a), (b) and 17(a), (b) compare the improvement by H_2 treatment of the four different MMSI fiber types at dose

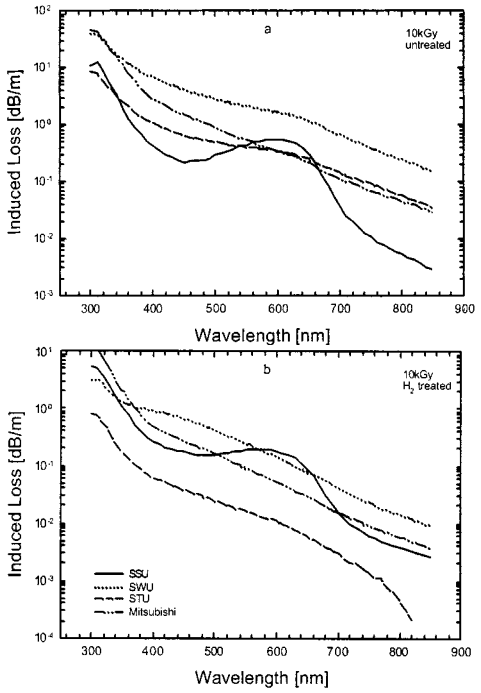


Fig. 17. Attenuation increase as a function of wavelength of (a) untreated and (b) H_2 treated samples of the four MMSI fibers at a dose of 10^4 Gy.

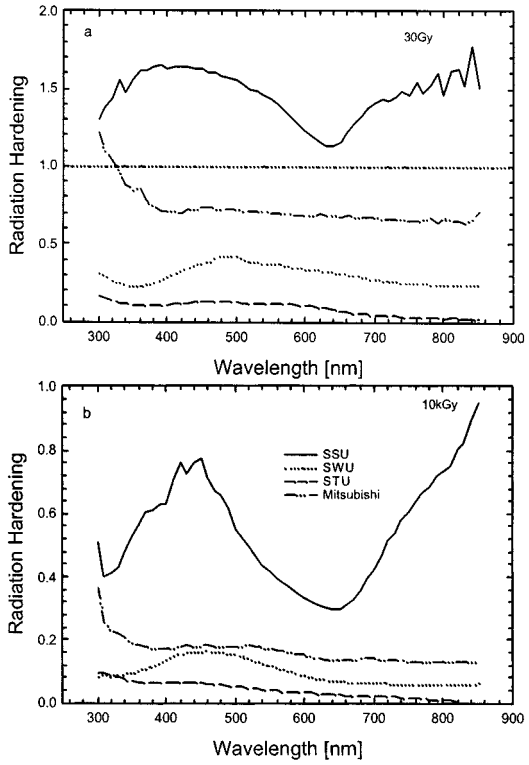


Fig. 18. Radiation hardening by H_2 treatment of the four MMSI fibers at dose values of (a) 30 Gy and (b) 10^4 Gy.

values of 30 Gy and 10^4 Gy. With increasing dose the H_2 treated STU fiber becomes superior in the whole wavelength range. Since the H_2 treatment has also improved the "initial" attenuation of the STU fiber to values comparable with those of the high OH SSU fiber [Figs. 5(b) and 6], H_2 (or D_2) treated STU fibers

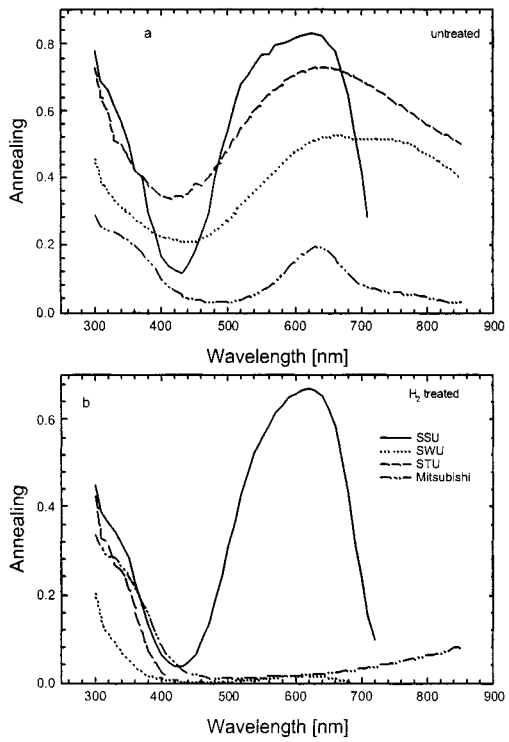


Fig. 19. Annealing (= ratio of the attenuation increase 10 h 45 min after the end and at the end of irradiation up to 10^4 Gy) of (a) untreated and (b) H_2 treated samples of the four MMSI fiber types.

are the best choice, e.g., for endoscopy in the visible wavelength range or as fiber optic "Cerenkov detectors" [10], where it is very essential to have good transmission at shorter wavelengths even after longer use (higher dose values) at an accelerator.

Fig. 18(a) and (b) compare the "radiation hardening" of the four fiber types by H_2 treatment at both dose values. All fibers show a higher improvement at the highest dose that might, however, decrease toward dose values of 10^5–10^6 Gy as indicated by some of the measurements at fixed wavelengths. Even the high OH SSU fiber shows distinctly improved radiation hardness at wavelengths $<$800 nm.

Fig. 19(a) and (b) compares the acceleration of the annealing of the four fiber types by H_2 treatment. Here annealing is defined as ratio of the radiation-induced attenuation 10 h 45 min after the end of irradiation and immediately before the end, at a dose of 10^4 Gy. All low OH fibers show significant improvement that might be the reason for their improved radiation hardness.

IV. Conclusion

Hydrogen (or deuterium) treatment consisting of H_2 (or D_2) loading under high pressure (125 bar) and immediate pre-irradiation up to optimized dose values, can considerably improve the radiation hardness of optical fibers with a pure SiO_2 core, especially of those with a low OH content. A factor of up to 50 was observed with the new Heraeus STU fiber type already showing superior radiation hardness of untreated fiber samples at high dose values [4]. Improved radiation hardness still could be observed during a second irradiation between six and 16 months later.

Hydrogen loaded fibers that were not pre-irradiated showed no improved radiation hardness when they were irradiated at least one month after the H_2 loading, i.e., after the H_2 has left the fiber. Pre-irradiation doses between 10 Gy and 10^3 Gy were not sufficient. A good compromise were dose values between 10^4 and $2 \cdot 10^4$ Gy for our investigated fiber types. Here the H_2 treatment even caused a distinct improvement of the "initial" attenuation of the low OH fibers at wavelengths $\leq$700 nm, compared with untreated fibers. At wavelengths $>$700 nm the H_2 treatment led to an increase of the initial attenuation, especially at the usual transmission wavelengths around 1300 and 1550 nm, caused by a strong growth of the OH absorption bands at 1380 nm and in the far IR, especially after a pre-irradiation dose of $2 \cdot 10^5$ Gy. This attenuation increase around 1300 nm and 1550 nm is higher than the radiation-induced attenuation of untreated fibers up to 10^6 Gy! Changing to deuterium (D_2) treatment reduces the increase of the initial attenuation considerably (if the H_2 contamination of the D_2 gas is low enough) and is at least as efficient as H_2 treatment in reducing the radiation-induced loss increase.

Most advantageous is H_2 treatment for increasing the radiation hardness in the visible wavelength range. A H_2 treated STU fiber shows a loss increase of only 0.1 to 0.001 dB/m in the wavelength range 350–800 nm even after a dose of 10^4 Gy ($=$1 Mrad). The reason might be the strongly accelerated loss annealing after the end of irradiation observed with H_2 treated low OH fibers. Therefore, H_2 or D_2 treated STU fibers are the best choice, e.g., for fiber optic endoscopes or fiber optic Cerenkov detectors at accelerators [10].

Since accelerated annealing might be the reason for the increase of radiation hardness, it should be investigated in more detail whether the observed radiation hardening will decrease toward lower dose rates.

Measurements during pre-irradiation, i.e., in the presence of H_2, showed extremely low radiation-induced loss values. Therefore, the best method for radiation hardening should be to capture a high amount of H_2 in a fiber—if possible. But this would only be useful for wavelength regions with negligible H and H_2 absorption bands.

Lyons and Looney explained the effect of H_2 treatment by a bonding of H atoms to color center precursors (Section I). Our observed increase of the OH content of low OH fibers and the fact that even untreated high OH SSU or KU fibers show good radiation hardness suggests that this increased OH content could be the reason for the improved radiation hardness. Therefore, it would be interesting to compare, e.g., the radiation sensitivity of Heraeus STU fibers of different OH content within the given margin of 1–20 ppm [4]. But increased OH content can not be the only reason for the improvement since also the high OH SSU fiber, with negligible increase of the OH content, shows improved radiation hardness (especially in the visible) and the H_2 treated STU fiber has nearly a factor of ten lower loss increase (in the visible at 10^4 Gy) than the SSU fiber, despite its much lower OH content.

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