



# Time dependence of silica optical properties during the implantation of fast hydrogen ions: Theory



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## ABSTRACT

Formation, excitation and passivation of defects by absorbed hydrogen have been extensively reported in the literature. Here we present a basic luminescence-diffusion model to simulate creation and chemical annealing behavior of non-bridging oxygen hole centers in silica by their treatment under a long-time hydrogen implantation. The model is in a good agreement with experimental data and explains the uncommon nonmonotonic time dependence of the non-bridging oxygen hole centers luminescence during the hydrogen implantation. The proposed model establishes the quantitative relation between the intensity dependence of luminescence on its intrinsic diffusivity, hydrogen concentration, defect concentration and cross-section of their creation. Possibilities to estimate these parameters based on the experimental data for the efficiency of silica luminescence are also discussed.

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## 1. Introduction

Balance of hydrogen in silica is very important for fabrication of optical fibers and modern interfaces [1]. Hydrogen saturation of silica can be performed by both implantation and thermodiffusion under hydrogen gas exposure of the sample.

There are a number of up-to-date models for formation, excitation and passivation of defects by absorbed hydrogen. The corresponding thermodiffusion process through the surface of bulk silica has been studied in [2] at different temperatures. Hydrogen can easily diffuse through the glass, however due to the reaction with the defect centers its effective  $D$  is smaller than the intrinsic one at the given temperature. The explanation of the experimental data has been also performed in [2] by solving the complete reaction–diffusion problem based on the hydrogen diffusivity, defects and hydrogen concentrations. In this case new defects are not formed and a chemical annealing of the existing defects is occurred. Let's note that the hydrogen implantation differs from the Tandon case of hydrogen gas exposure.

Along with the other methods ion implantation technique is widely used to develop novel functional materials based on silica

[3]. D. Fink et al. studied hydrogen diffusion induced by ion implantation by a special Nuclear Reaction Analysis often referred to as the “1 s N technique” [4]. The hydrogen implantation profiles were also measured in [4] and the hydrogen diffusion coefficients were calculated with «DIFFUS» code on the basis of the measured profiles.

The implantation is accompanied by formation, excitation and annealing of the defect structure in the matter. The defects can affect the performance of modern silica technologies (microelectronic devices, optical fiber communications and others) [2 and the references there in]. In particular, the understanding of radiation effects in silica-based materials is all-important for optical functional applications of materials [5].

Silica optical properties are to a considerable degree depend on the concentration and profile of different defect type [6]. It is well known that there are two types of intrinsic silica defects: ODC (oxygen deficiency center) and NBOHC (non-bridging oxygen hole center) (see [6] for example). It has been determined that in the visible spectrum range ODC and NBOHC are luminescent light source reaching the maximum intensity at 460 nm (2.7 eV) and 645 nm (1.9 eV), respectively.

According to [7,8] the hydrogen ion implantation changes luminescent spectra. At the present time, large experimental data array for defect formation under the ion beam exposure and concomitant ionoluminescence in silica has been obtained and models for defect

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formation and luminescent light generation have been suggested [9–11]. Defect formation occurs along the entire ion track resulting in luminescent light generation under the ion bombardment. Dynamics of luminescence efficiency are quite various at different regions of silica sample due to the opposite processes of defect formation and annealing. The chemical annealing of the oxygen hole centers by reacting them with hydrogen results in the formation of hydroxyl having a different absorption wavelength in comparison to NBOHC.

Luminescent light spectrum changes are associated with a defect balance taking into account hydrogen diffusion through the sample. In our case this diffusion toward the surface of the sample has radiation-enhanced character with diffusion coefficient larger than the one for the thermal case [4] especially for the higher absorption doses. Luminescent spectrum treatment not only permits to separate an influence of the defect on the luminescent spectrum, but also to obtain luminescent temporal characteristics under a long-time irradiation [12].

Long-time irradiation by the hydrogen ion causes changes in the luminescent radiation spectra of silica [5,9,13]. It was found, that the luminescent bands responsible for ODCs and NBOHCs change in different ways under a long-time irradiation since the implanted hydrogen ions interact with these defects in different ways. As mentioned above, several processes take place: not only the formation and excitation, but also modification and passivation of defects. These processes are closely connected and should be studied by solving the complete system of luminescence-diffusion equations. To determine the relation between luminescent radiation (LR) and defect formation processes one can use the model [8] for instance.

In this paper we develop a model of LR temporal changes associated with the cross-sections of the above-mentioned processes and a hydrogen diffusion from the implanted layer. By solving the complete luminescence-diffusion problem, we are looking to explain the experimental data and the uncommon nonmonotonic time dependence of luminescence during the hydrogen implantation observed in [14]. We also determine parameters, which can be estimated based on the LR time dependence for different combinations of defect concentration and the hydrogen ions flux density.

## 2. Theoretical models and results

### 2.1. Dynamics of hydrogen implantation

We consider the time dependence of hydrogen atoms concentration  $n_H(\vec{r}, t)$  during the implantation process. As noted in [4], in the case of low temperature and low-fluence implantation the depth distributions of energetic ions implanted into solids are accurately determined by the dedicated computer codes like SRIM/TRIM [15]. At high fluence implantation the secondary effects such as radiation enhanced mobility become important and lead to same deviations from the well-known distributions [4]. As a result, the dynamical equation for time dependence on hydrogen concentration should contain two major terms. The first one is to describe the initial local accumulation of hydrogen atoms in the sample of silica and the second one to account for their simultaneous diffusion. In the most simplified model of the one-dimensional implantation, which agrees well with experiments in [7], the proper dynamic diffusion equation can be written in the following form

$$\frac{\partial n_H(x, t)}{\partial t} = D_H \frac{\partial^2 n_H(x, t)}{\partial x^2} + (2\Delta R_p)^{-1} \phi [\theta(x - R_p + \Delta R_p) - \theta(x - R_p - \Delta R_p)], \quad (1)$$

where  $x$  is the coordinate along the ion beam axe,  $D_H$  is the diffusion coefficient of hydrogen,  $\phi$  is the flux density of ions,  $\theta(x)$  is the

Heaviside function and  $\Delta R_p$  is the width of initial hydrogen distribution so that the region of initial hydrogen accumulation due to the implantation is defined by the two-sided inequality

$$R_p - \Delta R_p \leq x \leq R_p + \Delta R_p.$$

Taking into account the well-known ratio [4,16]

$$\Delta R_p/R_p \ll 1, \quad (2)$$

which is satisfied for high-energy ions, the realistic distribution profile can be neglected and is replaced by the difference of the Heaviside functions [11] giving the uniform distribution in the region of initial localization, as shown in Fig. 1a.

For hydrogen implementation (1) is a subject to the initial and boundary conditions as follows:

$$n_H(x, 0) = 0, \quad (3)$$

$$n_H(x, t)|_{x=0} = 0, \quad \partial n_H(x, t)/\partial x|_{x=R_p+\Delta R_p} = 0. \quad (4)$$

We assume here that the ions are implanting through the sample surface with coordinate  $x = 0$ , at which the concentration of hydrogen is constantly zero due to vaporization. The boundary conditions can also be simplified at  $x = R_p + \Delta R_p$ , where the flux of hydrogen is negligibly small since the radiation-enhanced diffusion coefficient  $D_H$  is very large in comparison with the one of the thermal hydrogen mobility in the area  $x > R_p + \Delta R_p$ . In other words, the leak of the hydrogen into the unirradiated area is very small, as it is proved below.

Conditions (3) and (4) result in the following solution of Eq. (1) [17]:

$$n_H(x, t) = \sum_{n=0}^{\infty} \left[ \int_0^t e^{-D_H \left[ \frac{\pi}{2(R_p + \Delta R_p)} \right]^2 (2n+1)^2 \tau} f_n d\tau \right] \sin \frac{\pi(2n+1)}{2(R_p + \Delta R_p)} x, \quad (5)$$

where

$$\begin{aligned} f_n &= \frac{1}{R_p + \Delta R_p} \Delta R_p^{-1} \phi \int_0^{R_p + \Delta R_p} [\theta(x - R_p + \Delta R_p) - \theta(x - R_p - \Delta R_p)] \\ &\quad \times \sin \frac{\pi(2n+1)}{2(R_p + \Delta R_p)} x dx \\ &= \frac{1}{R_p + \Delta R_p} \Delta R_p^{-1} \phi \int_{R_p - \Delta R_p}^{R_p + \Delta R_p} \sin \frac{\pi(2n+1)}{2(R_p + \Delta R_p)} x dx. \end{aligned}$$

Due to the strong inequality (2) the coefficients  $f_n$  can be rewritten as

$$f_n = \frac{2}{R_p} \phi \sin \left( \frac{\pi}{2} + \pi n \right) = \frac{2}{R_p} \phi (-1)^n. \quad (6)$$

Substituting (6) into (5) and performing the integration with respect to  $\tau$  gives

$$\begin{aligned} n_H(x, t) &= \frac{2\phi}{R_p D_H} \sum_{n=0}^{\infty} (-1)^n \left[ \frac{2R_p}{\pi(2n+1)} \right]^2 \\ &\quad \times \sin \frac{\pi(2n+1)}{2R_p} x \cdot \left[ 1 - e^{-D_H \left( \frac{\pi}{2R_p} \right)^2 (2n+1)^2 t} \right] \end{aligned} \quad (7)$$

As follows from the Eq. (7), the general dependence of the hydrogen distribution on time of the implantation  $t$  is exponential and in the considered case the diffusion time scale can be defined as

$$t_D = D_H^{-1} \left( \frac{2R_p}{\pi} \right)^2. \quad (8)$$

In accordance with (7) the flux density of ions governs only the magnitude of the hydrogen distribution. For  $t \gg t_D$  the final distribution can be obtained directly from the Eq. (7):

$$n_H(x) = \frac{2\phi}{R_p D_H} \sum_{n=0}^{\infty} (-1)^n \left( \frac{2R_p}{\pi} \right)^2 \frac{\sin(2n+1)\frac{\pi x}{2R_p}}{(2n+1)^2}. \quad (9)$$

The sum of the functional series in (9) is tabulated [18], so that the final distribution is given by the following formula:

$$n_H(x) = \frac{\phi}{D_H} x. \quad (10)$$

This result, shown in Fig. 1b, has a clear physical meaning – the equilibrium state is established when the radiation induced afflux of hydrogen atoms is compensated by the leak due to their diffusion and further vaporization from the surface of the sample. The total number of hydrogen atoms per  $\text{m}^2$  in the irradiated part of the sample is given by the expression:

$$N_H = \int_0^{R_p + \Delta R_p} \frac{\phi}{D_H} x dx = \frac{\phi(R_p + \Delta R_p)^2}{2D_H} = \frac{1}{2} \left( \frac{\pi}{2} \right)^2 \phi t_D. \quad (11)$$

In the general case, other processes, such as diffusion into the unirradiated bulk of sample, can also contribute to the leak of hydrogen. The proper dynamic diffusion equation and the boundary condition for unirradiated bulk can be written in the following form:

$$\frac{\partial n_H^{(u)}(x, t)}{\partial t} = D \frac{\partial^2 n_H^{(u)}(x, t)}{\partial x^2}, \quad (12)$$

$$n_H^{(u)}(R_p + \Delta R_p, t) = n_H(R_p + \Delta R_p, t), \quad (13)$$

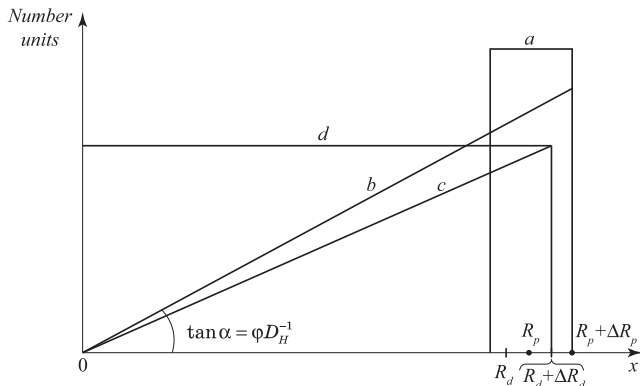
where  $D$  is the diffusion coefficient of the thermal hydrogen mobility. The boundary condition (13) stands for the continuity of the hydrogen's distribution function and means that the irradiated part of the sample represents an infinite source of hydrogen for the unirradiated bulk.

As a rule, the linear size of the unirradiated area is much larger than the representative value  $R_p$  of the irradiated area. Taking this into account, we use a well-known solution [17] for the semi-infinite space, which is defined by the following integral expression:

$$n_H^{(u)}(x, t) = \frac{D}{2\sqrt{\pi}} \int_0^t \frac{x - R_p - \Delta R_p}{[D(t - \tau)]^{3/2}} e^{-\frac{(x - R_p - \Delta R_p)^2}{4D(t - \tau)}} n_H(R_p + \Delta R_p, \tau) d\tau. \quad (14)$$

Integrating (14) with respect to  $x$  gives us the number of hydrogen atoms per  $\text{m}^2$ :

$$N_H^{(u)}(t) = \sqrt{\frac{D}{\pi}} \cdot \int_0^t \frac{n_H(R_p + \Delta R_p, \tau)}{\sqrt{t - \tau}} d\tau. \quad (15)$$



**Fig. 1.** Space distributions in the silica irradiated by ion beam: (a) hydrogen concentration for initial model; (b) final equilibrium distribution of hydrogen; (c) d) model damage distributions governed by two different cross-sections of the radiation production of NBOHCs.

Using Eq. (15) and involving expression (10) one can estimate the number of atoms as follows:

$$N_H^{(u)}(t) < \sqrt{\frac{D}{\pi}} \frac{\phi(R_p + \Delta R_p)}{D_H} \int_0^t \frac{d\tau}{\sqrt{t - \tau}} = 2\sqrt{\frac{Dt}{\pi}} \frac{\phi(R_p + \Delta R_p)}{D_H}. \quad (16)$$

Now we determine a time of the implantation  $t$ , at which the leak of the hydrogen into the unirradiated area is very small:  $N_H^{(u)}(t) \ll N_H$ . Using Eqs. (6) and (11) we obtain

$$t \ll \left( \frac{\pi}{4} \right)^3 \frac{D_H}{D} t_D.$$

Obviously, the latter condition is easily satisfied experimentally because the ratio  $D_H/D$  is of the order of  $10^3 - 10^4$  as it was shown in [4].

## 2.2. Dynamics of LR

Specific character of LR is conditioned by the fact that the luminescence centers are both created and excited by the radiation energy that can be deposited from the nuclear collisions or delivered by ionization stopping. This means that the physical mechanisms responsible for the defect production and luminescence excitation at any instant can differ essentially. As a rule the exciton luminescence in silica dioxide is treated as a result of the electronic excitation mainly [11]. Nevertheless, independently on the nature of the defect production and luminescence excitation, the following permanent factors are the most important in the explanation of the luminescence excited by hydrogen ions: creation of luminescence centers due to the radiation production of defects, for example in the shape of NBOHCs, and a passivation of non-bridging oxygen by atoms of hydrogen. Hence, the description of the luminescence dynamics can be done by using the kinetic equation

$$\frac{\partial n_o(x, t)}{\partial t} = \{n_2 - n_o(x, t)\} \sigma(x) \phi - kn_H(x, t)n_o(x, t) \quad (17)$$

and the initial condition

$$n_o(x, 0) = n. \quad (18)$$

Here,  $n_2$  is the total concentration of oxygen atoms,  $n_2 - n_o$  is the concentration of target oxygen atoms,  $\sigma(x)$  is the cross-section of the radiation production of non-bridging oxygen,  $k$  is the reaction rate constant for non-bridging oxygen passivation by the hydrogen and  $n$  is the initial concentration of non-bridging oxygen atoms in the silica sample. The cross-section in Eq. (17) depends on the coordinate  $x$  due to the dependence on the ion energy that changes as a function of the ion path. In the absence of the passivation term, (17) coincides with the usual equation for the radiation induced reactions [16].

The interpretation of LR dynamics is more complicated comparing with the hydrogen implantation because of the mentioned two-staged process. The intrinsic feature of the first stage is the negligible effect of passivation during the implantation time that we define for convenience as  $t < t_D$ . Contrary to the first stage, which is characterized by increasing of luminescence due to the radiation damage, the second stage, when  $t > t_D$ , may lead to the inhibition of the local luminescence.

To simplify the model we assume that for  $t < t_D$  there is no hydrogen diffusion and passivation of NBOHCs, whereas for  $t > t_D$  the final hydrogen distribution (10) sets up immediately. Under these assumptions the solution of Eq. (17) takes the form

$$n_o(x, t) = \begin{cases} n_2 + (n - n_2)e^{-\sigma(x)\phi t}, & 0 \leq t \leq t_D \\ n_2 \frac{\sigma(x)}{\sigma(x) + kD_H^{-1}x} + [n_o(x, t_D) - n_2 \frac{\sigma(x)}{\sigma(x) + kD_H^{-1}x}]e^{-[\sigma(x)\phi + k\phi D_H^{-1}x](t - t_D)}, & t > t_D \end{cases} \quad (19)$$

The limit of  $n_o(x, t)$  when  $t \rightarrow \infty$ , i.e. the saturation level of NBOHC concentration, equals to

$$n_o(x, \infty) = n_2 \frac{\sigma(x)}{\sigma(x) + kD_H^{-1}x} < n_2,$$

which is less than the saturation value  $n_2$  in the absence of passivation. Moreover, the saturation value may become lower than concentration of NBOHCs for  $t = t_D$  or even for  $t = 0$  resulting in the nonmonotonic dependence of the local concentration on time, as shown in Fig. 2 for two different values of  $n_o(x, \infty)$ .

For realistic values of ion flux density and cross-section of the radiation defects production, the strong inequality  $\sigma(x)\phi t_D \ll 1$  is satisfied giving the linear time dependence of solution (19) for  $t < t_D$ . Hence, the local concentration at the moment of time  $t = t_D$  is larger than  $n_o(x, \infty)$  if

$$n + n_2\sigma(x)\phi t_D > n_2 \frac{\sigma(x)}{\sigma(x) + kD_H^{-1}x}. \quad (20)$$

Finally,  $I(x, t)$ , the luminescence efficiency along the track at any instant, is a product of the NBOHCs concentration (19), which is formed by the processes of hydrogen diffusion and further passivation of luminescence centers, and the excitation efficiency  $i(x)$ :  $I(x, t) = i(x)n_o(x, t)$ . As a rule [10,11], the full width for exciton production is assumed to be larger and smoother than the one for damage. If so, the excitation density profile may be approximated for simplicity by the uniform distribution  $i(x) = i$ . Then the total output luminescence per  $m^2$  is the integral of the luminescence over the irradiated region:

$$I(t) = iS \int_0^{R_p + \Delta R_p} n_o(x, t) dx = iSN_o(t), \quad (21)$$

where  $N_o(t)$  is the number of non-bridging oxygen per  $m^2$  and  $S$  is the area of the sample. Thus, the number of defects determines the dynamics of luminescence.

### 3. Discussion

The shape of the rise curve conforms to a model of luminescence at a simple exponential rate (see Fig. 2, solid curve). The present dynamic model of the hydrogen implantation and LR assumes a nonmonotonic time dependence of the luminescence observed in

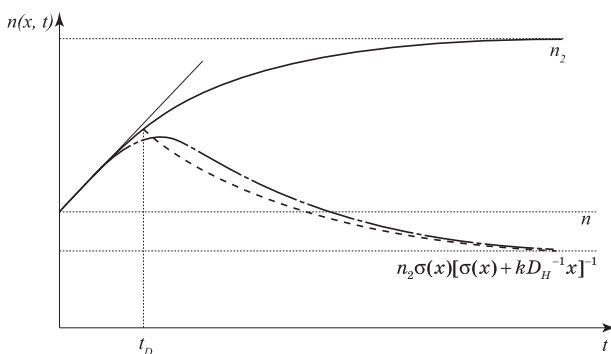


Fig. 2. Time dependence of the local NBOHC concentration: monotonic (solid line) and nonmonotonic (dashed line) dependence on time in the absence and the presence of hydrogen diffusion, respectively; realistic rise curve (dash-dot line).

silica [14]. According to the Eqs. (20) and (21), the nonmonotonic rise of luminescence will be observed, if

$$N + N_2\phi t_D(R_p + \Delta R_p)^{-1} \int_0^{R_p + \Delta R_p} \sigma(x) dx > N_2(R_p + \Delta R_p)^{-1} \int_0^{R_p + \Delta R_p} \frac{k^{-1}D_H\sigma(x)}{k^{-1}D_H\sigma(x) + x} dx,$$

where  $N = N(0) = n(R_p + \Delta R_p)$  and  $N_2 = n_2(R_p + \Delta R_p)$ . As expected this inequality is easily satisfied in the limiting case, when the passivation reaction rate constant is infinitely large, i.e.  $k^{-1} \rightarrow 0$ .

Based on the available experimental data the developed model allows estimation of the parameters necessary to describe the processes of hydrogen implantation and LR in silica. For example, the Eq. (8) can be rewritten in the form

$$D_H = \left(\frac{2R_p}{\pi}\right)^2 t_D^{-1}, \quad (22)$$

giving the estimated value of the diffusion coefficient, which is defined by the time  $t_D$  corresponding to the peak luminescence.

Furthermore, exploiting the strong inequality  $\sigma(x)\phi t_D \ll 1$  the following ratios can be derived directly from Eq. (19):

$$\frac{N(t_D)}{N} = 1 + \frac{N_2}{N}\phi t_D(R_p + \Delta R_p)^{-1} \int_0^{R_p + \Delta R_p} \sigma(x) dx = 1 + \frac{N_2}{N}\bar{\sigma}\phi t_D, \quad (23)$$

$$\frac{N(\infty)}{N} = \frac{N_2}{N}(R_p + \Delta R_p)^{-1} \int_0^{R_p + \Delta R_p} \frac{k^{-1}D_H\sigma(x)}{k^{-1}D_H\sigma(x) + x} dx. \quad (24)$$

Thus, (23) gives us the information about  $\bar{\sigma}$  the average value of the cross-section, if the initial portion of defects  $N/N_2$  is known. At the same time the right-hand side of the Eq. (24) only depends on the product  $k^{-1}D_H\sigma(x)$ , which may be used to estimate the reaction rate constant  $k$ .

A more accurate analysis of the experimental data is required as it could be explained in terms of the variation in the value of the cross-section  $\sigma(x)$ . The initial increase in luminescence intensity takes place mainly due to the build up of defects by nuclear collisions in a region centered around  $R_d$ , where the light ions are brought to rest [9]. The value of  $R_d$  is slightly smaller than  $R_p$ , i.e. closer to the surface. At the same time there is a small contribution of defects created and retained in the region where the ions are traveling at high energy. In this region the defect production rates from the electronic events and nuclear collisions may be comparable, although the efficiency of electronic events in producing damage is three orders of magnitude less than in the nuclear collisions [19–21]. However, by the time the average ion energy is reduced down to 100 keV the nuclear damage rate will be 10 times larger than the electronic one rising monotonically when approaching to  $R_d$ .

For these reasons the monotonic increase of the cross-section through the projected range may be roughly approximated by the linear function for  $H^+$  and  $H_2^+$  ions with the energies of about 200 and 400 keV, respectively. In this approximation, shown in Fig. 1c, the cross-section of the radiation production of NBOHCs takes the form:

$$\sigma(x) = \begin{cases} \sigma x / (R_d + \Delta R_d), & 0 \leq x \leq R_d + \Delta R_d \\ 0, & R_d + \Delta R_d < x \end{cases},$$

where  $\sigma = 2\bar{\sigma}$ . Substituting of this equation into (24) and integrating give

$$\frac{N(\infty)}{N} = \frac{N_2 R_d + \Delta R_d}{N R_p + \Delta R_p} \frac{2k^{-1} D_H \sigma}{2k^{-1} D_H \sigma + R_d + \Delta R_d}.$$

Taking into account the evident strong inequality  $N_2/N \gg 1$  we have

$$k = \frac{D_H \sigma}{R_d + \Delta R_d} \left[ \frac{N}{N(\infty)} \frac{N_2 R_d + \Delta R_d}{N R_p + \Delta R_p} - 1 \right] \cong \frac{N}{N(\infty)} \frac{N_2 D_H \sigma}{R_p}. \quad (25)$$

Another approach may be used for implantation of  $H^+$  and  $H_2^+$  ions with energies of about 50 and 100 keV, respectively. According to [20], the reasonable assumption for these beam conditions is that the energy deposited from the nuclear collision events is relatively uniform throughout the projected range. As a consequence, the cross-section can be chosen as a constant value equal to  $\sigma(x) = \sigma = \bar{\sigma}$  (see Fig. 1d). In this case we have the transcendental equation

$$\frac{N}{N(\infty)} \frac{N_2 R_p + \Delta R_p}{R_d + \Delta R_d} X = \ln(1 + X), \quad X = \frac{k(R_d + \Delta R_d)}{D\bar{\sigma}}, \quad (26)$$

which can be numerically solved with respect to  $k$  in each particular case. At that the lower limit of the reaction constant can be easily estimated as follows:

$$X = \frac{N}{N(\infty)} \frac{N_2 R_d + \Delta R_d}{N R_p + \Delta R_p} \ln(1 + X) > \frac{N}{N(\infty)} \frac{N_2 R_d + \Delta R_d}{N R_p + \Delta R_p},$$

so that

$$k > \frac{N}{N(\infty)} \frac{N_2 D_H \sigma}{R_p}. \quad (27)$$

Note, that the values of the  $R_p$  parameter are different in (25) and (27) since it depends on the initial velocity of ions [16].

#### 4. Summary

We have presented a basic luminescence-diffusion model to simulate the creation and chemical annealing behavior of non-bridging oxygen hole centers in silica by their treatment with hydrogen under a long-time hydrogen implantation. This model can also be used to simulate the transport of hydrogen through the sample of silica accounting for the leak of hydrogen into unirradiated part of the sample. The predictions based on the model are in a good agreement with the experimental data presented in [14] and explain the uncommon nonmonotonic time dependence of luminescence for the non-bridging oxygen hole centers during the hydrogen implantation. The proposed model establishes the

quantitative relation between the dependence of intensity of luminescence on its intrinsic diffusivity, hydrogen concentration, defect concentration and cross-section of their creation. Estimation of these parameters based on the experimental data for the luminescence efficiency is discussed.

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