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RADIATION DAMAGE KINETICS IN PWO CRYSTALS

A. Annenkov [1], E. Auffray [2], A. Fedorov[4], S. Gninenko [3], N. Golubev[3],

M. Korzhik[4], P. Lecoq[2], V. Ligun[1], A. Lobko[4], O. Missevitch[4],

J.-P. Peigneux[5], Yu.D. Prokoskin[6], A. Singovski[6].

1) *BTCP, Bogoroditsk, Russia* - 2) *CERN, Geneva, Switzerland*

3) *JNR, Moscow, Russia* - 4) *INP, Minsk, Belarus*

5) *LAPP, Annecy-le-Vieux, France* - 6) *IHEP, Protvino, Russia*

Abstract

This note proposed a model to describe the kinetics of the irradiation damage in PWO crystal and apply it to two well-known measurement methods of the optical transmission damaged by irradiation.

1. Introduction

It is known that in some PWO crystals damage to their optical properties appears under ionizing radiation. A decrease of the optical transmission and then of light yield has been detected. We have determined that the level of damage strongly depends on the peculiarities of the crystal growth technology. However, the magnitude varies from crystal to crystal even for crystals grown from purified raw materials. This suggests the existence of several contributors to the damage process which are mainly structural defects. The amount of such defects is influenced by even small technological changes. Although the origin of these defects may differ, they can be characterized by some universal set of parameters which describes their kinetics under irradiation. Thus motivated, we have analysed within a phenomenological model the transmission radiation damage in the PWO crystals. The model does not include detailed background about peculiarities of free carriers capture under irradiation and recovery mechanisms of the damage centres. However, the model describes well the kinetics of the damage and gives the values of the parameters needed to estimate the level and the relative influence of different damage centres in the crystals. Using this model we describe and analyse here two well-known measurement methods of the transmission damage by irradiation. A similar approach has already been used by others [1] to analyse optical bleaching in BaF_2 crystals, but in this case transmission and light output damage were not dependent on dose rate.

2. The model

Radiation damage in the crystals appears to be due to a charge-state change of the existing point structure defects. This means that creation of new defects due to inelastic scattering of damaging particles is negligible and that the concentration of impurities is considerably smaller than the amount of host defects. The radiation damage process is therefore driven by the change of the defects' electronic states which is at the origin of production of colour centres and the creation or suppression of their associated absorption bands. Let us suppose that the defects are randomly distributed in the crystal, that the interaction between them is negligible, and that the amount of each type of defect *i* is limited and considerably smaller than the amount of the lattice atoms. In this case the radiation damage will reach a saturation level after a certain time which is determined by the amount of pre-existing defects. Thus, the amount of damaged centres of type *i* is described by the following differential equation:

$$\frac{dN_i}{dt} = -\omega_i N_i + \frac{S}{d_i}(N_i^* - N_i) \; , \tag{1}$$

where N_i is the amount of damaged centres of type *i* at the time *t*, ω_i is the recovery rate of damaged centres of type *i* , *S* is the rate of irradiation, N_i^* is the amount of pre-existing defects of type i , and di is the damage constant of the mentioned centres which depends on the capture cross-section of free carriers by the centres of type *i*. Summing all types of centres we get the following equation:

$$\frac{dN}{dt} = -\sum_i \omega_i N_i + S\sum_i \frac{1}{d_i}(N_i^* - N_i) \; , \tag{2}$$

where $N = \sum_i N_i$ is the total amount of different types of damaged centres. Precise processing of the equation requires the values of the constants ω_i, N_i^*, d_i but the simultaneous determination is rather difficult and a practical solution can only be reached through approximations.

First, we can suppose that all centres have very similar parameters so that they can be characterized by average constants and by the total amount of centres N^*. Thus, Eq. (2) can be simplified as

$$\frac{dN}{dt} = -\omega N + \frac{S}{d}(N^* - N) \ . \tag{3}$$

This approximation is acceptable in the rare situation where one sort of damaged centre dominates.

In most of the cases of PWO, we observe two recovery time constants, which suggest two types of damaged centres. This is what we call the *fast-slow* approximation.

$$\frac{dN}{dt} = -(\omega_{\text{fast}} N_{\text{fast}} + \omega_{\text{slow}} N_{\text{slow}}) + S[\frac{1}{d_{\text{fast}}}(N^*_{\text{fast}} - N_{\text{fast}}) + \frac{1}{d_{\text{slow}}}(N^*_{\text{slow}} - N_{\text{slow}})] \ , \tag{4}$$

where $N = N_{\text{fast}} + N_{\text{slow}}$.

Equation (4) can be separated into two equations:

$$\frac{dN_{\text{fast}}}{dt} = -\omega_{\text{fast}} N_{\text{fast}} + S\ [\frac{1}{d_{\text{fast}}}(N^*_{\text{fast}} - N_{\text{fast}})] \ , \tag{5}$$

$$\frac{dN_{\text{slow}}}{dt} = -\omega_{\text{slow}} N_{\text{slow}} + S\ [\frac{1}{d_{\text{slow}}}(N^*_{\text{slow}} - N_{\text{slow}})] \ . \tag{6}$$

The *fast-slow* approximation is valid when the amount of each type of damaged centre is limited and if their recovery times differ by at least one order of magnitude. This corresponds to the observed situation of the recovery of damaged centres in PWO as described in Ref. [2]. From this approximation one can understand the influence of the fast and slow saturating centres on the kinetics of the PWO damage induced absorption.

Equations (3), (5), and (6) have similar solutions. For averaged parameters

$$N = N^* \frac{S}{S+\omega\ d}\{1 - \exp[-(\omega + \frac{S}{d})t]\} \tag{7}$$

and in the *fast -slow* approximation

$$N = N^*_{\text{fast}} \frac{S}{S + \omega_{\text{fast}} d_{\text{fast}}}\{1 - \exp[-(\omega_{\text{fast}} + \frac{S}{d_{\text{fast}}})t]\} + N^*_{\text{slow}} \frac{S}{S + \omega_{\text{slow}} d_{\text{slow}}}\{1 - \exp[-(\omega_{\text{slow}} + \frac{S}{d_{\text{slow}}})t]\} \ . \tag{8}$$

If irradiation is stoped the recovery of damaged centres of type *i* is described by the following equation:

$$\frac{dN_i}{dt} = -\ \omega_i N_i(t_0) \ , \tag{9}$$

where $N_i(t_0)$ is the amount of the damaged centres of type *i* at the time t_0 (irradiation time) at the end of the irradiation period.

The solution of this simple equation is, in the case of averaged parameters,

$$N = N^* \frac{S}{S+\omega\ d}\{1 - \exp[-(\omega + \frac{S}{d})t_0]\}\exp(-\omega\ t) \ , \tag{10}$$

or in that case of *fast-slow* approximation:

$$N = N^*_{\text{fast}} \frac{S}{S+\omega_{\text{fast}} d_{\text{fast}}}\{1 - \exp[-(\omega_{\text{fast}} + \frac{S}{d_{\text{fast}}})t_0]\}\exp(-\omega_{\text{fast}} t) + N^*_{\text{slow}} \frac{S}{S+\omega_{\text{slow}} d_{\text{slow}}}\{1 - \exp[-(\omega_{\text{slow}} + \frac{S}{d_{slow}})t_0]\}\exp(-\omega_{\text{slow}} t) \ . \tag{11}$$

3. Variable dose rate method

The induced absorption coefficient k produced by irradiation is proportional to the amount of absorbing centres N through $k = \sigma N$, where σ is the cross-section of the absorbing centre. Expressions (7 and 8) can be rewritten for the induced absorption as

$$k = k_{\rm sat} \frac{S}{S+\omega d}[1 - \exp[-(\omega + \frac{S}{d})t] \qquad (12)$$

and

$$k = k_{\rm sat}^{\rm fast} \frac{S}{S+\omega_{\rm fast} d_{\rm fast}}\{1 - \exp[-(\omega_{\rm fast} + \frac{S}{d_{\rm fast}})t]\} + k_{\rm sat}^{\rm slow} \frac{S}{S+\omega_{\rm slow} d_{\rm slow}}\{1 - \exp[-(\omega_{\rm slow} + \frac{S}{d_{\rm slow}})t\} , \qquad (13)$$

where $k_{sat} = N * \sigma$ is the saturated induced absorption coefficient when all centres are damaged.

Therefore, the parameters $k_{sat}^{fast,slow}$, $\omega_{fast,slow}$, $d_{fast,slow}$ which characterize the radiation damage can be determined by the measurement of the induced absorption at different S values.

4. Transmission recovery method

In the two above-mentioned approximations the recovery of the transmission after the end of irradiation at time t_0 is described by the following equations:

$$k = k_{\rm sat} \frac{S}{S+\omega d}\{1 - \exp[-(\omega + \frac{S}{d})t_0]\} \exp(-\omega \; (t - t_0)) \qquad (14)$$

$$k = k_{\rm sat}^{\rm fast} \frac{S}{S+\omega_{\rm fast} d_{\rm fast}}\{1 - \exp[-(\omega_{\rm fast} + \frac{S}{d_{\rm fast}})t_0]\} \exp(-\omega_{\rm fast}(t - t_0)) + k_{\rm sat}^{\rm slow} \frac{S}{S+\omega_{\rm slow} d_{\rm slow}}\{1 - \exp[-(\omega_{\rm slow} + \frac{S}{d_{\rm slow}})t_0]\} \exp(-\omega_{\rm slow}(t - t_0)) . \qquad (15)$$

This method also gives access to the required constants. However, in this case the crystal under investigation has to be irradiated several times with different accumulated irradiation doses.

5. Discussion

The two above methods for the measurement of radiation damage are equivalent. However, the first one is faster especially in the case of slow recovery. To make numerical calculations of the induced absorption in PWO crystals we have used the *fast-slow* approximation in case of variable dose rate. Let us estimate the values of the constants. The value of the d parameter depends on the absorption wavelengths of the defect and varies from 1500 to 100 rad [2]. For the numerical calculation we will use the average value of 800 rad for slow and fast recovery centres. Measurements on several crystal give recovery time constants of 3 h (180 min) for medium slow, 31 days (44 640 min) for very slow components [3]. A very fast recovery time constant of about 1 min is added in the analysis to study the influence of very shallow traps. To model the induced absorption, a saturation coefficient of $k_{sat} = 5\ m^{-1}$ at 500 nm was chosen for all centres and dose rates of 0.01 rad/min, 0.3 rad/min and 300 rad/min were selected to match the dose rates at X1, and H4 test beam lines and the Geneva Hospital ^{60}Co source, respectively.

Figures 1-3 show the induced absorption at different rates of irradiation for a crystal having fast, medium slow and very slowly recovering centres. At a low irradiation rate (0.01 rad/min) fast recovering centres do not contribute practically to the induced absorption, however, medium slow recovering centres reach a plateau and the main contribution to the induced absorption appears to be due to very slowly recovering centres which also saturated at longer times. At an irradiation rate of 0.3 rad/min the level of induced absorption increases for all centres, but the shape of the curves looks the same. At high irradiation rate (300 rad/min) all curves show saturation, but their values are different. Only the medium slow and the very slowly recovering centres reach the saturation level k_{sat}. This can be understood by the analysis of the amplitude in Eq. (13) which depends on the ratio of S and ω in the following way: $k = k_{sat} \frac{S}{S + \omega d}$. If $S >> \omega d$, in the case of a high rate of irradiation or very slow recovery, $k \approx k_{sat}$. On the contrary, if $S \leq \omega d$, k becomes smaller than $k_{sat}/2$ and falls with the increase of the recovery time or the d value. Thus, the value of the detected damage in the crystals depends mainly on the ratio of the irradiation rate and recovery time of the damaged centres. The main contributors to the induced absorption are the slowly recovering centres.

This conclusion is clearly illustrated by the correlation of the detected damage in H4 for the crystals N1266, 1283, 1318 and their transmission damage recovery times after the ^{60}Co source irradiation. Fig. 4 presents the recovery curves of these crystals after Co irradiation. The crystal N1318, which showed only 5% damage in the beam, contains faster recovering centres, however N1266 and 1283 with 20% and 8% damage level have considerably slower recovering centres [4].

Let us estimate now the influence of the defects on the induced absorption of PWO crystals with a damage constant of about 800 rad and a total density of defects leading to an absorption of 5 m^{-1} at full saturation. If we assume that the acceptable level of the induced absorption coefficient is 0.01 m^{-1}, this leads to a change of the transmission of 0.5% through a scintillator length of 0.23 m with an initial absorption length of 2 m. At the dose rate of 5 rad/min which is close to the real situation in the CMS ECAL, after 1000 min of irradiation fast recovering centres contribute according to Eq. (13) to the induced absorption for 0.03 m^{-1}, medium slow recovering centres produced an absorption of 2.7 m^{-1} and slowly recovering centres are saturated at a level of 5 m^{-1}. This indicates that even fast recovering centres contribute too much and have to be suppressed.

However, very fast recovering centres do not contribute significantly to the induced absorption under irradiation. Figure 5 shows the comparison of the contributions of defects with recovery times of 1 minute, 1 second and 1 millisecond at the dose rate 5 rad/min. One can see that compared to the minute's recovery, the second and millisecond defects do not contribute to a measurable absorption. It also means that very fast recovering centres which are the source of slow components in scintillation and afterglow do not contribute to the radiation damage.

The temperature also plays an important role in the radiation damage and the recovery kinetics. Decreasing the crystal temperature reduces the relaxation probability of the traps following an $\exp(-E_T/kT)$, where E_T is trap activation energy and T the absolute temperature. It therefore reduces the recovery probability of the damaged centres. This process is shown in Fig. 6 for crystal N1119 which has been irradiated at room temperature as well as at 0°C with a dose rate of 300 rad/min. Usually different types of damaged centres have different concentrations in the crystals and the k_{sat} parameters are also not the same. Thus the curve of the transmission damage shows a fast saturating regime followed by a long growing tail due to slowly recovering centres.

The proposed model leads to the following conclusions.

a) Very fast recovering centres with $1/\omega \sim$ seconds or less do not contribute practically to the induced absorption at the irradiation rate up to a few rad/min. It means that in the PWO scintillators to be used in the CMS ECAL their suppression is not required.

b) Recovering centres with 1/ω ~ minutes already contribute to the induced absorption at the irradiation rate of a few rad/min. Moreover their saturation level is very much dependent on the irradiation dose rate, which makes the crystal unstable under variable beam conditions. They have to be suppressed.

c) The main contributors to the induced absorption are the centres with slow recovery time and their presence in the PWO scintillators has to be minimized as much as possible. They are responsible for the accumulation of the damage and for the permanent degradation of the crystal transparency.

References

[1] D.A. Ma, R.Y. Zhu and H. Newnan, NIM A356 (1995) 309.

[2] A. Annenkov, S. Baccaro, B. Borgia, F. Cavalari et al., "Vacuum annealing and radiation hardness of PWO crystasls" (to be published).

[3] A. Lobko, B. Borgia, M. Korzhik et al. (to be published).

[4] E. Auffray "Results on radiation damage of PWO crystals". 22 CCC Meeting, CERN, Geneva, October 18, 1996.

Figure captions

Fig. 1 Induced absorption in PWO crystals due to different centre recovery at a low rate of irradiation, 0.01 rad/min. Change of the 0.23 m scintillator's transmission with initial absorption length of 2 m, after 1000 min of irradiation is 0.004% due to fast; 0.5% due to medium slow; 3% due to very slowly recovering centres.

Fig. 2 Induced absorption in PWO crystals due to different centre recovery at an irradiation rate of 0.3 rad/min. Change of the 0.23 m scintillator transmission with initial absorption length of 2 m, after 1000 min of irradiation is 0.1% due to fast; 17% due to medium slow; 55% due to very slow recovering centres.

Fig. 3 Induced absorption in PWO crystals due to different recovering centres at an irradiation rate of 300 rad/min full damage of transmission.

Fig. 4 Induced absorption recovery kinetics (500 nm) of different PWO crystals at room temperature after 500 Gy irradiation at a rate of 300 rad/min.

Fig. 5 Induced absorption in PWO crystals due to very fast recovering centres at a dose rate of 5 rad/min.

Fig. 6 Kinetics of the induced absorption recovery (500 nm) of the PWO crystal at 293 K and 273 K.

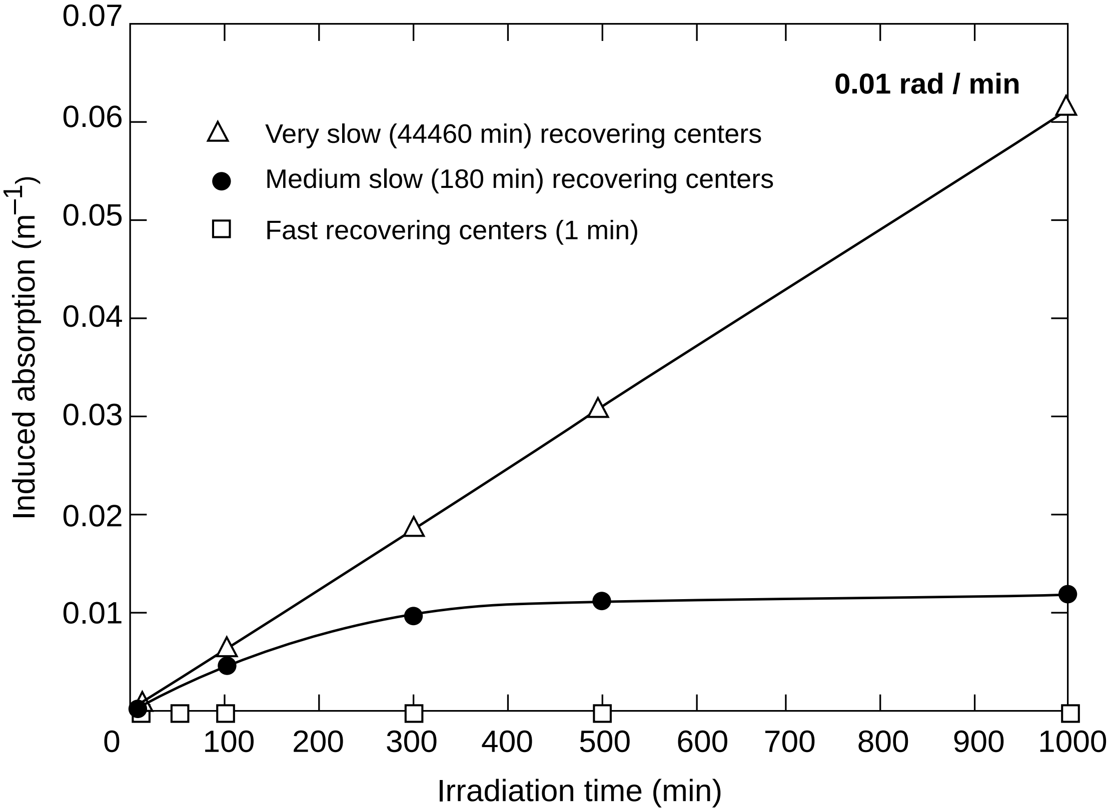
0.01 rad / min
Very slow (44460 min) recovering centers
Medium slow (180 min) recovering centers
Fast recovering centers (1 min)
Induced absorption (m^{-1})
Irradiation time (min)
0
0.01
0.02
0.03
0.04
0.05
0.06
0.07
100
200
300
400
500
600
700
800
900
1000

Fig. 1

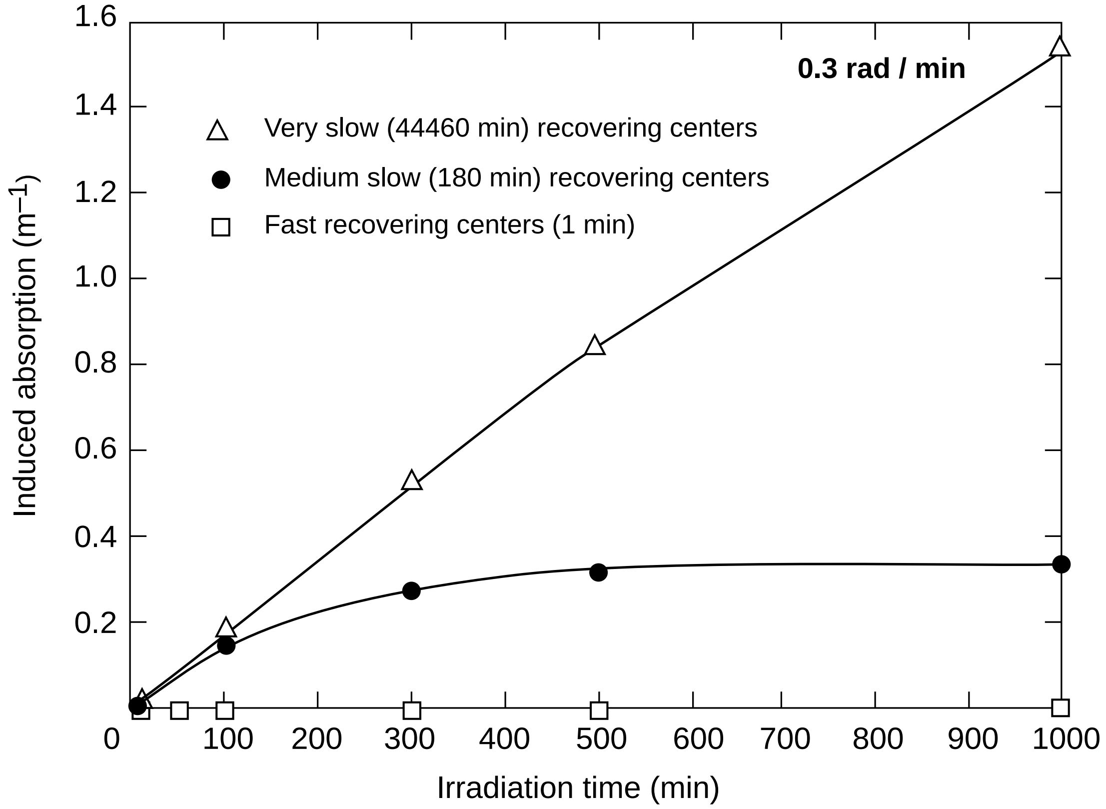
0.3 rad / min
Very slow (44460 min) recovering centers
Medium slow (180 min) recovering centers
Fast recovering centers (1 min)
Induced absorption (m^{-1})
Irradiation time (min)
0
0.2
0.4
0.6
0.8
1.0
1.2
1.4
1.6
100
200
300
400
500
600
700
800
900
1000

Fig. 2

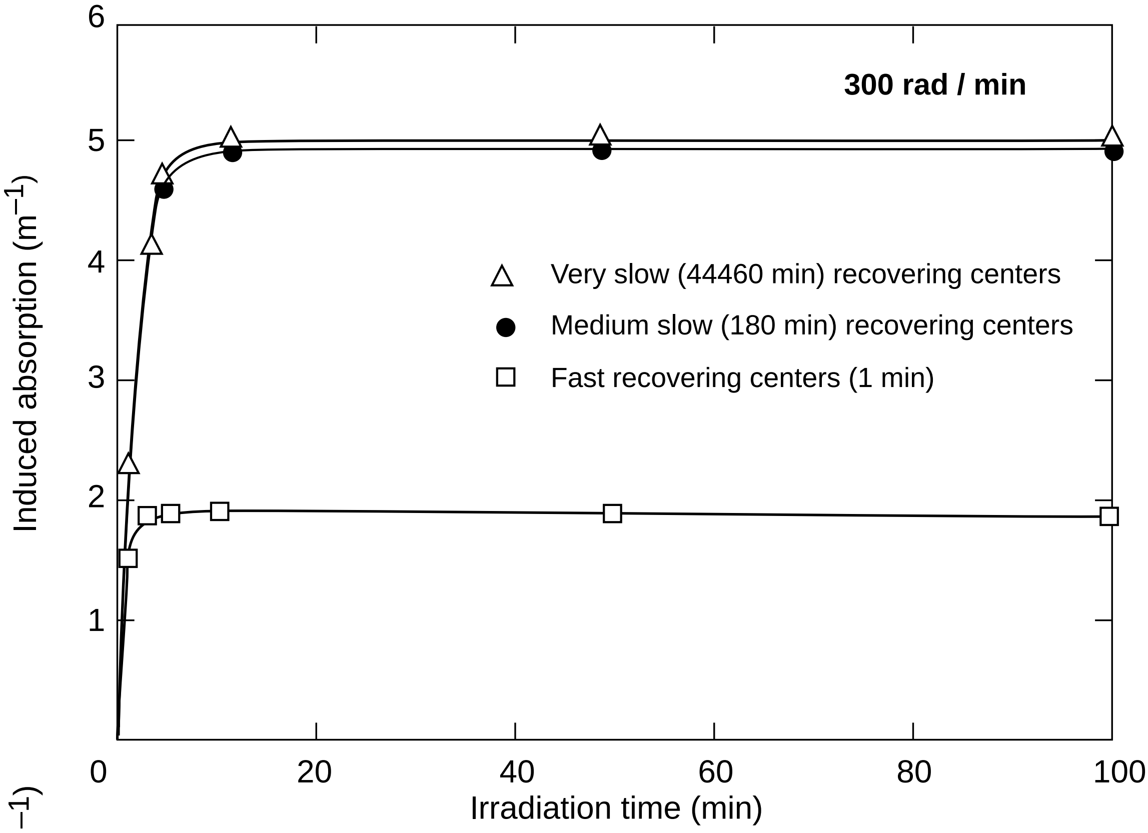
300 rad / min
Very slow (44460 min) recovering centers
Medium slow (180 min) recovering centers
Fast recovering centers (1 min)
Induced absorption (m–1)
Irradiation time (min)
0
1
2
3
4
5
6
20
40
60
80
100

Fig. 3

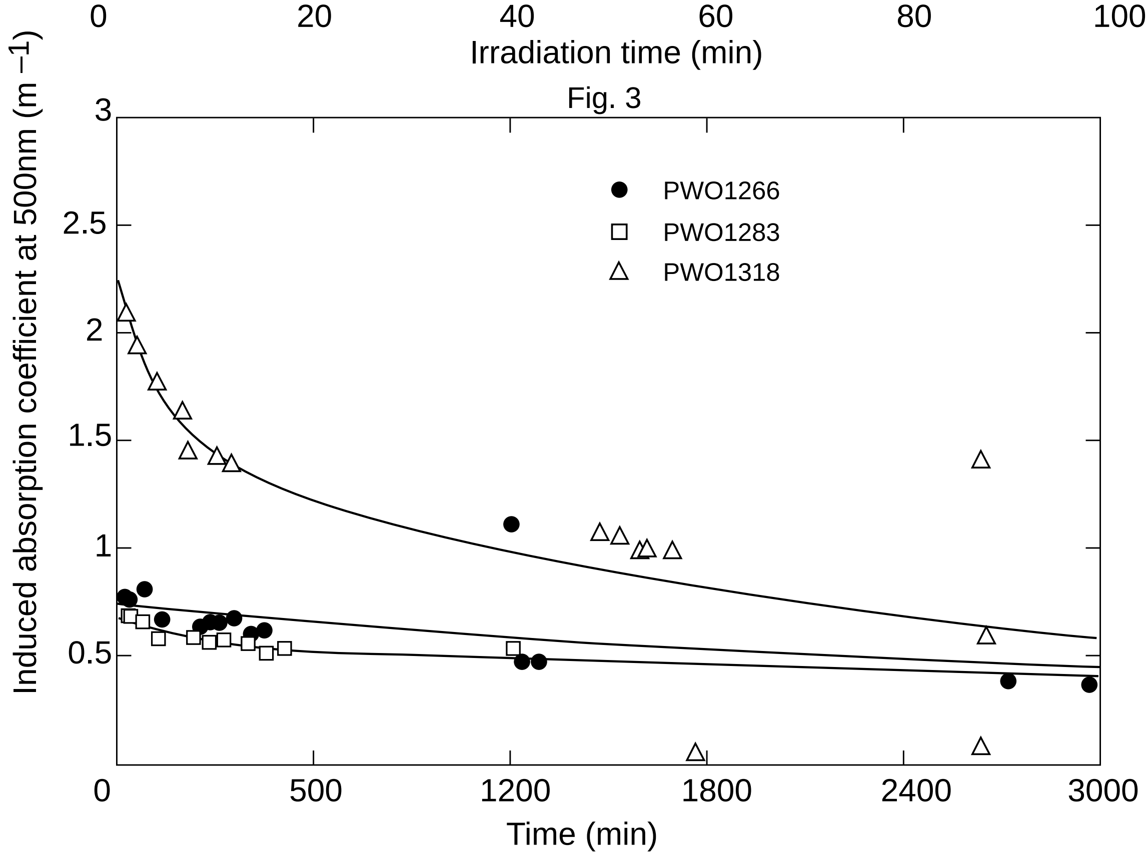
PWO1266
PWO1283
PWO1318
Induced absorption coefficient at 500nm (m –1)
Time (min)
0.5
1
1.5
2
2.5
3
0
500
1200
1800
2400
3000

Fig. 4

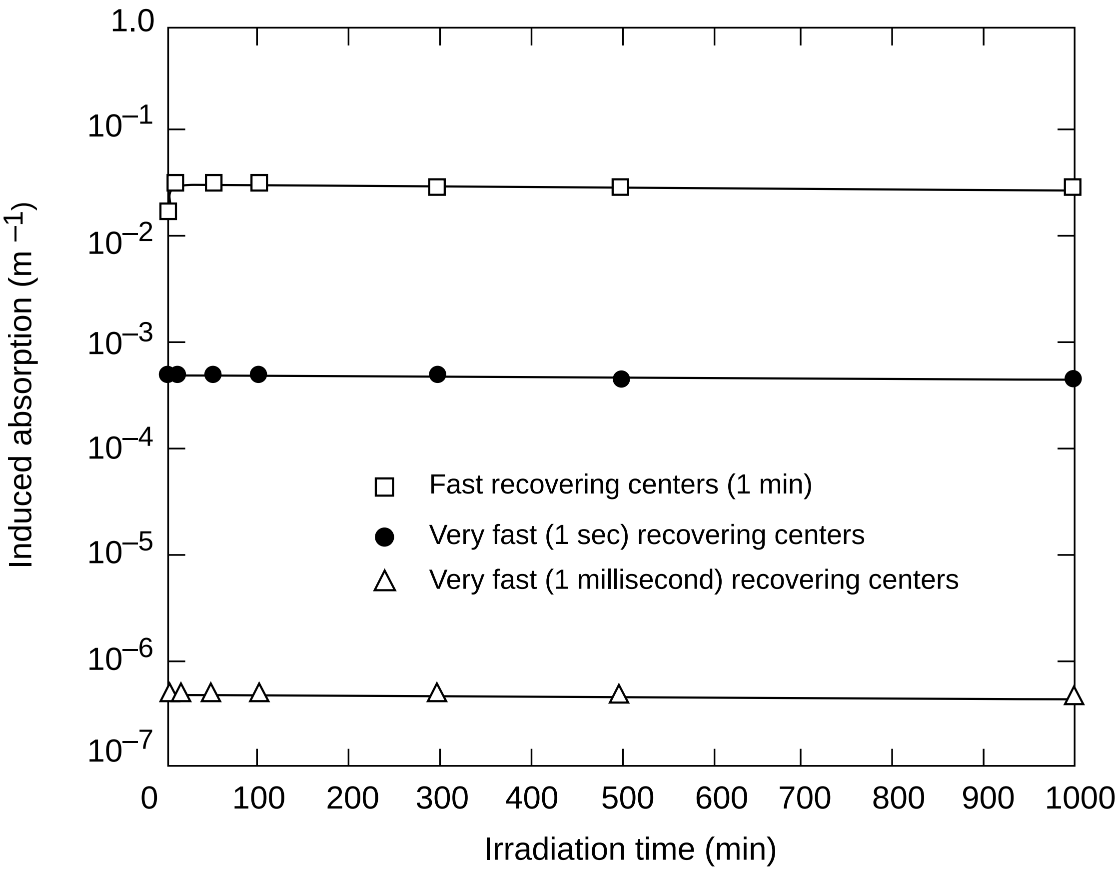
1.0
10–1
10–2
10–3
10–4
10–5
10–6
10–7
Induced absorption (m –1)
Fast recovering centers (1 min)
Very fast (1 sec) recovering centers
Very fast (1 millisecond) recovering centers
0
100
200
300
400
500
600
700
800
900
1000
Irradiation time (min)

Fig. 5

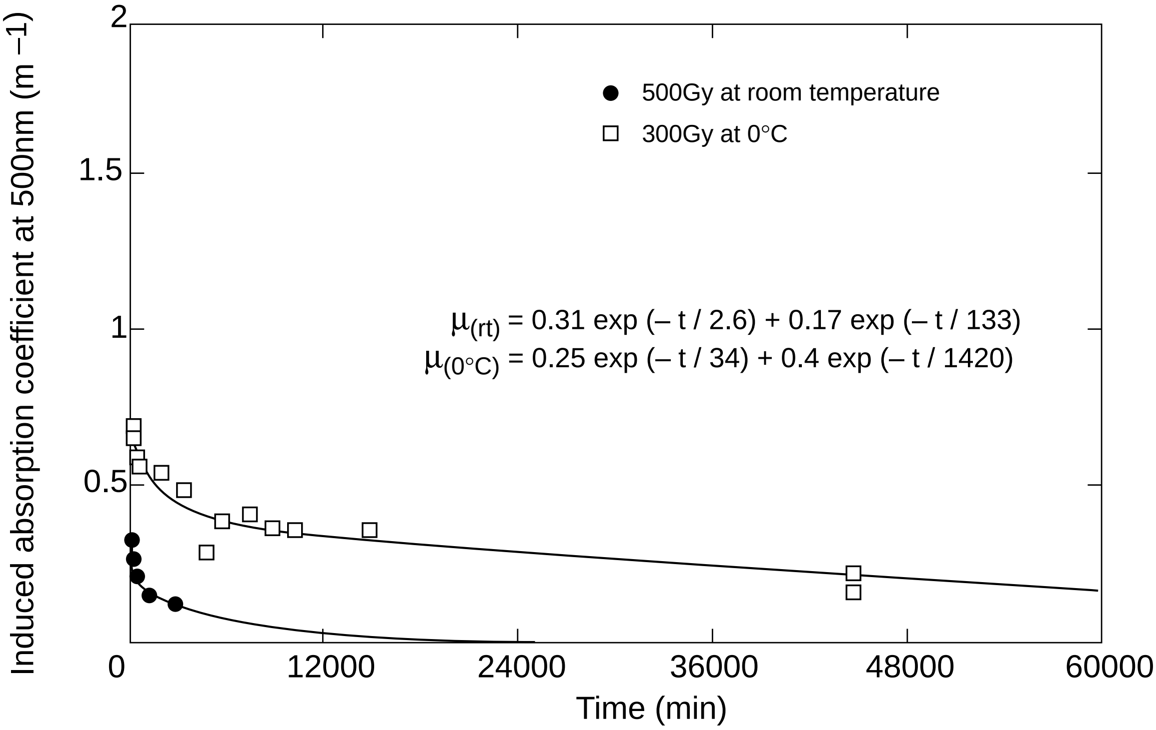
2
1.5
1
0.5
Induced absorption coefficient at 500nm (m –1)
500Gy at room temperature
300Gy at 0°C
μ(rt) = 0.31 exp (– t / 2.6) + 0.17 exp (– t / 133)
μ(0°C) = 0.25 exp (– t / 34) + 0.4 exp (– t / 1420)
0
12000
24000
36000
48000
60000
Time (min)

Fig. 6