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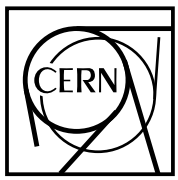

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Radiation damage mechanism in $PbWO_4$ crystal and radiation hardness quality control of PWO scintillators for CMS

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Abstract

The optical damage induced by UV light in $PbWO_4$ crystals is found to be similar to that induced by γ radiation. Due to the peculiarities of optical absorption in $PbWO_4$, the damage induced by UV light is a bulk process. This fact has important consequences for the approach to be adopted both for the use of the crystal as scintillator and for the qualification methods foreseen in the Regional Centres of the ECAL CMS Collaboration.

1. Introduction

The radiation resistance is one of the most important requirements for the $PbWO_4$ (PWO) scintillators to be used for the construction of the Electromagnetic Calorimeter (ECAL) of the Compact Muon Solenoid (CMS). The definition of a measurable parameter able to characterize the radiation hardness of PWO scintillators as well as a technological solution for the quality control of it are therefore compulsory.

Though the color centers induced by γ irradiation are intensively studied in the last period and their nature is quite well known [1, 2, 3], a reliable method to detect the crystal defects responsible for the bad radiation hardness of some PWO crystals is missing. The classification [4] of full length PWO scintillators in type 1, 2 and 3 defined by the shape of the optical transmission near the absorption edge allows only a crude characterization of their radiation resistance. A better understanding of color center production under γ radiation exposure is necessary if a reliable way to characterize the full length PWO scintillators from radiation hardness point of view is wanted. On the other hand it is obvious that one cannot imagine to test for radiation hardness all the 110 000 PWO crystals to be used for the construction of the ECAL exposing them to γ radiation.

Two are the possibilities to find out whether or not a PWO crystal is radiation hard:

1. test if the lattice defects responsible for the color center production are there;
2. try to create the color centers in a way previously certified as equivalent to γ radiation.

A careful analysis of the mechanism of color center production correlated with the peculiarities of the optical absorption in PWO led us to the conclusion that the damage induced by UV light in this crystal is similar to that induced by γ irradiation. UV light might then be used in a radiation hardness qualification line for PWO scintillators.

2. Experimental methods

A set of 8 PWO scintillators coming from mass production lots of three producers: Bogoroditsk Technochemical Plant, Russia (Bogoroditsk), "Preciosa" Crytur Company, Turnov, Czech Republic (Crytur) and Shanghai Institute of Ceramics, China (SIC) were chosen to be studied for their radiation hardness properties. The choice criterion was their different scintillation (decay time, emission spectrum, light yield) and radiation hardness characteristics. The list of the samples is given in Table 1. All faces of the samples were optically polished.

The details of γ irradiation facilities used in this work as well as the spectrophotometers used for optical transmission measurements were described elsewhere [5]. The samples were thermally treated for 2 hours at 200°C in the dark before each γ radiation or UV light exposure in order to bleach the effect of previous exposures. As UV light source, a Xe lamp (ILC Technology, ILC 201, 115V, 400VA emitting in the 200 - 1100 nm spectral range) was used.

The concentration of colour centres created by different procedures is estimated from optical transmission measurements since it is proportional to the induced absorption coefficient defined as:

$$\mu_{treat} = \left(\frac{1}{d}\right)\ln\left(\frac{T_{initial}}{T_{treatment}}\right) \qquad (1)$$

where $T_{initial}$: optical transmission before the treatment

$T_{treatment}$: optical transmission after the treatment

d : sample width

The generation of free carriers by UV light in the bulk of the material was checked by photoconductivity measurements. The photoconductivity spectra were made on a home made set-up made in such a way that the light exposure was possible both on the electrodes side (front) and the opposite side (back) of the sample.

Table 1

Crystal samples cut from mass production PWO scintillators

sample ID full length crystal ID	**Producer**	**Growth method**	**Doping X:ppm**	**transm. type [2]**	**Dimenssions mm^3**
1371 1266	Bogoroditsk	Czochralski	Nb:30	3	17x11x11
1377 1332	Bogoroditsk	Czochralski	Nb:30	1	24x11x10
1397 1037	Bogoroditsk	Czochralski	Nb:30	2	21x11x10
1404 1013	Bogoroditsk	Czochralski	Nb:30	3	18x11x9
1411 1310	Crytur	Czochralski		1	20x10x10
1461 1001	Bogoroditsk	Czochralski	Nb:30	1 (3)	21x10x10
1463 1126	SIC	Bridgman		3 (1)	23x12x11
1465 1262	SIC	Bridgman		1 (3)	23x12x11

3. Results and discussion

3.1 Production of colour centres in PWO by gamma irradiation

The eight samples studied in this work were exposed to γ radiation of different doses in the range 0.3 - 30 Gy at a dose rate of 7.4 Gy/h and previously noticed [6, 7] characteristics of the γ radiation damage process in PWO were confirmed:

1. the amplitude and weight of each absorption band differs from crystal to crystal
2. for a given crystal the relative amplitude of different absorption bands remain constant with the absorbed dose, i.e. the shape of the radiation induced absorption spectrum remains unchanged with the absorbed dose

Fig.1 gives the γ induced absorption spectra for the eight samples for a radiation dose of 3 Gy. The spectra are in fact the superposition of absorption bands [1] attributed respectively to:

- an oxygen vacancy which captured an electron (F^+ centre : 670 - 705 nm) or two electrons (F centre : 525 - 575 nm)
- a hole captured by the WO_4^{2-} group ($(WO_4)^-$ centre : 410 - 430 nm)
- a hole captured by a Pb^{2+} ion (Pb^{3+} centre : 340 - 360 nm)

These colour centres are produced by γ radiation exposure in a three steps process:

1. creation of hot electron-hole pairs
2. cooling down and diffusion of separated electrons and holes
3. thermalised electrons and holes are fixed in suitable lattice sites, thus creating the above mentioned colour centres

At this stage of understanding of colour centres production process it is well possible that the same kind of colour centres with the same relative concentration as in the case of γ exposure can be produced as soon as free electron-hole pairs are created in the bulk of the PWO crystal , no matters the way in which the free carriers were created. The smoothed shape of the fundamental absorption edge in PWO with values of the absorption coefficient of the order of 10 - 100 m^{-1} makes it possible the penetration of light deep in the material for distances of the order of centimeters. In this case pairs electron-hole are created in the bulk of the crystal and if the necessary defects in the lattice are present, colour centres are created. The thermal decomposition of bound exciton state into free electron and hole pairs [8] at 300 K may also play a role in a "deep penetration" of free carriers into bulk and color centres formation. All this will be possible if the diffusion length of photocarriers is sufficiently high.

3.2. Colour centres production in PWO by UV light exposure

It was already noticed that UV light can damage the PWO crystals but the effect was looked as a surface process only, given the huge values of the absorption coefficient of PWO in this wavelength range. In order to check if the colour centres produced in different PWO samples by UV light exposure are the same as those produced by γ irradiation, the eight samples studied in this work were exposed to a Xe lamp. No filter was used and no estimation of the light flux value at the level of the crystal sample holder was made. Nevertheless it is possible to say that the light flux was the same for all samples since they were strictly kept in the same position for the same time interval. Transmission measurements were made on a direction perpendicular to the face exposed to the Xe lamp before and after the exposure and the induced absorption coefficient was calculated. The resulting spectra are given in fig. 2. The similitude with the γ radiation induced absorption spectra is striking for all samples. In both cases not only the colour centres but also their relative concentration in the bulk of the crystals are the same. Fig. 3 gives the correlation between the γ radiation induced absorption coefficient (for a 3 Gy radiation dose) and the corresponding (same wavelength) induced absorption coefficient after the Xe lamp exposure of the PWO samples studied in this work.

Even if the correlation between γ and UV induced colour centres concentration is observed, it should be stressed that the two processes though similar in their final stage, are different in many aspects. For the colour centres that generate the absorption bands in the 300 - 700 nm region it is important to note that:

-in the case of γ irradiation, for a given dose acummulated at a given dose rate the concentration of the colour centres produced in the crystal depends only on the concentration of the defects (crystal sites able to form the Pb^{3+}, $(WO_4)^-$, F and F^+ colour centres)

- in the case of UV light exposure, for a given incident energy at a given flux value, the concentration of the colour centres produced in the crystal depends not only on the concentration of the above mentioned defects but also on the values of the optical constants (absorption and reflection coefficients) of the sample.

Due to these peculiarities it might happen that a PWO crystal seem UV hard only because the absorption coefficient in the UV wavelength region is very high and the damage occurs on the surface of the material. It also means that the proportionality factor between the UV induced and γ induced absorption coefficients will be the same for two different crystals only if the optical constants of these two crystals are the same in the wavelength region used for UV light exposure. Once the PWO scintillator production technology will be well defined, precise values of the optical constants might be imposed to the producer so that a reliable qualification method could be defined (see further).

3.3. Photoconductivity measurements

In order to get more information on the way that colour centre are produced by UV light exposure, photoconductivity spectra were made on the sample PWO1465 (23x12x11 mm^3) on which 5 x 5 mm^2 Al electrodes were vacuum deposited at 2 mm distance on one of the major faces of the sample. The photoconductivity spectra obtained for front and back exposure of the sample are given in fig. 4 where the optical density:

$$\varepsilon = \frac{1}{d} \cdot \ln\left(\frac{100}{T(\%)}\right) \ (m^{-1}) \qquad (2)$$

of the sample is also given in the inset. The relatively sharp photocurrent peak in the case of back exposure correlated with the shape of the optical density near the absorption edge of the sample indicates that the diffusion of photocarriers is not the leading factor in the process. Photocarriers are created deep (in the proximity of electrodes) in the PWO crystals by UV light in a 50 nm range peaking at 350 nm. It means, in the frame of the model for the colour centre production exposed above, that light with wavelengths in this range will induce in the bulk of the material the same colour centres as γ radiation.

5. Revealing the lattice sites responsible for γ radiation damage in PWO scintillators by UV light exposure

Given the huge number of crystals foreseen to be used for the construction of the ECAL-CMS detector, it is a hard challenge to find the most efficient qualification procedure for the PWO scintillators. The method to be adopted for radiation hardness quality control in the regional centres where parts of the detector will be assembled and tested, has to be:

1. non destructive
2. no time consuming
3. done in the whole volume of the crystal
4. compatible with other tests foreseen for the quality control procedure
5. cheap

On the other hand the experience of first industrial scale produced PWO crystals may imply that the control of the radiation hardness characteristics has to be done in a more systematic way than initially foreseen. Given the fact that UV light can produce the same optical damage in $PbWO_4$ crystals as γ radiation, the radiation hardness test could be made in a three steps sequence at the beginning of the quality control procedures foreseen for the PWO scintillators:

1. longitudinal transmission measurement $T_0 = F(\lambda)$ in the wavelength range 300 - 800 nm for a number of wavelengths to be defined. Comparison of $F(\lambda)$ with a $F_0(\lambda)$ caliber in order to discard crystals having cracks or other major optical defects

2. exposure of the crystal to UV light (most probably 300 - 400 nm) for an equivalent γ dose to be defined

3. longitudinal transmission measurement $T_{UV}(\lambda) = G(\lambda)$ in strictly the same conditions as 1.

4. Define a quality factor:

$$Q(\lambda) = \frac{F(\lambda) - G(\lambda)}{F(\lambda)} \cdot 100 \quad (\%) \qquad (3)$$

and compare with a predefined $Q_0(\lambda)$ caliber in order to discard crystals having $Q < Q_0$ in the wavelength range of interest (400 - 550 nm).

Preliminary tests made on full length PWO scintillators show that the proposed method is promising. The longitudinally measured induced absorption coefficient in crystals laterally exposed to a Xe lamp (fig. 5) are identical in shape with those induced by γ exposure. In the inset of the plot for each crystal the correlation between the induced absorption coefficients for γ and UV exposures are given. The values of the γ radiation induced absorption coefficient correspond to a 500 Gy radiation dose at 180 Gy/h dose rate. Absorption spectra are measured 30 - 45 min after irradiation. The UV induced absorption coefficient correspond to approx. 10 minutes of Xe lamp lateral exposures of the crystal. Absorption spectra are measured immediately after the Xe lamp exposure. Before both gamma and Xe lamp exposure the crystals were thermally bleached.

Conclusion

Lattice sites favorable to colour centre formation are present in PWO crystals as grown. A method to reveal the presence of these sites would be sufficient to qualify the PWO scintillators for γ radiation hardness. The UV exposure test is a possible solution since the present work proves that the colour centres induced in PWO crystals by UV light exposure: (i) might be produced in the bulk of the crystal; (ii) are of the same nature and (iii) are produced in the same relative concentration as for γ irradiation. Further studies in order to completely understand the UV induced damage process in PWO are necessary in order to set up a reliable qualification procedure to be applied in the Regional Centres of the CMS-ECAL Collaboration.

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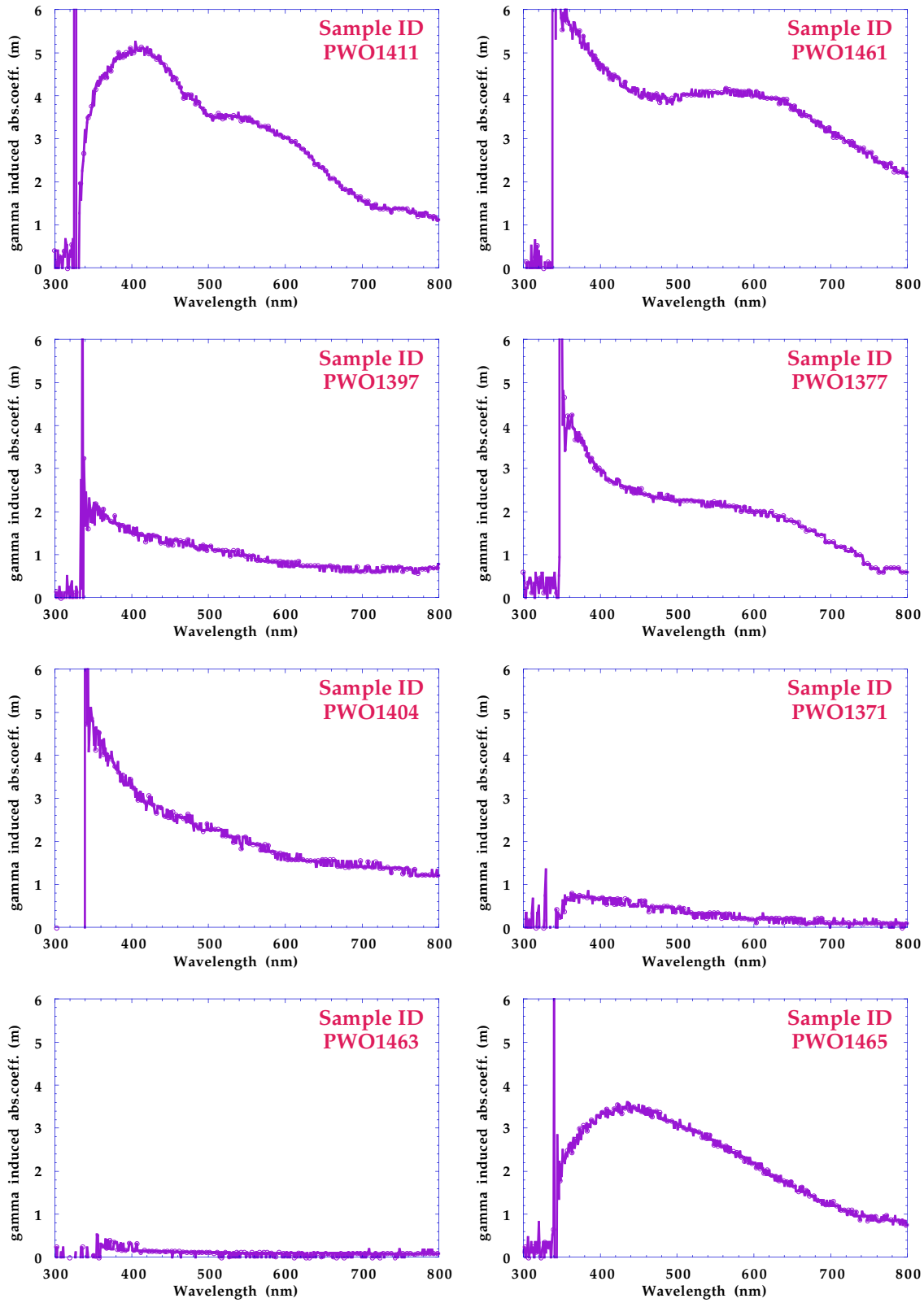

Fig.1 Colour centres production in different PWO crystals by γ irradiation (absorbed dose 3 Gy)

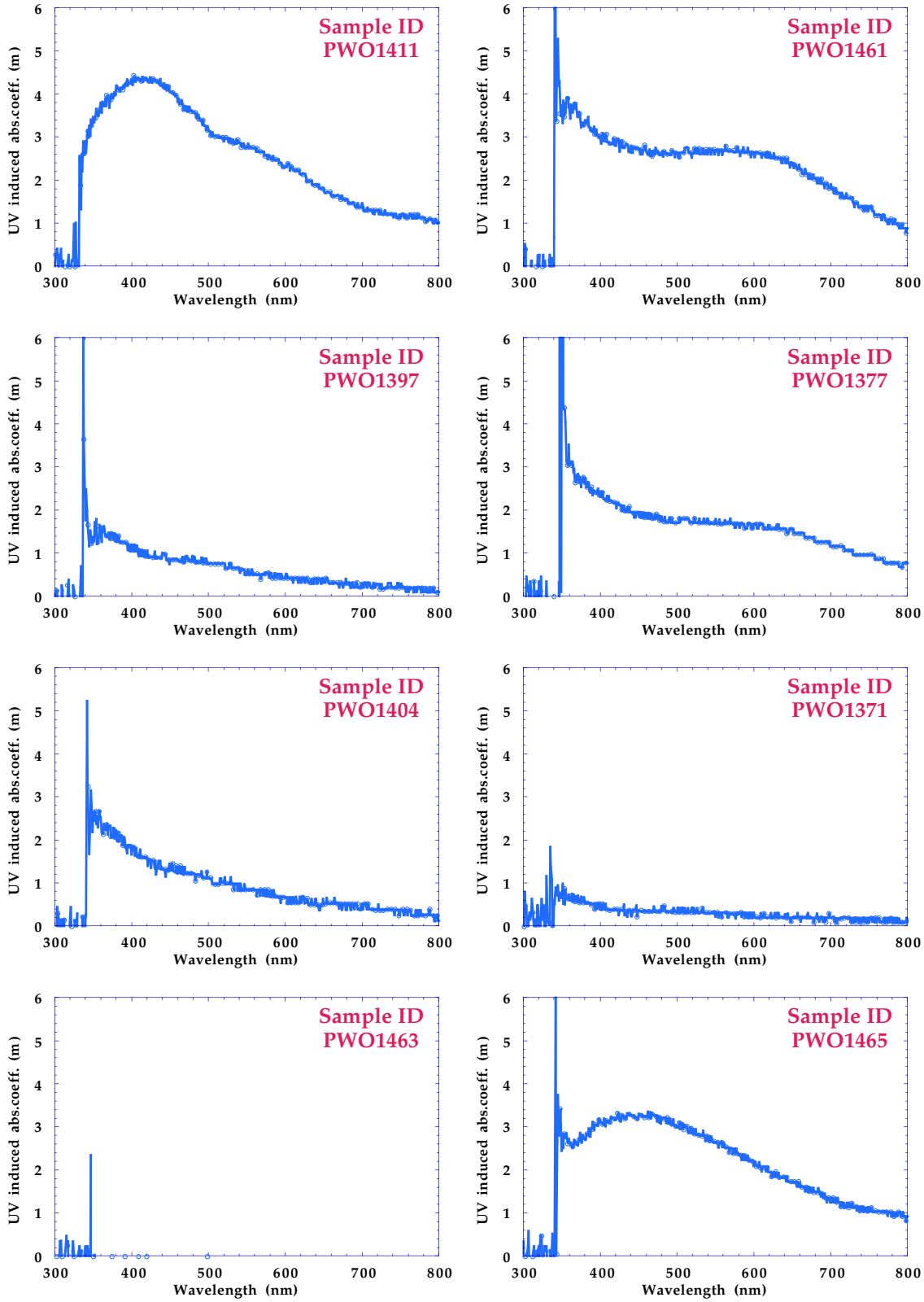

Fig.2 Absorption coefficient induced by a Xe lamp exposure in different PWO crystals

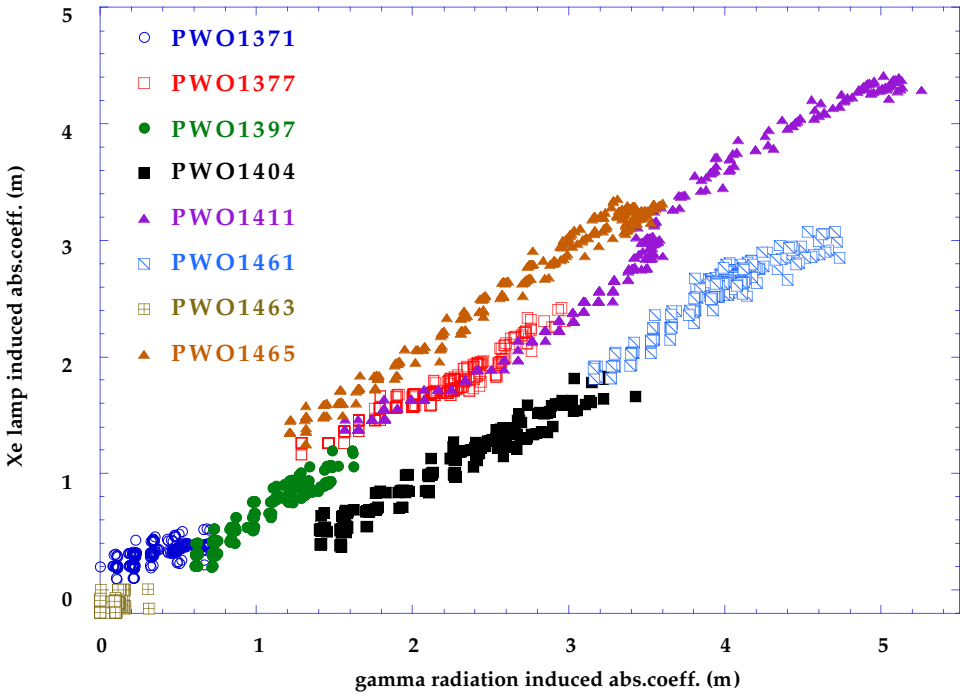

Fig. 3 Correlation between γ radiation and UV light induced absorption coefficients in the (400 - 700) nm spectral region for different PWO crystals

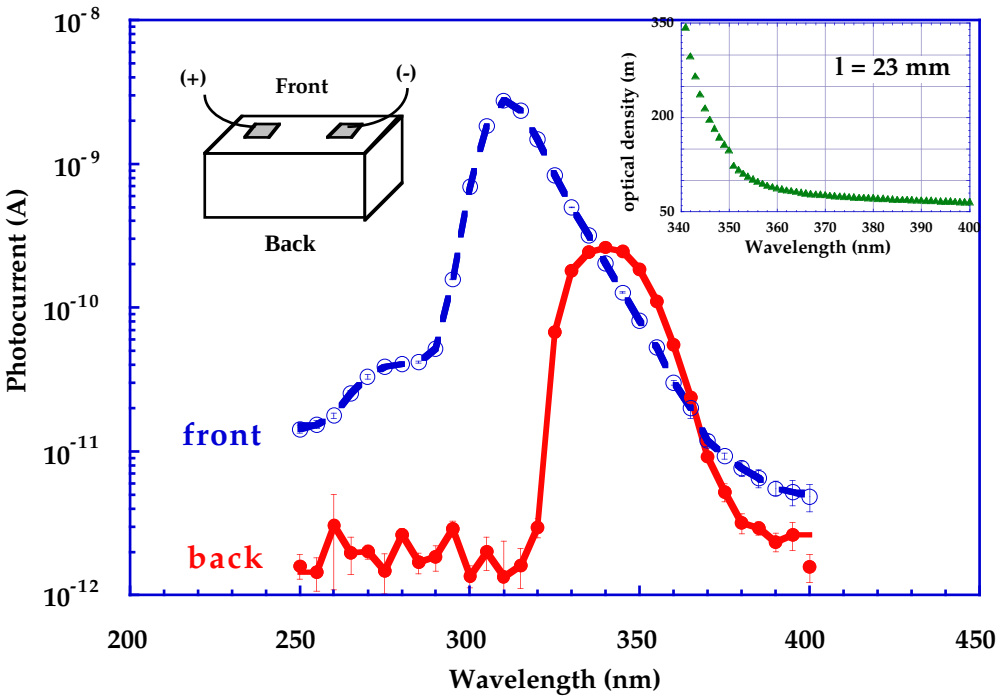

Fig.4 Photoconductivity spectrum for a PWO sample (see the text)

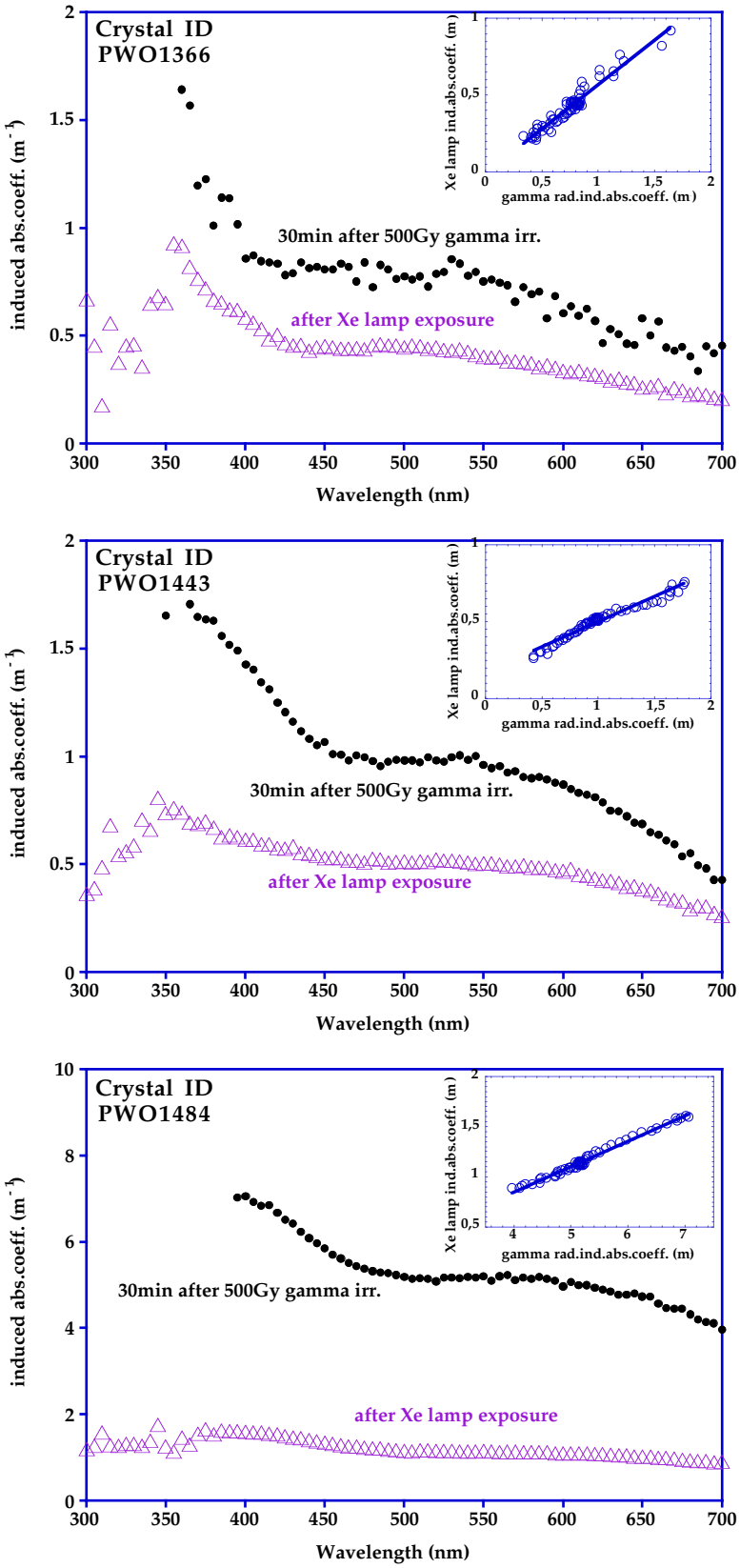

Fig. 5 Comparison between a Xe lamp lateral exposure and γ radiation effects upon full length PWO scintillators. The correlation between γ radiation and UV light induced absorption coefficients are given in the insets