Behaviour of PWO scintillators after high fluence neutron irradiation*

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Abstract

We report here the results of lead tungstate single crystal irradiation with a fast neutron fluence of about 10^{19} cm^{-2}. Definitely, such fluence is extreme and will be never achieved at high energy physics experiments. However we were motivated, firstly, to study with this heavy irradiation a new approach that we call "nuclear doping" to obtain impurity-activated centers in the crystal. The transmutation of tungsten to rhenium by neutron irradiation was supposed to be a way to introduce rhenium in the tungstate lattice. Re ions are well known luminescence centers and may change scintillation properties of the $PbWO_4$ crystal. Secondly we aimed to study the degradation of the material and possible loss of the scintillation properties due to an eventual amorphisation of the material by fast neutrons, and to investigate recovery of the damage under heat treatment.
We present in this paper the study of the optical and scintillation properties of the irradiated sample compared to a control sample. Initially very degraded, the irradiated sample recovers partially after annealing at 600 °C. Even after a 900 °C annealing, the recovery is not complete. For the irradiated sample, the X-ray luminescence spectrum intensity is approximately 35 % of these of the control sample. The optical transmission is not totally recovered as well, inducing a shift toward the red in the photoluminescence emission spectrum. No trace of Re or other center emission is detected. If $PbWO_4$ survives quite well to such an extreme fluence of neutrons, this method does not seem to be practical to induce Re-activated luminescence centers and to enhance its luminescence yield.

1. Introduction

Scintillation single crystals of lead tungstate, $PbWO_4$, now are widely applied for high energy physics experiments. Stability of its parameters in radiation environment is one of the most important features for their application in precise electromagnetic calorimetric detectors [1].

In another hand, improvement of its light yield is actively researched in view of application in other fields, in which lower energies are to be detected. Various additives have been tested, most of them coming in substitution to the tungsten or lead ions [2].

In that frame, it has been proposed to use rhenium, but by no way this element could be introduced in sizable quantity in the $PbWO_4$ lattice during crystal growth. A only potential way to introduce rhenium in the lead tungstate lattice was to use the transmutation of tungsten in rhenium by thermal neutrons capture.

Therefore we have undertaken a explorative research program, submitting a $PbWO_4$ sample to a high flux of neutrons, and measuring it's optical and luminescence properties. The results of this work are reported here.

2. Neutron irradiation

The crystal sample, of mass 0.84 g and approximate size $10 \times 10 \times 1$ mm^3, was irradiated in June 2000 in the channel β1/H2 of the reactor Osiris in Saclay, during 110 h. A similar sample was kept apart as reference. The fluences reached were about $7.8\ 10^{18}$ cm^2 for fast neutrons, $1.2\ 10^{19}$ cm^2 for epithermal neutrons and $4.0\ 10^{19}$ cm^2 for thermal neutrons [3]. The dose received is estimated to $3.3\ 10^8$ Gy (33 Grad) [4]. In that condition, the main nuclear reactions occurred with the following natural isotopes of tungsten: ^{186}W (28.4 % natural abundance), ^{184}W (30.7 %) and ^{180}W (0.12%):

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$$^{186}\mathrm{W} + \mathrm{n_{th}} \xrightarrow{\sigma=36\mathrm{b}} {}^{187}\mathrm{W} \xrightarrow{\tau=24\,\mathrm{h}} {}^{187}\mathrm{Re} \qquad (1)$$

$$^{184}\mathrm{W} + \mathrm{n_{th}} \xrightarrow{\sigma=2\mathrm{b}} {}^{185}\mathrm{W} \xrightarrow{\tau=75\,\mathrm{d}} {}^{185}\mathrm{Re} \qquad (2)$$

$$^{180}\mathrm{W} + \mathrm{n_{th}} \xrightarrow{\sigma=4\mathrm{b}} {}^{181}\mathrm{W} \xrightarrow{\tau=121\,\mathrm{d}} {}^{181}\mathrm{Ta} \qquad (3)$$

The other natural isotopes (^{182}W (26.50 %) and ^{183}W (14.31%)) gives stable nucleus and, as well as lead and oxygen, do not participate significantly to the induced radioactivity.

Assuming the published thermal neutron cross-section parameters and neglecting reaction with epi-thermal and fast neutrons, the quantity of rhenium formed (dominantly ^{187}Re, which decays in ^{187}Os with a 4.2 10^{10} years half life.) should be around to be 0.041 % of tungsten content.

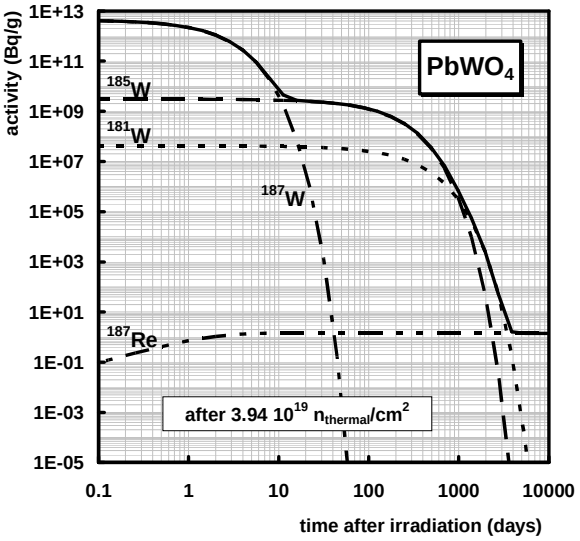

Fig. 1. Calculated induced activity of the $PbWO_4$ sample irradiated.

After irradiation the sample was highly radio-active. Figure 1 shows the estimated activity in function of time. The decay of ^{187}W dominates in the first days, then ^{185}W during three years, and then ^{181}W emerges. The sample was then kept in its irradiation container in a repository place for three years, and became assessable safely for study at the middle of 2003. It appears at that time dark red and its surface was slightly damaged, probably because of the heat suffered in, a not controlled atmosphere, during neutron irradiation. Unfortunately it has been impossible yet to polish it because of its internal radioactivity.

3. Annealing and Measurements

After irradiation the irradiated sample was annealed in air in successive steps, from 100 °C to 900 °C by 100 °C, (ramp: ± 1 °C/mn, plateau at T_{anneal}: 2 h). optical transmission and UV luminescence excitation and emission spectra of the irradiated and a control sample of the same size were measured before, between and after annealing steps using respectively a *Lambda 19* and a *LS50B* Perkin-Elmer spectrometers, X-ray luminescence spectra were measured at LPCML before annealing, and after the 600 °C and 900 °C steps. The optical absorption coefficient at step *i*, μ_i is calculated from the initial measured optical transmission T_0 and the optical transmission T_i at step *i*, using the usual formula (*e* being the thickness of the sample, here 1 mm):

$$\mu_i = \ln\left(T_0/T_i\right)/e \qquad (4)$$

4. Results

Before annealing, optical absorption was very sever (see Fig. 1) and luminescence (either UV and X-ray below the sensitivities of our instruments. Optical transmission began to recover beyond 200 °C, and, as seen on Fig. 1., three or four absorption bands emerged, centered at 473 – 500, 642 and 745 nm. No further improvement occurred between 700 and 900°C, but the already-noted effect of surface degradation might be important (especially in the low wavelength region) and could not be distinguished from bulk attenuation.

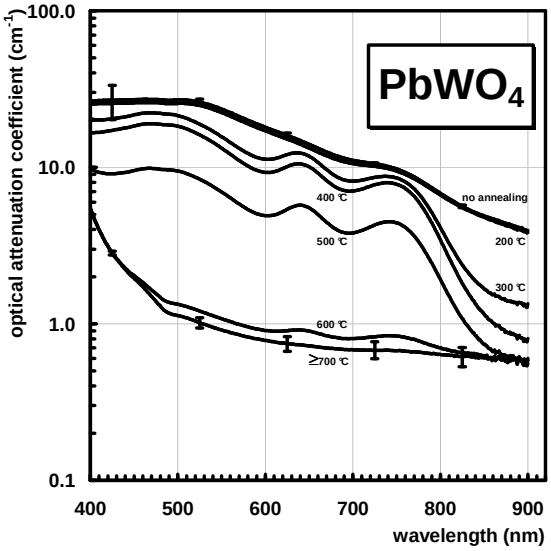

Fig. 2. Optical attenuation coefficient of the irradiated sample before annealing and after successive annealing temperatures.

UV-Luminescence was very low at any steps below 600 °C. It reappears after 600 °C annealing and remains stable beyond. As shown on Fig. 2, the overall intensity of emission is almost one fifth of the one of the initial $PbWO_4$. Compared to the initial spectra, the excitation spectra remain similar, when the emission spectra are shifted toward the red.

The X-ray luminescence spectra were measured only before annealing and after the 600 and 900 °C steps. As shown on Fig. 3, they have almost the same shape as the initial spectrum, and a sensible improvement is seen between the two annealing steps. After 600 °C, the intensity is around one fourth of intial one. It reaches one third after 900 °C.

The difference in the photo and X-ray luminescence spectra is rather unusual, and can not be explained by optical absorption in the material. In that case this is the X-ray spectrum which should be shifter in the red part. This difference can be due to the deterioration of the surface, already seen in the optical transmission measurements. Unfortunately, we still have to wait some months before to polish the sample safely.

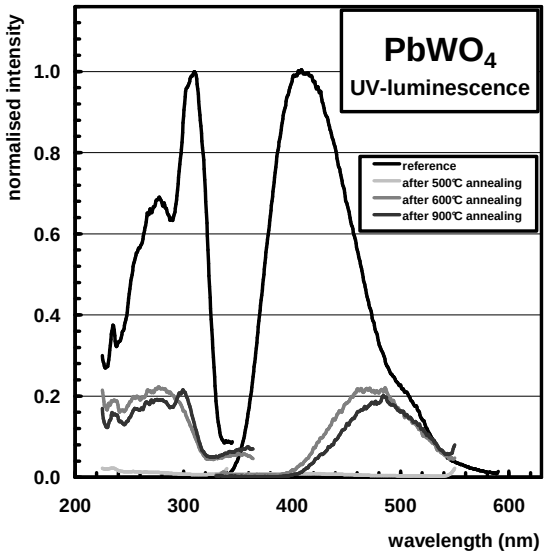

Fig. 3. UV luminescence spectra of the irradiated sample before irradiation (black) and after 500 °C (light grey) 600 °C (medium grey) and 900 °C (dark grey) annealing temperatures.

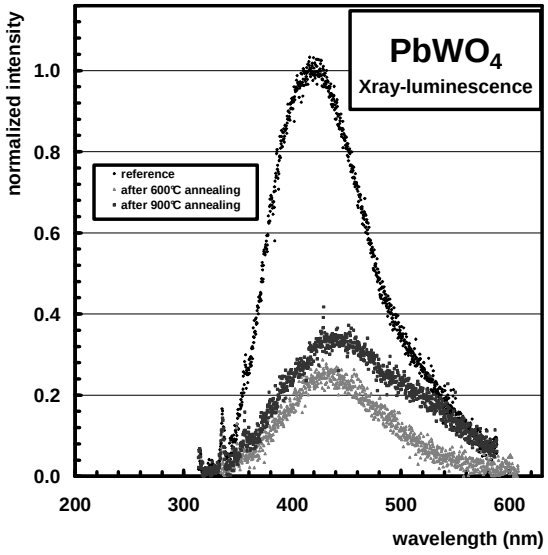

Fig. 4. X-ray luminescence of the irradiated sample before irradiation (black) and after 600 °C (medium grey) and 900 °C (dark grey) annealing temperatures, normalized to the .

However, the recovery, at least partial, of both transmission and X-ray luminescence spectra indicates that the structure of lead tungstate has not been much deteriorated by this heavy irradiation. No amorphisation of the material, which would not have been suppressed at the temperature of annealing reached, occurred. On an other hand, no indication of rhenium luminescence is seen and neutron induced rhenium doping seems to be neither practical neither efficient to increase the lead tungstate light yield.

5. Conclusion

After a drastic neutron (and gamma) irradiation, lead tungstate is not destroyed nor “amorphised”. The optical transmission and luminescence spectra are partly recovered, in two steps, around 200 and 600°C. The surface state, altered during irradiation, is responsible for a part of the permanent damage, and further studies will be done when the radioactivity decrease would allow the polishing of the sample. No trace of rhenium or other center emission is detected. If $PbWO_4$ survives quite well to such an extreme fluence of neutrons, the method of neutron transmutation does not seem to be practical to induce rhenium-activated luminescence centers and to enhance its luminescence yield.

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