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Cathodoluminescence-Based Quantitative Analysis of Radiation Damage in Powellite Single Crystals

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Powellite ($CaMoO_4$) is a phase which appears to crystallize in high waste loading glass (HWL glass) developed for the immobilization of nuclear wastes. Taking into account that radioisotopes tend to segregate towards powellite precipitations, it is of prime importance to assess the stability of this compound upon irradiation. Damage accumulation was reproduced by using an ion irradiation method, which is the most convenient method of simulation of radiation defects created during the exposure of material to a radiative environment. Although the radiation damage build-up in single crystals is extensively studied since early fifties of the 20^{th} century, mainly by the use of Rutherford Backscattering/channeling (RBS/c) method, the very recent concept is to use luminescence techniques to study the radiation damage, which constitutes a novel approach for this topic.

$CaMoO_4$ single crystalline samples were irradiated with 320-keV Ar ions at fluences ranging from $1x10^{13}$ to $1x10^{16}$ cm^{-2}. Cathodoluminescence (CL) measurements were performed using an EMSystems setup mounted in Auriga CrossBeam Workstation (Carl Zeiss NTS). The electron beam energy was reduced to 10 kV in order to limit the analyzed volume of the material as much as possible to the damaged layer (~200 nm). A broad luminescent band consisting of five peaks positioned at ~462, 498, 534, 593 and 681 nm was observed in the CL response to electron irradiation (Fig.1a). This multi-component broad green emission band is known to be due to electronic transitions of charge transfer type within the $(MoO_4)^{2-}$-anion complex and is interpreted as an emission of self-trapped molecular excitons. Electric dipole allowed transitions $^1A_1 \rightarrow (^1T_2, {}^1T_1)$ is responsible for the fundamental absorption edge of $CaMoO_4$ while the radiative transitions originate from the lower-lying triplets $^3T_2$ and $^3T_1$ down to the $^1A_1$ level. The short wavelength luminescence (498 and 534 nm) of $CaMoO_4$ is generally attributed to the intrinsic emission of the $(MoO_4)^{2-}$-centers whereas that at long wavelength (593 nm) can be ascribed to defect centers.

The ionoluminescence (IL) was measured using a homemade system based on the use of Hamamatsu spectrometer collecting the light from sample installed inside a target chamber of an ion implanter (Balzers MPB 202RP). A H_2^+ ion beam of 50 keV energy (selected so to correspond to the projected range of 320 keV Ar ions) was used to excite luminescence. Again a broad band of green emission was observed, showing three peaks (at 482, 543 and 626 nm) after deconvolution.

The areas of each peak (the integrated intensities, I_p) in the registered CL and IL spectra and their variation with the irradiation fluence were analyzed for each sample in the study. The CL and IL spectra observed before and after irradiation were fitted with Gaussians after background subtraction. An

example of fitting procedure used to analyze the spectra is presented in Fig.1a for the case of unirradiated $CaMoO_4$ sample. The integrated intensities of each peak were then plotted with the ion fluences, so that the kinetics of the damage accumulation were determined (Fig. 1b). For single crystals of powellite the damage-build up as a function of accumulated ion fluence was established through RBS/c. The results of CL, IL and RBS/c analysis were then processed using Multi-Step Damage Accumulation (MSDA) model [1-3]. That allowed for the determination of damage build-up kinetics, and finally cross-section for radiation damage build-up. The direct correlation between the defects formation and the rapid decrease of the luminescence of the material was observed, which in the case studied mostly occurs in the first stage of the defects accumulation and is probably caused by creation of small clusters of defects or isolated interstitial atoms.

The data acquired by the use of CL and RBS/c method confirmed the two-step character of damage accumulation process in powellite crystals, both methods pointed to similar values (~1.3 x 10^{15} cm^{-2}) of threshold irradiation fluence required to induce structural transformation. These results point out the possible use of luminescence techniques for quantitative analysis of damage accumulation in polycrystalline materials. Such a method may offer thus interesting practical applications, as polycrystalline materials are the only form of solids used as construction materials in nuclear engineering [4].

References:

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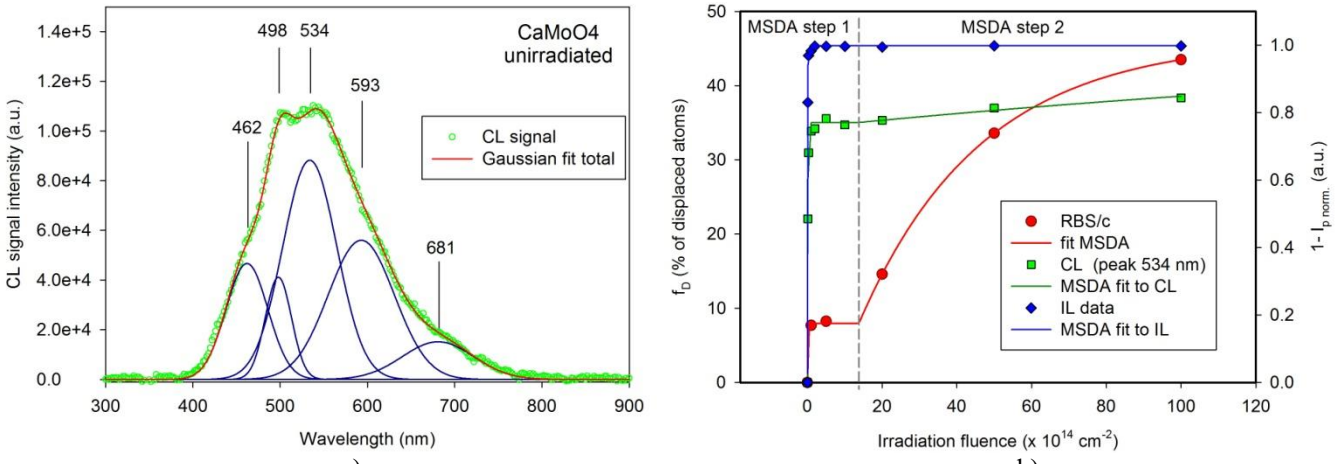

a) b)

Figure 1. Analysis of CL spectra recorded on $CaMoO_4$ unirradiated sample (a), and radiation damage accumulation kinetics based on CL, IL and RBS/c measurements analyzed with MSDA model (b).

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