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X-ray induced radiation damage in CLYC(Ce)

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ABSTRACT

The radiation hardness of the ^{6}Li loaded scintillator CLYC(Ce) to X-rays was investigated. Two crystals were studied; one crystal was irradiated with X-rays and one was kept as a control and only exposed to a moderated ^{241}Am–Be source. The control crystal was used as a reference sample for photoluminescence excitation and emission measurements. The exposed crystal was given two doses of X-rays, the first was 2.3 Gy and the second was 118.2 Gy after a 22 week annealing period. The total dose was 120.5 Gy and was found to significantly reduce the light yield of the crystal. Pulse height spectra from the moderated ^{241}Am–Be taken with the irradiated crystal showed a 54% decrease in pulse height and a broadening of the FHWM of the peak from 14.6 % to 44.2%. Analysis of the pulse shapes showed no change to the median decay time to 37% of the scintillator, with values of ~350 ns for the irradiated and control crystal. Photoluminescence excitation and emission measurements revealed a reduction in the relative intensity of the main Ce^{3+} emission with respect to the self-trapped exciton luminescence. This indicated a modification to the Ce^{3+} luminescence center. There was little recovery at room temperature over the course of 22 weeks after the first irradiation. The irradiated crystal was also found to display significant room temperature luminescence after X-ray irradiation with it being measurable up to 120 min after the X-ray irradiation.

1. Introduction

New and upgraded neutron scattering facilities such as the European Spallation Source in Sweden [1], J-PARC in Japan [2], the Spallation Neutron Source in the United States [3] and the ISIS pulsed neutron and muon source in the United Kingdom [4] produce high neutron fluxes for materials science. ISIS is a neutron spallation source which is used to generated thermal and epithermal neutrons and muons. There are currently 30 neutron scattering instruments in ISIS which are dedicated to applications including neutron reflectometry, powder and single crystal diffraction, small angle scattering and imaging. Neutron sensitive scintillators such as ZnS:Ag/^{6}LiF and ^{6}Li-glass are currently used on ISIS [5]. These scintillators have disadvantages like long afterglow for ZnS:Ag/^{6}LiF and high sensitivity for the ^{6}Li-glass [4]. Because of these disadvantages, new neutron sensitive scintillators are regularly evaluated to determine their suitability for use at ISIS.

Due to inaccessibility and cost restrictions in neutron scattering instrumentation, instruments are expected to have a lifetime of ~20 years. Radiation hard detectors are therefore required. High neutron fluxes also give rise to high backgrounds of γ-rays. Therefore, damage from both of these sources of radiation must be evaluated. The group of van Eijk at the Delft University of Technology have performed a thorough, though non-exhaustive, development and characterization of thermal neutron scintillators for multiple applications including spallation neutron sources [6–8].

One of the promising neutron sensitive scintillators is Cerium activated Cs_2LiYCl_6 (CLYC(Ce)) isotopically enriched with ^{6}Li . CLYC(Ce) is typically used for dual mode γ-ray and neutron detection where the ^{6}Li is used for thermal neutrons and the ^{35}Cl(n,p)^{35}S reaction allows for fast neutron detection. CLYC(Ce) has been shown to have energy resolution in the order of 4%–5%, a 1 μs decay time allowing for operation at rates in the order of 100's of kcps, and excellent pulse shape discrimination (PSD) properties [9–13]. PSD arises from the complex decay characteristics of scintillation light in CLYC(Ce) where a γ-ray will induce 3 ns core-valence luminescence in addition to the typical 50 ns and 1000 ns decay components while a neutron event will scintillate with a greater probability of the slower 1000 ns decay. This difference

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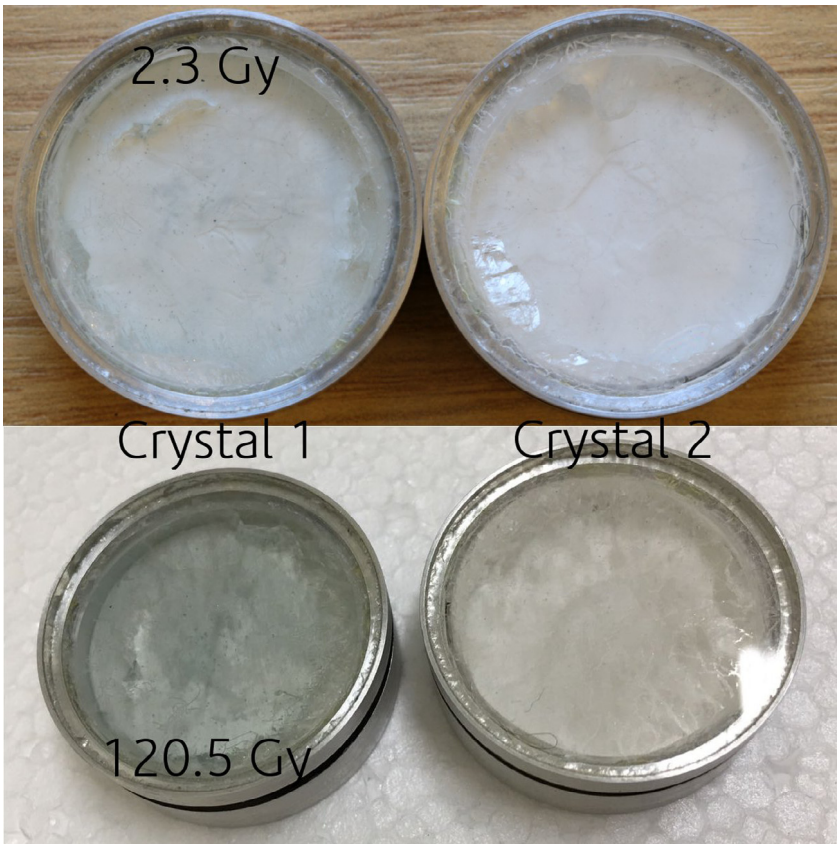

Fig. 1. A photograph of both commercially bought CLYC(Ce) crystals after the 1st irradiation (top) and the 2nd irradiation (bottom), note the discoloration and the macro-defects.

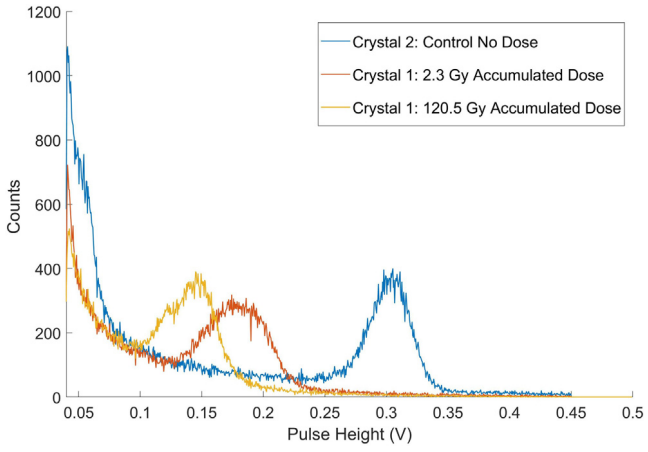

Fig. 2. Pulse Height Spectra of the CLYC(Ce) crystals, showing the control crystal and the irradiated crystal after both sets of X-ray irradiation.

of decay enables PSD, allowing for clear discrimination between neutron events and γ-ray events. The different scintillation mechanisms observed in CLYC(Ce) that gives rise to PSD have been thoroughly studied with substantial data on the decay characteristics and emission wavelengths available [11,12,14].

The authors could only find 2 published studies on the radiation hardness of CLYC(Ce). Both studies focused on the suitability to CLYC(Ce) for space radiation environments and using proton beams to characterize radiation hardness. Both studies found that proton irradiation induced the formation of color centers in CLYC(Ce) resulting in a reduction of performance [12,13].

This work presents measurements on X-ray induced changes to the luminescent properties of commercially purchased CLYC(Ce). Suitability of CLYC(Ce) for use in instruments at neutron science facilities where long term stability under significant neutron and γ-ray exposures is assessed.

2. Method

Two 2.54 cm diameter 5 mm thick CLYC(Ce) crystals canned in aluminum with a white reflector and quartz windows were purchased from the same supplier from the same batch so they would be of similar quality. The irradiated crystal was denoted Crystal 1 and the control was denoted Crystal 2. The components of the study were; X-ray irradiations, neutron measurements and luminescence measurements. The crystals were stored in a dark, temperature controlled, dry nitrogen cabinet between irradiations and measurements to investigate room temperature annealing and to ensure there was no optical stimulation of any filled traps.

2.1. X-ray irradiations

Two sets of X-ray irradiations were performed using tungsten anode X-ray sets. The dose was measured using a Radcal Accu–Gold + with the Radcal 10×6-6 6 ionization chamber. The first irradiation was to a dose of 2.3 Gy at 160 kVp, 50 μA for 45 min at a source to crystal distance of 10 cm. Crystal 1 was then given a second X-ray irradiation after a 22 week annealing period. This second and final irradiation was to a dose of 118.2 Gy delivered over 10 min at 60 kVp 50 mA with a source to crystal distance of 33.5 cm.