Effect of stoichiometry on the timing characteristics of Ce doped YAlO$_3$ single crystal scintillator

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Introduction

Ce doped YAlO$_3$ (YAP:Ce) is one of the excellent scintillators with many advantageous properties like fast decay (~30 ns), good light yield (20-25k photons/MeV), moderate density (5.3 g/cc) and proportional scintillation response. It is useful in various radiation detection applications like imaging, gamma-ray detectors, timing applications and many others [1]. Single crystals of YAP are generally grown by the Czochralski technique. The scintillation properties of the materials are highly dependent on the crystal growth parameters [2].

In this paper we are presenting the effect of variation in stoichiometry of the melt on the scintillation properties of YAP:Ce.

Experimental

Single crystals of YAP:Ce of 60 mm length and 25 mm diameter were grown from the melt using the Czochralski crystal growth technique. Y$_2$O$_3$, Al$_2$O$_3$ and CeO$_2$ powders of 99.99% purity were used as starting materials. Powders were mixed in the stoichiometric ratio of 50%Y$_2$O$_3$-50%Al$_2$O$_3$ (Crystal-I) and 51%Y$_2$O$_3$-49%Al$_2$O$_3$ (Crystal-II) for the two types of different experiments. The cerium concentration was kept at 0.5 mol% in each experiment. Mixed powders were loaded into an iridium crucible that was placed inside a growth station made from Zirconia based ceramic cylinders. An RF heating mechanism based crystal puller (Cyberstar, France make Oxypuller) was used to grow the single crystals from the melts. An un-oriented YAP:Ce single crystal of about 6 mm diameter and 40 mm length was used as the seed. A pull rate of about 1 mm/h and a rotation rate of 15 RPM was used in each growth experiment. High pure Ar was used to create inert ambient inside the crystal puller chamber. For scintillation characterization 10x10x10mm$^3$ samples were cut from the crystal ingots. Radio-luminescence of the samples was recorded using a white X-ray source (Cu-target) operating at 40kV-30mA and a mono-chromator. All the experiments were carried in 45 degree reflection geometry. The recorded spectra are not corrected for the instrument response.

For pulse height and decay time measurements, one face of the cube was polished to optical finish and on remaining faces 5-6 layers of Teflon tape were wrapped. The polished face was then coupled to a PMT (R6095) using silicon based optical grease. An AMPTEK-DP5G MCA was used to record the pulse height spectra ($^{22}$Na source) while a TEKTRONIX (MDO3102) digital storage oscilloscope was used to record decay profile.

Result and Discussion

Large size high quality transparent single crystals of YAlO$_3$:Ce were grown successfully.

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A photograph of an as-grown crystal is shown in Fig.1. Grown crystals were free from any visible defects like inclusion, bubbles etc. Single phase formation was confirmed from the powder X-ray diffraction pattern (not shown in the manuscript). The radio-luminescence spectra for both types of crystals are shown in Fig.2. The luminescence spectra exhibited the characteristic Ce$^{3+}$ emission (370 nm) corresponding to 5d$\rightarrow$4f transition [2]. An emission at longer wavelength (600nm) accompanied the Ce emission in both types of crystals. Though the 370 nm emission remains insensitive to the stoichiometric variation in the melt, the contribution of longer wavelength emission is relatively less in type-II crystals. That may be because of less number of Y$^{3+}$ related defects in the type-II crystals.

The recorded decay profiles (for gamma excitation) of the type-I and type-II crystals are shown in Fig.3. It may be clearly seen that the decay profile of type-I crystal is faster (~30 ns) than that of type-II crystal (~38 ns). This difference in the decay profile may be explained on the basis of different kinds of traps in the two types of crystals. These are also observed in the RL spectra. The pulse height spectra of both types of crystals were almost identical and a typical gamma spectrum recorded for $^{22}$Na is shown in Fig.4. This is comparable with reported spectrum for YAP:Ce [1]. From these results it may be concluded that the stoichiometric variations in the melt considerably affect the timing and other scintillation properties of Ce doped YAP crystal scintillator.

References