Crystal growth of $KCaI_3$:Eu and KSr_2I_5:Eu scintillators using the micro-pulling-down method

We demonstrated crystal growth of inorganic ternary scintillators $KCaI_3$:Eu and KSr_2I_5:Eu using a unique micro-pulling-down method that was developed for highly hygroscopic halides. These novel materials are very promising for use in gamma-ray spectroscopic detector applications related to nuclear nonproliferation and domestic security due to their superior energy resolution and scintillation light output. Fast pulling rates offered by this growth method also allow to study and improve axial segregation of the Eu luminescent activators.

We conducted crystal growth experiments using the micro-pulling-down (m-PD) method at Tohoku University. We explored experimental parameters such as the thermal gradient of the hot zone, pulling-rates (0.03 -0.09 mm/min), geometry of the crucibles and after-heaters, and melt history. We also compared two different forms of raw materials: pre-synthesized ternary compounds vs stoichiometric mixtures of binary halides. Raw materials were anhydrous, 4N-5N pure binary halides and were obtained commercially.

As a result, we successfully developed reliable growth protocols. Figure 1 shows grown single crystals of $KCaI_3$:Eu and KSr_2I_5:Eu scintillators. The crystals were 3 mm dia. and 15-20 mm long cylinders with high translucency and adequate shape control. We determined that fine powders are not suitable for pre-synthesis, and that larger chunks of pre-synthesized material must be used instead. We found that vibrations in the pulling mechanism caused unstable growth conditions and prevented from using pull rates above 0.09 mm/min. Additionally, due to high hygroscopicity of the crystals, it was challenging to handle sample preparation for characterization.

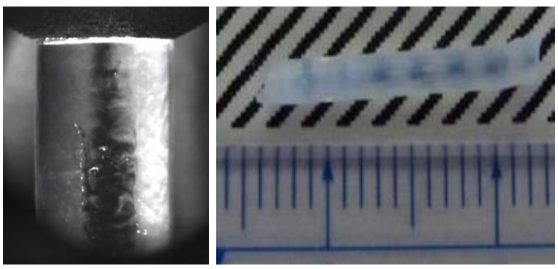

Fig. 1 *In-situ* view of stable growth conditions during the micro-pulling-down experiment and the resulting crystal of $KCaI_3$:Eu.

crystals in our laboratory at the University of Tennessee, USA. Photo- and radioluminescence as well as luminescence kinetics were measured. Figure 2 shows an important characteristic for gamma-ray spectroscopy applications, I.e. the energy resolution at ^{137}Cs (662 keV). Based on the energy resolution, light output and scintillation non-proportionality, both $KCaI_3$:Eu and KSr_2I_5:Eu are highly competitive with novel commercial scintillators such as SrI_2:Eu and $LaBr_3$:Ce. Good scintillation non-proportionality and energy resolution are also indicative of high crystalline quality obtained via the m-PD method.

The research results of this work will be presented at the American Crystal Growth

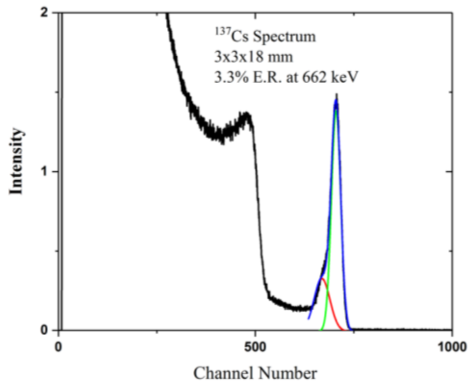

Fig. 2 Gamma-ray spectrum of $KCaI_3$:Eu showing energy resolution of 3.8% at 662 keV and ~70,000 ph/MeV light output.

Conference, July 30 – August 4, Santa Fe, NM, USA. The abstract entitled “Crystal growth of $KCaI_3$:Eu and KSr_2I_5:Eu scintillators using the micro-pulling-down method” has been accepted for a poster presentation.

Keywords: crystal growth, radiation effects, luminescence
Mariya Zhuravleva (University of Tennessee, Scintillation Materials Research Center)
E-mail: mzhuravl@utk.edu
http://www.engr.utk.edu/smrc/