Thermal Neutron Detection by Entrapping $^6$LiF Nanocrystals in Siloxane Scintillators

S.M. Carturan$^{1,2}$, T. Marchi$^2$, G. Maggioni$^{1,2}$, F. Gramegna$^2$, M. Degerlier$^{1,3}$, M. Cinausero$^2$, M. Dalla Palma$^{2,4}$, A. Quaranta$^{2,4}$

$^1$Department of Physics and Astronomy, University of Padova, Padova, Italy.
$^2$INFN, Laboratori Nazionali di Legnaro, Legnaro (Padova), Italy.
$^3$Science and Art Faculty Physics Department, Nevsehir Haci Bektaş Veli University, Nevsehir Turkey.
$^4$Department of Industrial Engineering, University of Trento, Trento Italy.

INTRODUCTION

The importance of revealing nuclear weapons concealed in trucks or boats at the borders, using Radiation Portal Monitors (RPM) has grown up, unfortunately, in the last years. Furthermore, a renewed interest in neutron detection has grown up again both in nuclear physics, mainly related to the development of Radioactive Ion Beam facilities, and in applied physics related fields, such as materials analysis by neutron imaging and diffraction spectroscopy.

The high cost of $^3$He has triggered a widespread and intensive research work aimed at its replacement [1]. Siloxane-based scintillators are, in principle, very simple and economic tools for particle detection. Their capability to detect thermal neutrons by loading with boron has been demonstrated, but the limited solubility of the organoboron compound hampered their further development [2].

In this preliminary work, we report on the production of a previously studied siloxane scintillator, added with a suitable combination of primary dye and wavelength shifter, where $^6$LiF nanocrystals have been embedded. The neutron reaction with $^6$Li nucleus produces ionizing particles (alpha and triton) and, in turn, scintillation light.

EXPERIMENTAL

Polysiloxane based scintillators are obtained by Pt catalyzed addition reaction, as described previously [2,3]. In case of scintillators, one can add suitable dyes and neutron converter to the base resin, before vulcanization. In this case, 2,5-diphenyloxazole (PPO) was chosen as primary dye and Lumogen Violet (BASF, LV) as wavelength shifter.

As neutron absorber $^6$Li was chosen because of its unique and clean decay channel consisting of a 2.73 MeV triton and 2.05 MeV alpha particle. Among the various lithium compounds, lithium fluoride displays the lowest water absorption and, very recently, LiF nanoparticles synthesis has been reported [4]. The approach of nanoparticles synthesis can be advantageous since the use of nano-sized particles can minimize light scattering and increase the transparency. In this work, $^6$LiF nanoparticles were prepared with a shell of oleic acid, hereafter called $^6$LiF OA, starting from $^6$Li trifluoroacetate (TFA). $^6$LiTFA was mixed with oleic acid and octadecene (1:1 mixture) and the solution was heated under argon at 320°C for 1h. After cooling, excess ethanol was added and the nanocrystals with oleic acid functionalization were recovered by centrifugation.

Samples of polysiloxane with added $^6$LiF were prepared with amounts of 0.5, 1 and 2 wt% of $^6$Li by simple dissolution of oleic acid-capped particles. $^6$LiF OA containing samples, which are about 0.5 mm in thickness, appear as homogeneous, though as the amount of $^6$Li increases the optical transparency decreases remarkably.

Scintillation measurements were made by exciting the samples with a $^{241}$Am α source, with a $^{60}$Co γ-rays source and with a properly thermalized neutron emitted from an AmBe source. Pulse height spectra were obtained by coupling the scintillator samples to an H6524 Hamamatsu PMT and wrapping them with aluminized Mylar on the front-face and Teflon tape on the side walls in order to maximize the light collection. The yields were compared with that obtained from EJ-212 plastic scintillator (Eljen Technology Products, 1 inch diameter, 1 mm thickness) and lithium glass GS20 (Applied Scintillation Technologies, 1 inch diameter, 1 mm thickness) in the same experimental conditions.

RESULTS

SEM inspection of the synthesized nanoparticles evidences flower-like structures, as visible in figure 2a.
Oleic acid may promote aggregation of nanocrystals during growth at high temperature, with formation of different shapes and size in the order of some microns.

High-resolution X-ray diffraction analysis has been performed on powders (figure 2b) and the typical XRD pattern of crystalline LiF (griceite) has been observed. As for the particle size, the FWHM of the main peak (200) can be observed to get an estimate of the particle size from the Scherrer formula (about 25 nm).

Fig. 2. a) SEM image of 6LiF OA capped aggregate and (b) XRD spectrum of the powder.

The samples with 6LiF OA have been tested under alpha irradiation and the results are reported in figure 3. As compared to EJ212, the light output reaches high values, up to 80% of EJ212, almost the same value of GS20. Sample thickness detrimentally affects the light collection, owing to reabsorption effects. However, the sample 5 mm thick still displays a moderately high light output (about 50% of EJ212).

Fig. 3. Pulse-height spectra collected under alpha irradiation.

Exposure to thermal neutrons allowed to record the spectra reported in figure 4 where the response of samples with 0.5% 6Li and different thickness is shown. The 0.5 mm thin samples obtained by adding higher amount of 6LiF, did not produce any significant signal, owing to the severe lack of transparency and low active volume for neutron capture.

Neutron absorption within a scintillating material is described by the Beer-Lambert equation: the attenuation level depends on the absorbing nucleus concentration and its capture cross-section.

The neutron absorption length \( l \) is defined as the material thickness necessary to attenuate the original fluence of the impinging neutron beam down to a factor 1/e (i.e. about 63% absorption) [5]. In the case of GS20, which contains higher amount of 6Li than our 0.5 wt% loaded samples the absorption length is about 0.7 mm for thermal neutrons, while in the case of siloxane is about 20 mm. Therefore, it can be inferred that the higher thickness in siloxane based samples accounts for higher peak intensity and better signal-to-background ratio in the pulse-height spectrum, as visible in figure 4. The response of GS20 appears as composed of more than one component. This can be ascribed to edge effects, where the produced ionizing particles, either alpha or triton, escape from the disk borders with incomplete energy deposition. However, the observed behavior is still under discussion.

Fig. 4. Pulse-height spectra obtained under irradiation with thermal neutrons. The spectrum of GS20 has been reported in the inset, for the sake of clarity.

The \( \gamma \)-rays background from the AmBe source not completely shielded is visible as a shoulder in the low channels region. The thicker the sample the more significant becomes the contribution from Compton interactions. Nevertheless, the neutron signal is clearly well separated from the shoulder due to \( \gamma \)-rays background, thus indicating that n-\( \gamma \) separation is possible simply on the basis of pulse height discrimination.

REFERENCES AND NOTES

The work was supported by INFN Vth Commission (experiment HYDE) and by the Ministry of Education (Prim 2010-2011 -2010TPSCSP_005- Developments of new detectors and analysis techniques for experiments with radioactive beams at the National Laboratories of INFN, with special reference to the SPES project).