ALGERIAN JOURNAL OF ENGINEERING AND TECHNOLOGY

Algerian Journal of Engineering and Technology
ISSN: 2716-9227 (Print)
EISSN: 2716-9278 (Online)
DOI: http://dx.doi.org/10.57056/ajet.v9i2.184
Zahra et al / vol. 09, n°2 - 2024

Original Article Open Access

Development and characterization of novel fast nanosized scintillator Y_2SiO_5: Ce^{3+} prepared by polymer-assisted Sol–Gel method for radiation detection applications

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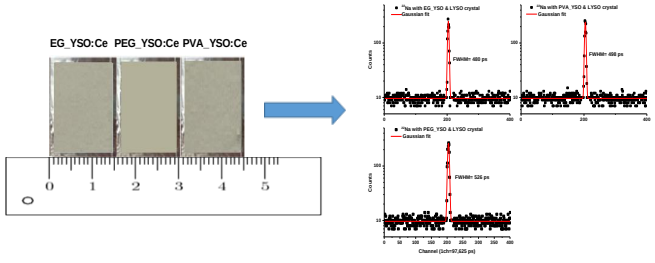
Article history: Received 09 June 2024, Revised 17 December 2024, Accepted 20 December 2024

ABSTRACT

In this study, three sample detectors were meticulously crafted using cerium-activated X_1-Y_2SiO_5: Ce^{3+} powder prepared via the monomer and polymer-assisted sol–gel method. The investigation aimed to assess how ethylene glycol (EG) monomer, polyethylene glycol (PEG) polymer, and polyvinyl alcohol (PVA) polymer influence the Coincidence Timing Resolution (CTR) of Ce^{3+} (xCe = 0.01)-doped Y_2SiO_5, with the goal to enhance radiation detection technologies. An advanced nuclear instrumentation system was set up to measure the coincidence timing resolution using 511 keV annihilation photons emitted by a ^{22}Na radioactive source. Results showed that complexing agents significantly affected the CTR of YSO: Ce^{3+} nanoscintillators, with the EG-prepared sample detector exhibiting the most favorable CTR of 480±21 ps. These findings enhance our understanding of YSO: Ce^{3+} nanoscintillators' synthesis and optimization, underscoring the pivotal role of the chemical environment and emphasizing the superior performance of ethylene glycol. These insights provide valuable avenues for further advancements in radiation detection and medical imaging applications.

Keywords: Nuclear instrumentation. Radiation detector. X_1-Y_2SiO_5:Ce^{3+} nanomaterial. Scintillation properties. Coincidence Timing Resolution.

Graphical abstract



Recommended Citation
Zahra B, Guerbous L, Hamroun MSE. Development and characterization of novel fast nanosized scintillator Y2SiO5: Ce3+ prepared by polymer-assisted Sol–Gel method for radiation detection applications. *Alger. J. Eng. Technol. 2024*, 9(2): 194-193., http://dx.doi.org/10.57056/ajet.v9i2.184

1. Introduction

Scintillation materials, recognized for their luminescent reaction to ionizing radiation, play pivotal roles in critical sectors, including radiation detection, medical imaging, and high-energy physics [1–5]. Within radiation detection, these materials form crucial components of advanced detectors deployed in security measures such as cargo, luggage, and individual screening for radioactive substances at ports, airports, and border checkpoints. In medical imaging, scintillation materials are fundamental to technologies like positron emission tomography (PET), single-photon emission computed tomography (SPECT), and gamma camera imaging. By converting incoming radiation into detectable light signals, scintillation detectors facilitate the visualization of internal anatomical structures, tissues, and physiological processes with remarkable clarity and precision. This non-invasive imaging capability, combined with the provision of quantitative data, has revolutionized medical diagnosis and treatment planning, leading to improved patient outcomes and advancements in healthcare delivery systems. In both radiation detection and medical imaging applications, the distinct properties of scintillation materials equip practitioners with essential tools for accurate and efficient radiation detection and imaging, thus contributing significantly to public health and safety initiatives.

This study is motivated by the necessity to develop scintillation materials with enhanced properties to meet the evolving demands of radiation detection and medical imaging applications. There is an increasing necessity for more efficient and reliable radiation detection systems, as well as advancements in medical diagnostics that require imaging technologies with higher sensitivity, improved spatial resolution, and reduced radiation exposure for patients. By focusing on the development of new scintillation materials with superior performance characteristics, this research aims to contribute to advancements in both fields, offering innovative solutions to improve public safety, national security, and medical care delivery.

The single crystals Lu_2SiO_5:Ce (LSO:Ce) [6] and $(Lu,Y)_2SiO_5$:Ce (LYSO:Ce) [7] are exceptional scintillators used in positron emission tomography (PET) scanners due to their high density, non-hygroscopic nature, superior light yield, and rapid scintillation decay. Despite these advantageous properties, these inorganic scintillators face drawbacks such as high production costs, lengthy processing times, fragility, and limitations in crystal size [8]. To overcome these challenges, the development of more cost-effective scintillator nanocrystals offers a promising alternative. In recent years, various chemical methods have been successful in synthesizing YSO nanopowder, providing better control over the manufacturing process. [9–13]. Notably, the sol-gel method, in particular, has gained significant attention due to its versatility, high purity, low processing temperature, and ability to control the size and morphology of the resulting grains [14].

Recently, N. Verma et al. [15] conducted an extensive review focusing on the luminescence properties of $Y_2SiO_5:Ce^{3+}$ synthesized through various methods and doped with elements such as Eu, Ce, and Lu. Their main goal was to elucidate the complexities of the luminescent behavior exhibited by these materials, aiming to leverage their potential for the development of Y_2SiO_5 nanophosphors with enhanced characteristics. In addition, K. Wantong et al. [16] investigated the luminescence and scintillation properties of Y_2SiO_5:Ce single crystalline films grown via liquid phase epitaxy. The scintillation light yield of the examined material was measured at 12,410 ph/MeV, which is lower than the 20,150 ph/MeV calculated for bulk Czochralski-grown YSO: Ce single crystals. However, to our knowledge, there has been no research into the scintillation properties of nano-sized Y_2SiO_5: Ce, particularly in terms of coincidence timing resolution (CTR), a key parameter for evaluating scintillator performance in medical imaging techniques such as PET and CT scans. [17].

To address this knowledge gap, this study examines the use of X_1-Ce-doped Y_2SiO_5 silicate nanopowders, synthesized through an ethylene glycol (EG) monomer, polyethylene glycol (PEG), and polyvinyl alcohol (PVA) polymer-assisted sol–gel process, for designing radiation sample detectors. The primary objective is to assess the impact of various monomers and polymers used as fuels and complexing agents during crystallization on the CTR characteristic of X_1-Y_2SiO_5 nanophosphors. To achieve this, we implemented a nuclear instrumentation system to measure the CTR of the designed sample detectors, with measurements conducted using 511 keV annihilation photons emitted from a ^{22}Na radioactive source.

2. Materials and Methods

In our previous study [18], we synthesized Cerium (Ce^{3+})-doped monoclinic X_1–Y_2SiO_5 (YSO)-type oxyorthosilicate powders using the monomer and polymer-assisted sol–gel method. After synthesis, we extensively investigated the

structural and photoluminescent properties of these samples. In this research, we utilized the synthesized YSO: Ce^{3+} powders to fabricate radiation detectors. Our primary goal was to identify the optimal fuel for designing highly efficient radiation detectors. The fabrication process involved employing two transparent Mylar sheets. The first sheet, served as the substrate for distributing the powder. It was tailored to dimensions of 20×15 mm² and molded to yield detectors with a 1 mm thickness. To ensure uniform powder distribution and adherence, we sprayed an ethanol solution onto the Mylar substrate before depositing the YSO particles. Subsequently, gentle pressing action was applied to spread and compact the YSO nanocrystals on the substrate, resulting in thin-film-like samples. After firm powder adhesion, we carefully removed the samples from the mold to prevent disruption and covered the opposite side with the second Mylar sheet. To shield the samples and aid handling during experimentation, we wrapped the edges in aluminum foil. During experimental assessments, the side covered by transparent Mylar was coupled with the photomultiplier tube window. Figure 1 illustrates the developed samples, showcasing the final product. All sample detectors in this study underwent the same fabrication procedure and adhered to identical dimensions of 20×15×1 mm³. For simplification, the prepared sample detectors of X_1-Ce^{3+}-doped Y_2SiO_5 (YSO) were named: EG_YSO, PEG_YSO, and PVA_YSO, according to the ethylene glycol (EG) monomer, polyethylene glycol (PEG) and polyvinyl alcohol (PVA) polymer used during the synthesis process.

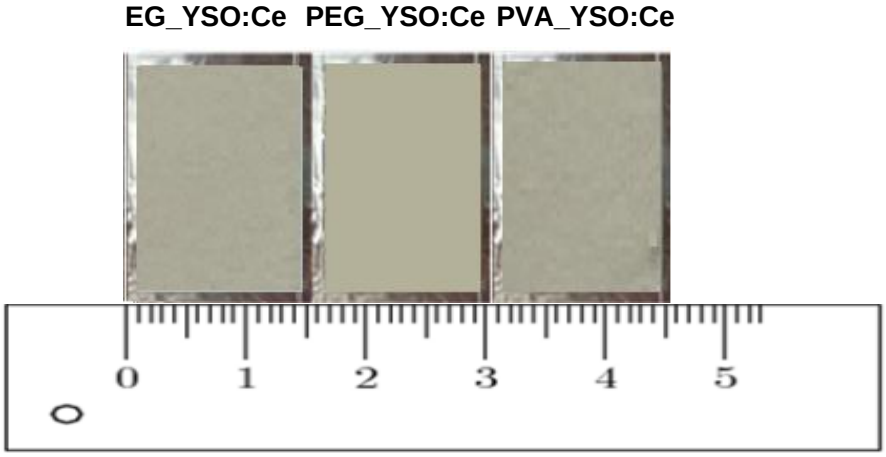


Fig 1. Photograph of prepared sample detectors

Coincidence timing resolution of designed sample detectors was meseared using 511 keV annihilation quanta emitted by a ^{22}Na source. Each specimen under examination was paired with a photonic XP2020Q PMT to establish a "Stop" channel within the timing setup. Additionally, a fast scintillating LYSO single crystal, coupled to a Photonis Hamamatsu R5320 PMT, served as the reference detector, boasting a time resolution of 269 ps for the 511 keV full-energy peak [19]. Upon irradiation of the crystals, which were coupled to each PMT, with 511 keV annihilation quanta emitted by a ^{22}Na source positioned between them, signals from each PMT were processed using an ORTEC 584 Constant-Fraction Discriminator (CFD). Coincidence time spectra were collected utilizing a CANBERRA TAC model 2145 and recorded via an ORTEC 927 MCA onto a PC. The fast-slow coincidence setup was consistently employed in all measurements to ensure precise selection of the required (511 keV) energy windows. Figure 2 illustrates the schematic diagram of the setup used to measure the coincidence timing resolution of investigated samples.

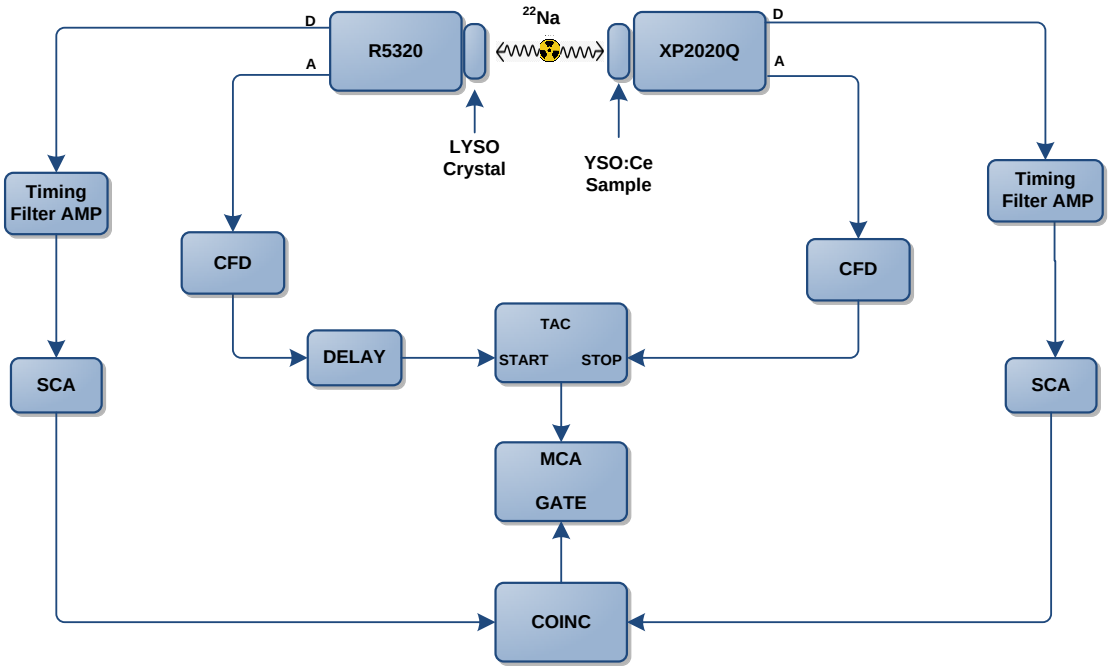


Fig 2. Schematic diagram of setup implemented to measure the coincidence timing resolution CTR of developed sample detectors.

3. Results and Discussion

The coincidence timing resolution of each sample detector under study was measured as described earlier. Figure 3 illustrates the coincidence timing resolution spectra obtained. Following this, the spectra underwent Gaussian fitting to derive the CTR values, determined by the Full Width at Half Maximum (FWHM).

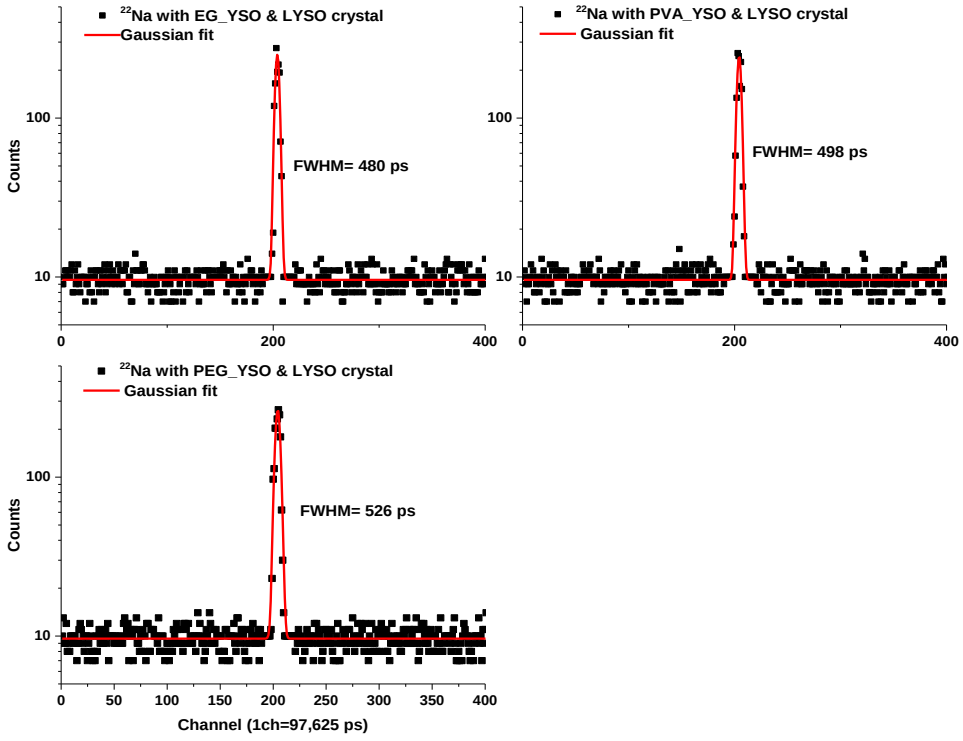


Fig 3. Coincidence time spectra measured for investigated sample detectors

The results outlined in Table 1 highlight the substantial impact of acidity on Coincidence Timing Resolution (CTR). Specifically, the data indicates that the EG_YSO sample detector exhibited the most favorable CTR value (480±21 ps). Our previous research [18] indicated that the morphology of Y_2SiO_5:Ce particles is significantly affected by the choice of monomer and polymer. Notably, the powder synthesized with EG monomer exhibited agglomerated particles with irregular morphology, spherical shape and devoid of pores. On the other hand, samples prepared with PEG polymer displayed a less dense and broader asymmetrical distribution of particle sizes, characterized by irregular morphology and an open structure with interstitial spaces several microns in diameter, forming small irregular aggregates. Meanwhile, observations of samples prepared with PVA polymer revealed arrangements of agglomerate particles with a narrow size distribution ranging from a few to tens of microns, displaying some porous morphology. In fact, phosphor particles with a spherical shape minimize light scattering on their surfaces, enhancing light emission efficiency and brightness [20]. This characteristic could potentially result in an increased scintillation light yield in the EG_YSO sample, thereby improving coincidence-timing resolution. Additionally, it was observed that the crystallite size, calculated using Debye–Scherrer's formula, is larger in samples prepared with EG complexant compared to those prepared with other polymers such as PEG and PVA complexants [18]. Figure 4 illustrates the relationship between the crystallite size of Y_2SiO_5 and the CTR values as function of the type of complexing agents, revealing an inversely proportional correlation.

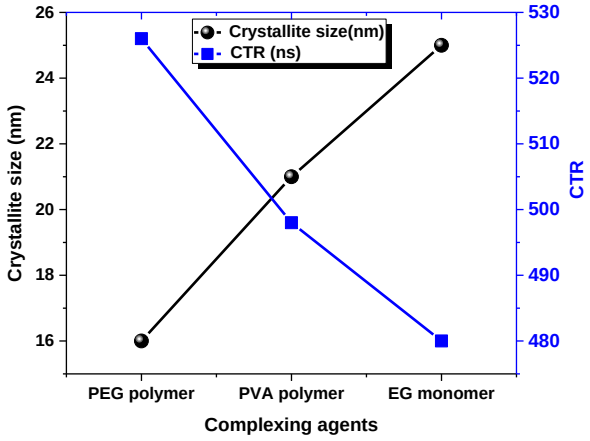


Fig 4. CTR and crystallite size relationship under different type of complexing agents

In fact, the larger crystallite size obtained with EG monomer results in a decreased surface-to-volume ratio (S/V) compared to the smaller size obtained with PEG and PVA polymers. This reduction in S/V leads to a decrease in the density of defects at grain boundaries. Higher defect densities are known to facilitate non-radiative recombination of electron–hole pairs [21]. Consequently, the reduced S/V ratio obtained with the EG_YSO sample allows for the improvement of scintillation light yield compared to the PEG_YSO and PVA_YSO samples, thereby enhancing the CTR.

Table 1. The coincidence time resolution CTR of studied samples.

Sample detectors	**Timing resolution (ps)**
EG_YSO	480±21
PVA_YSO	498±19
PEG_YSO	526±10

4. Conclusion

In conclusion, this study has provided valuable insights into the influence of complexing agents on the scintillation properties of Y_2SiO_5: Ce nanoscintillators. Results reveal the profound influence of the chemical environment, especially the complexing agent, on optimizing scintillation performance and coincidence timing resolution (CTR) of the studied

samples. Specifically, the findings highlight the superior performance of ethylene glycol (EG) as a complexing agent in optimizing the temporal response of the scintillator, leading to improved CTR values.

The observed correlation between crystallite size and CTR further emphasizes the importance of morphology control in achieving desired scintillation properties. The larger crystallite size obtained with EG monomer results in a lower defect density at grain boundaries, facilitating enhanced scintillation light yield and, consequently, improved CTR.

These promising results pave the way for the practical application of the developed detectors in radiation detection and medical imaging. The enhanced scintillation properties, coupled with improved CTR values, make the EG-based Y_2SiO_5: Ce nanoscintillators well-suited for various applications, including positron emission tomography (PET), single-photon emission computed tomography (SPECT), and computed tomography (CT) scans. The potential for high sensitivity and temporal resolution offered by these detectors holds great promise for advancing diagnostic imaging techniques and improving patient outcomes in the field of medical imaging.

Overall, our study underscores the importance of optimizing scintillation properties through careful selection of complexing agents and morphology control. By leveraging these advancements, we can contribute to the development of more efficient and reliable radiation detection systems and medical imaging technologies, ultimately benefiting healthcare delivery and public safety initiatives.

Funding Support

This research received no external funding.

Ethical Statement

This study does not contain any studies with human or animal subjects performed by any of the authors.

Conflict of Interest

The authors declare that they have no conflict of interest.

Data Availability Statement

Not applicable.

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