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Study on the Dependence of Energy Resolution on Reflector Material for Inorganic Crystal Scintillators Using the Geant4

Murat DAĞ*[1]

[1]*Vocational School of Health Services, Kırşehir Ahi Evran University, 40000, Kırşehir, Turkey*

**corresponding author e-mail:muratdag71@hotmail.com.tr*

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Abstract: The scintillator has played a primary role as the ideal device for the detection and measurement of particles and radiation in modern physics. With the development of experimental physics, the demand for new improved scintillating materials for several types of applications has kept increasing. High efficiency, fast scintillation and good energy resolution are among the most desired specifications as to a good scintillator. Yet, a variety of scintillators can be preferred depending on the precise specifications of the application considered. If the case is that the detection of gamma rays and high-energy electrons or positrons, inorganic crystals are exceptionally suitable scintillator because highly intense light outputs and the strong stopping power enable these type of crystals to provide better energy resolution among all scintillators. In this study, a scintillation detector consisting of inorganic crystal scintillator material (NaI:Tl and CsI:Tl) was modeled with the help of Geant4 scientific toolkit to determine if the energy resolution of the inorganic crystal scintillator detector is dependent on crystalline size and reflector material. In each simulation, different sized crystal covered with a variety of reflector type was exposed to the same energy gamma radiation; the resulting energy spectrum was evaluated and compared to others obtained.

Key words: Scintillation detector, Inorganic crystal, Reflector material, Geant4

Geant4 ile İnorganik Kristal Sintilatörler için Enerji Çözünürlüğünün Reflektör Malzemesine Bağımlılığı Üzerine Çalışma

Özet: Sintilatör, modern fizikte parçacıkların ve radyasyonun saptanması ve ölçülmesi için ideal bir cihaz olarak birincil bir rol oynamıştır. Deneysel fiziğin gelişmesiyle birlikte, çeşitli uygulama türleri için yeni geliştirilmiş parıldayan malzemelere olan talep artmaya devam etti. İyi bir sintilatör için yüksek verimlilik, hızlı parıldama ve iyi enerji çözünürlüğü en çok istenen özellikler arasındadır. Yine de, dikkate alınan uygulamanın kesin özelliklerine bağlı olarak çeşitli sintilatörler tercih edilebilir. Durum, gama ışınlarının ve yüksek enerjili elektronların veya pozitronların tespiti ise, inorganik kristaller son derece uygun sintilatördür çünkü yüksek yoğunluklu ışık çıkışları ve güçlü durdurma gücü, bu tür kristallerin tüm sintilatörler arasında daha iyi enerji çözünürlüğü sağlamasını sağlar. Bu çalışmada, inorganik kristal sintilatör dedektörünün enerji çözünürlüğünün kristal boyutuna bağlı olup olmadığını belirlemek için inorganik kristal sintilatör materyalinden (NaI:Tl ve CsI:Tl) oluşan bir sintilasyon dedektörü Geant4 bilimsel araç kiti yardımıyla modellenmiştir. Her simülasyonda, çeşitli reflektör tipi ile kaplanmış farklı büyüklükteki kristal aynı enerji gama radyasyonuna maruz bırakılmış, elde edilen enerji spektrumu değerlendirilmiş ve elde edilen diğerleriyle karşılaştırılmıştır.

Anahtar kelimeler: Sintilasyon dedektörü, İnorganik kristal, Reflektör materyal, Geant4

Murat DAĞ, muratdag71@hotmail.com.tr, ORCID: https://orcid.org/0000-0002-0503-6067

1. Introduction

The scintillator makes use of the fact that certain material interacting with radiation emits a scintillation light. The earliest use of scintillator for particle detection was in the 1900's [1]. Due to its tedious use, it was never very popular at that time. However, with the newly developed photomultiplier tube, new improvements in the field of scintillator were quickly followed, so by the mid-1950s scintillation detector became the most reliable and conveniently available. Even now scintillation materials and detectors are one of the prevalent devices for the detection of ionizing radiation and have been used in many applications from nuclear and high-energy physics to medical imaging area today [2].

There are certain properties that the ideal scintillation material should have such as short decay time, good optical quality, a high scintillation efficiency with detectable light, high light yield proportional to deposited energy, good transmission of light from crystal to a photomultiplier tube (index of refraction matching as closely as possible glass refractive index ~1.5) and reasonable cost. There are various types of scintillator materials in use at present; crystals, liquids, plastics, gases and glasses. No scintillator however can meet all these criteria at the same time. Therefore, the intended application has a great influence on the choice of scintillator [1,3].

One of the widely used scintillating materials for the detection of ionizing radiation is inorganic crystals [4]. They are mainly alkali halides, but sometimes contain a small activator impurity due to the fact that self-activated scintillators exhibit better scintillation properties when they are dopes by suitable ions [5]. The mechanism involved in the production of scintillation light differs based on the types of scintillator. The scintillation mechanism occured in inorganic scintillators is basically the characteristic of the electronic structure of the crystal. As a particle interacts with a crystal, mostly two processes may occur. Crystal is ionized either by exciting an electron from the valence band to the conduction band, resulting in creating a free electron and hole, or by exciting an electron to the exciton band created locally just below the conduction band, producing an exciton pair (see Figure 1). This created electron and hole (exciton) pair remain bound together, and can move freely in this state. If the crystal is doped by the suitable ions called impurity atoms (such as Thallium in NaI and CsI crystals), that contributes to create new electronic level in the forbidden energy gap. A migrating free hole or a hole from an exciton pair meet an impurity center is able to ionize the impurity atom. The subsequent electron can fall into the vacancy left by the hole if it arrives and make a transition from an excited state to the ground state, emitting radiation. If the transition is radiationless, the energy is lost to other processses and the impurity center becomes a trap [6].

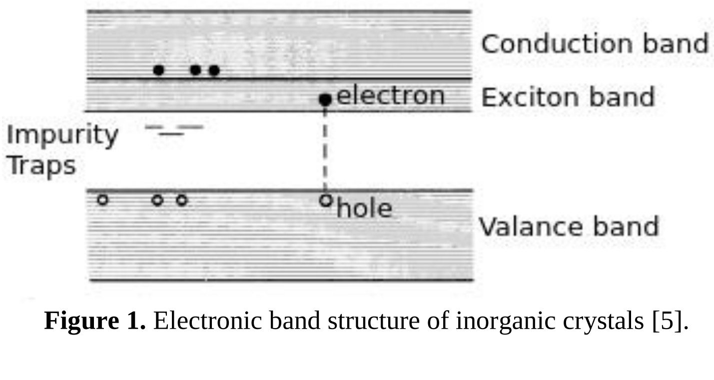


Figure 1. Electronic band structure of inorganic crystals [5].

Compared to organic crystals, inorganic crystals are 2-3 orders of magnitude slower in response due to phosphorescence. Additionally, hygroscopicity is the primary disadvantage of this type scintillating crystals so they need to be protected attentively from moisture in the air [7]. However, the strong stopping power and the highly intense light outputs give a great advantage, enabling this type of crystals to provide better energy resolution among all the scintillators.Thus, inorganic crystals are especially well suited to the detection of gamma and charged particle. The most common and actively used inorganic crystals are NaI:Tl and CsI:Tl, where Thallium (Tl) is the impurity activator [8,9].

The energy resolution is of the significance parameter to refer to the ability of a detector to distinguish between radiations of very close energies. For most applications of scintillation detectors, a good energy resolution is one of the most important parameters to be considered. But, non-propotional light yield, poor light reflection in the encapsulate, index of refraction mismatch, and crystal transparency and imperfections in scintillator seems the basic limitation of energy resolution [7,10,11]. Therefore, investigation of energy resolution for inorganic crystal scintillators on different reflector material using scientific simulation toolkit may help to indicate another factor that broaden energy resolution in the inorganic scintillator.

2. Material and Method

Geant4 (GEometry ANd Tracking) [12] is an open source and publicly available Monte Carlo toolkit developed in the European Organization for Nuclear Research (CERN) to create accurate simulations of particle propagation and interaction with matter. Basically, Geant4 obtains a solution to the problem by simulating individual particle paths and recording some aspects of their average behavior. Individual probabilistic events, which are the process of interaction of a particle with a material, are sequentially modeled. Each particle can generate additional particles or tracks at the collision site. The story of each particle (from birth to death) is considered an independent random event. The physical processes that Geant4 provides (hadronic, electromagnetic, and optical) cover a complex set of particles and materials in a wide range of energies. Besides physics, it also offers a full suite of features like tracking, visualization, and description of geometry, and widely used in various areas of application including high energy, nuclear, accelerator physics, medical, and space science.

A inorganic scintillation detector consists of two main components: scintillating material and a photomultiplier tube that is optically coupled to it. The scintillator is covered up with a reflector except on the side connected to the photomultiplier tube. As the incident radiation passes through the scintillator, it excites certain states in the scintillating material causing light to be emitted. This emitted light is transmitted to photomultiplier tube, where it is converted into a current of photoelectrons. By amplified the weak current, the resulting current signal is ready to analyze.

The same main components for the purpose of this study hav been created using the scientific simulation toolkit (see Fig 2). In this designed framework inorganic scintillator material is in green, the reflector material which covers up the crystal is in blue, the photomultiplier tube (PMT) is in red, and the cathode is in white.

Dimensions of inorganic crystals used in this study are 10x10x10 mm, 15x15x15 mm, 20x20x20 mm, 25x25x25mm and 30x30x30 mm. Aluminium foil, MgO and TiO2 were used as the reflector material covering the crystal.

Each crystal was exposed to the same gamma radiation (^{137}Cs at 662 keV). 10000 particles were registered in each run of the simulation. Reflected photons are marked in green as shown in Figure 2. The resulting energy spectrum for each run was collected and energy resolution were calculated.

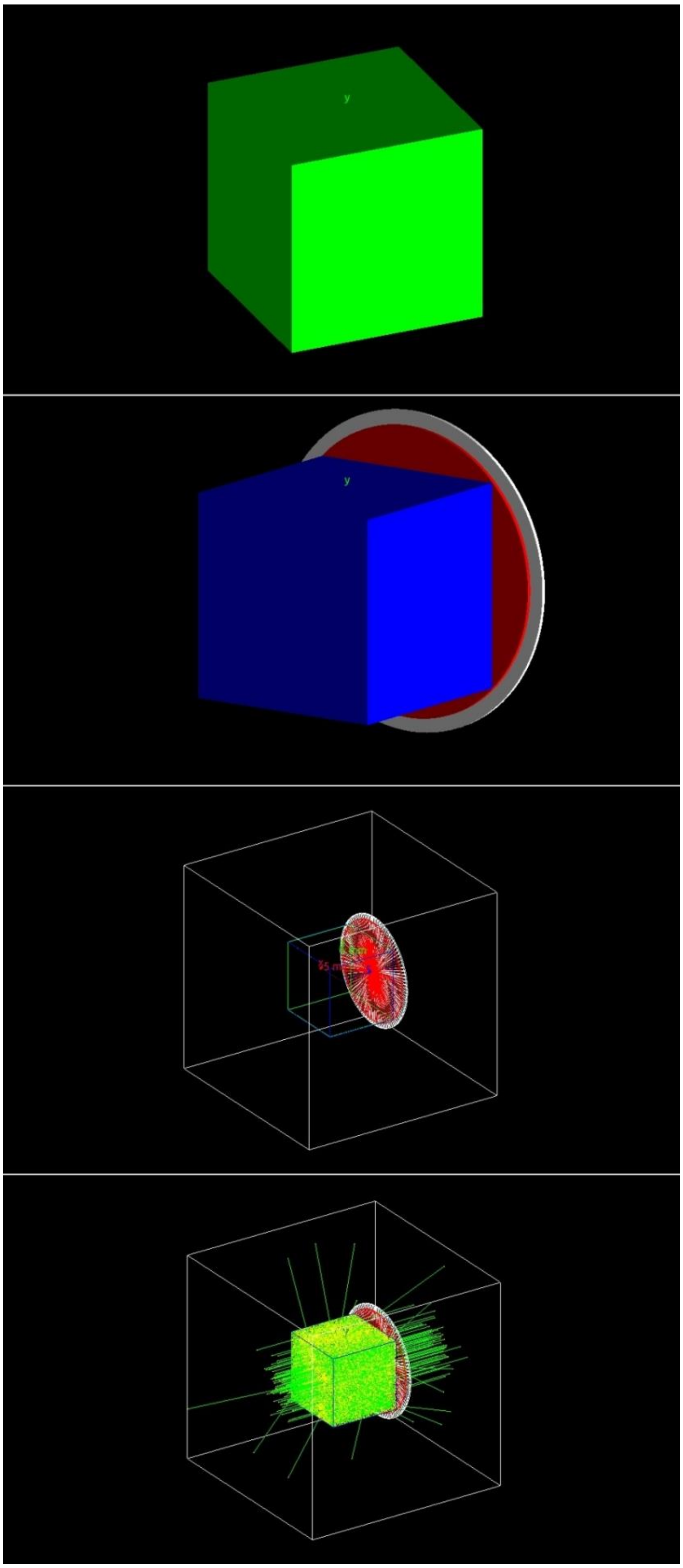


Figure 2. In the framework of the simulation using Geant4 toolkit, inorganic scintillator material is in green, the reflector material which covers up the crystal is in blue, the photomultiplier tube (PMT) is in red and the cathode is in white, all objects are in wire-fram and reflected photons are marked in green (top to bottom).

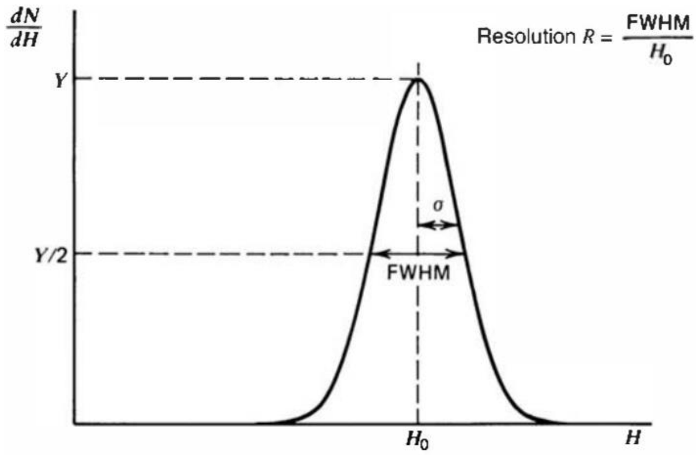


Figure 3. The full width at half maximum (FWHM) is illustrated and defined as the width of the distribution at a level which is just half of the maximum ordinate of the peak (H_0) [3].

If n events contribute to the overall response function for the gamma-ray detection system, the shape of that tends towards a Gaussian shape as seen in Figure 3. The shape of a peak in the gamma-ray spectrum thus can usually be approximated by a Gaussian distribution. The width of a Gaussian curve is generally indicated by the parameter σ. The square of the overall standard deviation,σ^2,can be expressed by

$$\sigma^2 = \sum_{i=0}^{n} \sigma_i^2, \tag{1}$$

where σ_i^2 is the variance of the event i. The full with at the half maximum (FWHM) of a Gaussian distribution, which indicates the difference between the variables corresponding to the half of the maximum of the peak height as seen in Figure 3, is connected to σ in the following way:

$$FWHM = 2{,}35\sigma = 2{,}35\sqrt{\sum_{i=0}^{n} \sigma_i^2}. \tag{2}$$

It is defined as the ratio of the full width at half maximum (FWHM) of the measured peak over the energy value of the peak centroid (H_0). The energy resolution of the scintillator is denoted by R, and is a dimensionless fractions expressed as a percentage.

$$Energy\ Resolution\ (R) = \frac{FWHM}{H_0}. \tag{3}$$

Table 1. Specifications of NaI:Tl and CsI:Tl inorganic scintillating crystal.

	Specific Gravity	**Wavelength of Max. Emission**	**Refractive Index**	**Decay Time (µs)**	**Absorbed Light Yield in Photons/MeV**
NaI:Tl	3.67	415	1.85	0.23	38000
CsI:Tl	4.51	420	1.84	0.46 – 4.18	39000

3. Results

A multiple simulation consisting of a combination of 5 different-sized inorganic scintillation crystals (NaI:Tl and CsI:Tl) were created using the specifications stated in Table 1. Each crystal was covered with 3 different reflective materials (Aluminium foil, MgO and TiO_2), and the thickness of the reflector material was decided not to change minimize the contribution. For photomultiplier tube components, quartz and photocathode materials were designed from fused silica K_2CsSb, respectively. Surface and boundary processes were set. EMStandartPhysics and OpticalPhysics were added in PhysicsList in Geant4. In each run of the simulation, 10000 particles were registered to created the same conditions. The full-energy photo-peak of gamma spectrum of ^{137}Cs which in case a single gamma line of 662 keV was determined and energy resolution calculated using the ROOT program [13] as seen in Fig 4. The details of the photo-peak for each combination can be seen in Table 2.

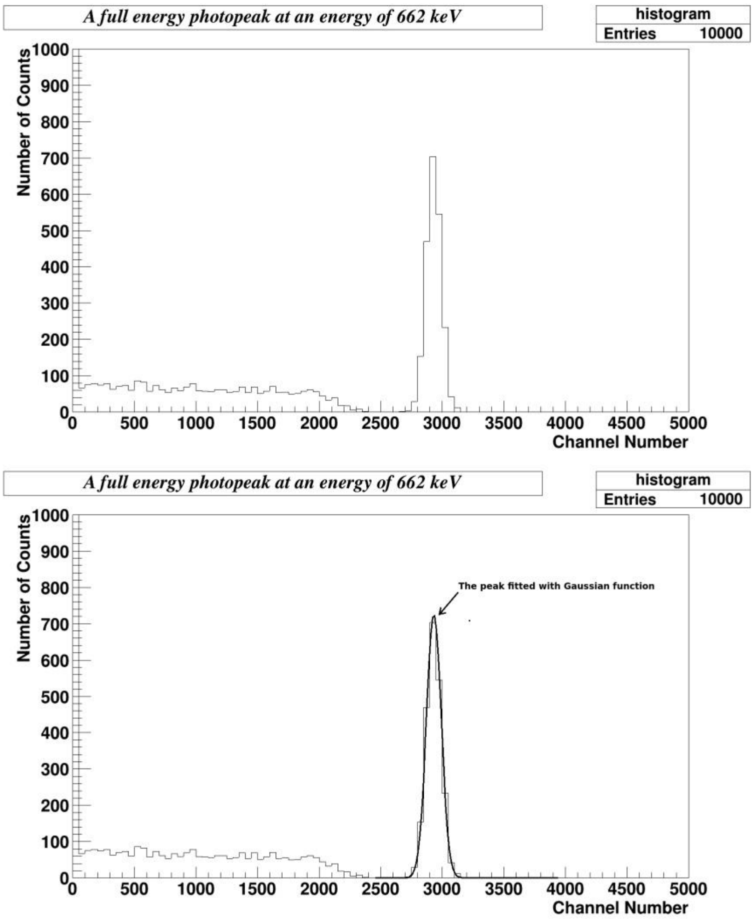
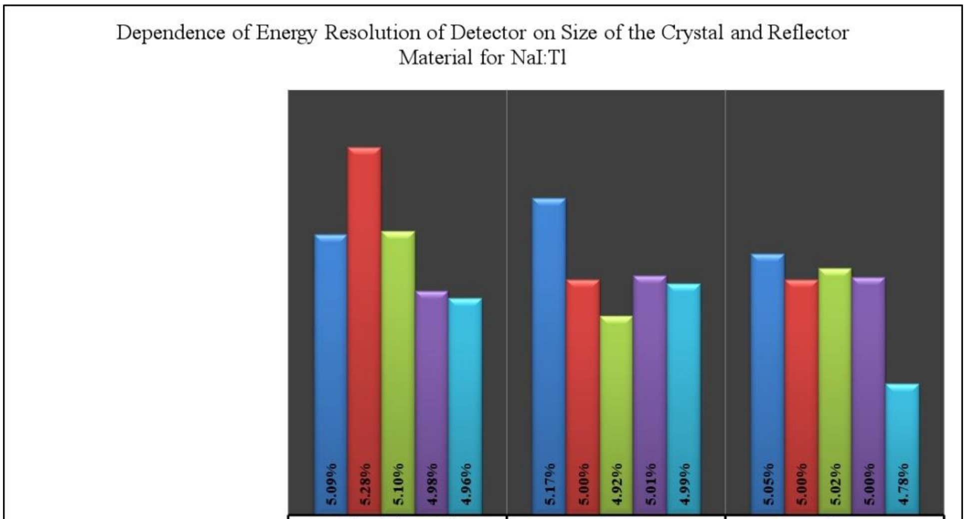


Figure 4. The gamma-ray spectrum obtained with NaI:Tl crystal exposed to 662 keV gamma-rays of ^{137}Cs (top), the peak fitted with Gaussian function (bottom).

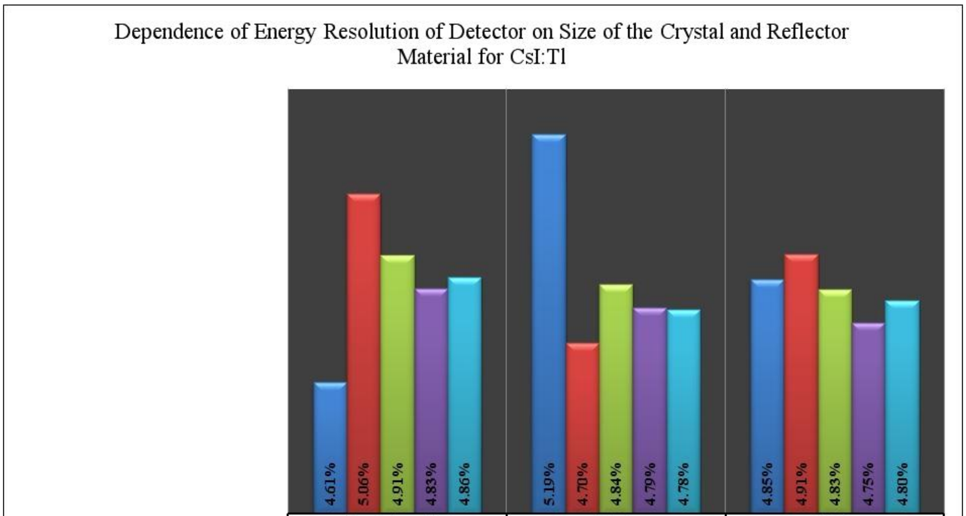
4. Conclusion

Intense light outputs and the strong stopping power have increased the interest in the inorganic scintillator, especially due to the detection of gamma and charged particle. To enhance the light output of a scintillator, a variety of approaches have been considered such as the use of a PMT with a window refractive index matching as closely as possible the one of the crystal [14–18], the modification of doping concentration in the crystal [19–21] or the modification of the crystal shape [22,23]. In addition to that, the coating of the crystal with different reflector type may be explored which allow us to significantly enhance the light collection on the detector. Therefore the framework was modeled with the help of Geant4 scientific toolkit to determine the dependence of the energy resolution of the inorganic crystal scintillator detector on crystalline size and reflector material. It is concluded that different reflector materials help to increase light reflection resulting in narrowing the FWHM by this way improving energy resolution, however this increase was within the error limits (see Figure 5 and Figure 6).



	Aluminium Foil	MgO	TiO2
Crystal Size (10 x 10 x 10 mm)	5.09%	5.17%	5.05%
Crystal Size (15 x 15 x 15 mm)	5.28%	5.00%	5.00%
Crystal Size (20 x 20 x 20 mm)	5.10%	4.92%	5.02%
Crystal Size (25 x 25 x 25 mm)	4.98%	5.01%	5.00%
Crystal Size (30 x 30 x 30 mm)	4.96%	4.99%	4.78%

Figure 5. Comparison of results of the calculation of the energy resolution of the full-energy photopeak with different crystalline sizes and reflector materials obtained in simulation for NaI:Tl.



	Aluminium Foil	MgO	TiO2
Crystal Size (10 x 10 x 10 mm)	4.61%	5.19%	4.85%
Crystal Size (15 x 15 x 15 mm)	5.06%	4.70%	4.91%
Crystal Size (20 x 20 x 20 mm)	4.91%	4.84%	4.83%
Crystal Size (25 x 25 x 25 mm)	4.83%	4.79%	4.75%
Crystal Size (30 x 30 x 30 mm)	4.86%	4.78%	4.80%

Figure 6. Comparison of results of the calculation of the energy resolution of the full-energy photopeak with different crystalline sizes and reflector materials obtained in simulation for CsI:Tl.

Table 2. Results of the calculation of the energy resolution of the full-energy photopeak with different crystalline sizes and reflector materials obtained in simulation.

Crystal Type	Crystal Size (mm)	Reflector Type	Resolution (%)
CsI:Tl	10 x 10 x 10	Aluminium Foil	4.61
CsI:Tl	10 x 10 x 10	MgO	5.19
CsI:Tl	10 x 10 x 10	TiO_2	4.85
CsI:Tl	15 x 15 x 15	Aluminium Foil	5.06
CsI:Tl	15 x 15 x 15	MgO	4.70
CsI:Tl	15 x 15 x 15	TiO_2	4.91
CsI:Tl	20 x 20 x 20	Aluminium Foil	4.91
CsI:Tl	20 x 20 x 20	MgO	4.84
CsI:Tl	20 x 20 x 20	TiO_2	4.83
CsI:Tl	25 x 25 x 25	Aluminium Foil	4.83
CsI:Tl	25 x 25 x 25	MgO	4.79
CsI:Tl	25 x 25 x 25	TiO_2	4.75
CsI:Tl	30 x 30 x 30	Aluminium Foil	4.86
CsI:Tl	30 x 30 x 30	MgO	4.78
CsI:Tl	30 x 30 x 30	TiO_2	4.80
NaI:Tl	10 x 10 x 10	Aluminium Foil	5.09
NaI:Tl	10 x 10 x 10	MgO	5.17
NaI:Tl	10 x 10 x 10	TiO_2	5.05
NaI:Tl	15 x 15 x 15	Aluminium Foil	5.28
NaI:Tl	15 x 15 x 15	MgO	5.00
NaI:Tl	15 x 15 x 15	TiO_2	5.00
NaI:Tl	20 x 20 x 20	Aluminium Foil	5.10
NaI:Tl	20 x 20 x 20	MgO	4.92
NaI:Tl	20 x 20 x 20	TiO_2	5.02
NaI:Tl	25 x 25 x 25	Aluminium Foil	4.98
NaI:Tl	25 x 25 x 25	MgO	5.01
NaI:Tl	25 x 25 x 25	TiO_2	5.00
NaI:Tl	30 x 30 x 30	Aluminium Foil	4.96
NaI:Tl	30 x 30 x 30	MgO	4.99
NaI:Tl	30 x 30 x 30	TiO_2	4.78

Author Statement

Murat Dağ: Investigation, Resource/Material/Instrument Supply, Original Draft Writing, Visualization.

Acknowledgment

As the authorsof this study, we declare that we do not have any support and thank you statement.

Conflict of Interest

As the author of this study, we declare that we do not have any conflict of interest statement.

Ethics Committee Approval and Informed Consent

As the authors of this study, we declare that we do not have any ethics committee approval and/or informed consent statement.

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