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PHOTOPEAK EFFICIENCY RESPONSE FUNCTION OF AN UNDERWATER GAMMA-RAY NaI(Tl) DETECTOR USING MCNP-X

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ABSTRACT

This work presents a study to calculate the response function of a 1.5"x 1" NaI(Tl) scintillation detector when it is used in the marine environment in the energy range from 20 keV to 662 keV. The method takes into account both the scattering of photons in the water and the detection mechanism of the detector. In addition, the calculation of the response function of the whole system is essential for suppressing the background of the measurement and for estimating the concentration of the involved radionuclides, especially given the greater probability of primary gamma photons undergoing multiple scattering events before they interact with the detector. The experimental photopeak efficiency measurements for point sources were compared with the simulated results under the same conditions of the experimental setup to validate the simulation of the detector. Monte Carlo simulations were performed using the MCNP-X code for the investigation of gamma-ray absorption in water in different brines. The energy resolution curve was used to improve the response of the mathematical simulation of the detector. The detector's simulation was based on information obtained from the gammagraphy technique. Both dimensions and materials were used for the calculation with the MCNP-X code. The photopeak efficiency of a NaI(Tl) detector for different radionuclides in the aquatic environment with different salinities was calculated.

Keywords: MCNP-X code, photopeak efficiency, NaI(Tl) detectors, marine environment.

1. INTRODUCTION

been improved significantly to detect leakages due to nuclear power plants and it is considered to be essential. In the case of a nuclear accident, the monitoring program must provide fundamental information on abnormal liberation of radioactivity, for security measures. In this case, it is indeed crucial to obtain detailed information rapidly to react adequately. It is important that detectors have a fast response time, distinguish the signal from background, reliably characterize gamma-rays from different sources and clearly discriminate between them [1]. Consequently, gamma-ray detector material has to meet the stringent national security

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[1] The in situ measurements offer many advantages over the laboratory analysis with conventional sampling, such as monitoring on a continuous basis, recording of propagation of the pollutant, as well as mapping of large areas of seawater.

requirements. The development of a underwater gamma-ray spectroscopy for the investigation of radioactivity in the water environment is today of important scientific priority for the marine sciences and especially for the operational oceanography [1].

The ^{137}Cs is of special interest among the artificial radionuclides, because it is a long-lived radionuclide (30.05 years); it is used as a radionuclide tracer in seawater and constitutes the artificial radionuclide of greatest radiological significance in the marine environment. The recorded gamma-ray spectra in seawater are characterized by high background induced by the natural radioactive constituents of seawater. This makes the system difficult to precise peak identification gamma-ray contributions, especially for low-level radioactivity studies and in the low-energy region, where the 661 keV gamma-ray of ^{137}Cs, is situated in the spectra is the most important radioactive contaminant, radiological assessment, produced from the atmospheric bomb tests, discharges from nuclear reactor accident [2-7]. Both properties are importants in the low-level gamma-spectrometry of environmental radioactivity research.

The systems most commonly used for gamma-ray spectroscopy in seawater, are based on NaI(Tl) detectors[2] [8-11], because of high[3] efficiency for high-energy gamma rays detection [18], being rugged to thermal and mechanical shock, relatively low cost spectrometric system (detector + associated electronics), availability in wide variety of sizes and is used at room temperature, therefore, can be used in various applications in field under unfavorable weather conditions. However, they have the disadvantage of relatively poor energy resolution and high background mainly originating from the Compton Effect during the detection process of the high-energy gamma-rays from the deexcitation of natural radionuclides, such as ^{40}K and ^{214}Bi, that reduce the capability of the whole system for underwater operational use [12-14]. Despite their lack of energy resolution, which strongly limits their use in gamma spectrometry, inorganic scintillators, and especially NaI(Tl), are the most common crystals for long-term in situ measurements since they combine high efficiency and capability of measuring with low power consumption [14].

This detectors have been used for a number of applications in the marine environment in the last few years, but these systems involve a high level of background radiation, rendering them incapable of detecting low-level radioactivity (<100 Bq.m^{-3}), especially in cases where the radionuclides emit low-energy gamma rays (<600 keV) [1]. Moreover, these detectors show a elevated sensitivity to the salinity of the water at low energy gamma-ray, mainly due to the high atomic number of chlorine atoms, which modify the photoelectric absorption region [15-16]. One approach to solve this problem is to measure regularly the salinity of the water component, but this solution will not pick up sudden changes in salinity.

In order to improve the use of the NaI spectrometer in seawater, Monte Carlo (MC) simulations has been developed for the calculation of its response function induced by salinity of seawater in the NaI(Tl) detector as well as spectrum unfolding techniques [17-18]. The Monte Carlo method is a widely used simulation tool for radiation transport, mainly in

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[2] When the gamma radiation interacts with the NaI(Tl) detector it yields scintillation that are transformed into electric signal, using a photomultiplier tube that consists of a photocathode that converts photons in the visible light range produced by radiation interaction within the scintillation crystal into electrons that are properly focused and accelerated by the dynodes with which they collide with enough kinetic energy, resulting in secondary electrons. The electron cascade resulting from this multiplication process produces a current pulse that reach the anode of the tube, which is collected with sufficient intensity to be processed in a gamma ray spectrometry system. The number of electrons converted is proportional to the energy of radiation incident on the crystal and is fairly linear for a meaningful range of energy. This allows relating the amplitude of the signal current with the energy absorbed by the crystal.

[3] Tthe relatively high atomic number of Iodine ($Z = 53$) and, due to the crystal's density, the NaI(Tl) detectors show large absorption efficiency, in other words presents a high photopeak to Compton ratio.

situations where measurements are inconvenient or impracticable. The MCNP-X code is a general purpose Monte Carlo radiation transport code developed at the Los Alamos National Laboratory and designed to track different types of particles (neutrons, electrons, gamma-rays, etc) over a broad range of energies [19]. MCNP-X code[4], based in MC method is a general purpose computer code which is useful for phenomena which are random[5] in nature such as interactions of nuclear particles with materials. The code rather obtains the solution of the problem by simulating individual particle trajectories and recording some aspects of their average behavior (it does not solve the Boltzmann particle transport equation). The MCNP-X code can be used, as in the case of this work, to simulate gamma-rays interactions which comprise: i) incoherent and coherent scattering; ii) the possibility of fluorescent emission after photoelectric absorption; iii) pair production with local emission of annihilation radiation and Bremsstrahlung effect [19]. When performing the mathematical simulation (using the MCNP-X code) of NaI(Tl) detectors, in order to obtain their response curves, some corrections should be made to improve the simulation in order the approach to the real case. Two of the main corrections are essential: the determination of the photon detection efficiency to quantify the radiation field and; the energy resolution, $\Delta E/E$, which is related to distinguish between different peaks very close to each other in the energy spectrum, their determination is of great importance when performing the identification of radionuclides. The simulating the detecting mechanisms of NaI(Tl) detectors, comprises the calculate the energy distribution of photons in seawater since primary gamma photons, which undergo multiple scattering that interact with the detector, contribute significantly to the measured spectrum.

The pulse height distribution (PHD), which is the output spectrum that reflects the interaction that occurs in the sensitive volume of the detector, does not reflect exactly the true photon flux due to the variation of the detector's energy response and other physical phenomena, as the occurrence of K-shell X-ray escapes and Compton scattering. Although the PHDs can be measured experimentally with the use of several calibrated monoenergetic radiation sources with energies of emission covering the entire range of interest, the number of these sources can be limited[6] and time consuming. Therefore, to calculate the response at intermediate energies between those obtained by isotopes measurements, either complicated interpolation methods of the experimental spectra or simulation by using Monte Carlo technique should be used [2; 21-32]. The Monte Carlo methods make it possible to calculate the response function for detectors with good accurate results [26], since precise and sufficient data to describe the various parts constituents of the detector are provided, in good agreement with those obtained experimentally. The photon detection efficiency curve, when calculated by simulation, is not influenced by several parameters, such as uncertainty in the concentration of activity and gamma energy yield, decay correction or peak sum effect [27].

This work uses the Monte Carlo N-Particle eXtended (MCNP-X™) code [19] to simulate the NaI(Tl) scintillation detector and the methodology presented improves[7] the determination of

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[4] The process consists of following each of many particles since its emission from a source until it reaches an energy threshold; the particle energy is transferred to the medium by absorption, escape, physical cut-off, etc. Probability distributions are randomly sampled using transport data to determine the outcome at each step of its trajectory. The quantities of interest are tallied, along with estimates of the statistical precision of the results.

[5] The individual probabilistic events that comprise a process of interaction of nuclear particle with material are simulated sequentially. The probability distributions governing these events are statistically sampled to describe the total phenomenon and the sampling process is based on the selection of pseudo-random numbers.

[6] It is also necessary radiation sources with high enough intensity to get a good counting statistics for the desired source-detector distance. Equipment and facilities are also required to ensure accurate positioning of the source, since small variations in distance or source-detector alignment may influence the response of the detector, thus increasing the uncertainty of the measurements.

[7] Even though the computer code does not simulate the scintillation process, but scores the energy deposited in any material, and considering that the photomultiplier tube accounts satisfactorily for the photon interaction within the detector's sensitive volume, the results obtained by

the response function of this type of detector for use on underwater applications. Thus, this work presents a study to calculate the response function of a 1.5"x 1" NaI(Tl) scintillation detector when it is used in the marine environment in the energy range from 20 keV to 662 keV.

2. METHODOLOGY

The methodology consists in mathematical modeling of the NaI(Tl) detector and simulation by the Monte Carlo method. The relationship between resolution and energy was determined experimentally for the correction of the spectra obtained by simulation. The parameter used for experimental validation of the detector's model was the comparison of the experimental efficiency curve with the calculated one by the MCNP- X code. The activity of the point radiation sources were measured with the NaI(Tl) detector to well defined distance between source-detector, in the axial direction of the detector and, afterwards, the same geometry was reproduced in the MCNP-X code. The spectrometric system response on energy resolution and efficiency curves was determined by measuring the maximum number of secondary standard sources (^{241}Am, ^{60}Co, ^{152}Eu, ^{22}Na e ^{137}Cs) available which were supplied and certified either by the Institute of Radioprotection and Dosimetry (IRD) or the International Atomic Energy Agency (IAEA) [33].

2.1. Mathematical Detector Model

The detector's simulation was based on information obtained from the gammagraphy technique. Both dimensions and materials were used for the calculation with the MCNP-X code [34]. The NaI(Tl) crystal density used was 3.667 g.cm^{-3}, the MgO powder density used was 2.0 g.cm^{-3} [30] and the aluminum density was 2.7 g.cm^{-3}. The photomultiplier tube on the back of the crystal was treated as a 30 mm thick aluminum disk to account for backscattering [30].

The mathematical model considered NaI(Tl) scintillator detector as a homogeneous cylinder [2; 24; 30-34] with 31 mm (diameter) and 19 mm (thickness). The information (dimensions and materials) of a real NaI(Tl) scintillator detector was considered in the model for calculating the MCNP-X code from gammagraphy technique. A special treatment provided in the MCNP-X code: the Gaussian energy broadening (GEB) (card FTn) option has been used to fit the full energy peak shape of PHD [19]. The GEB parameters have been set taking into account the resolution of the detector by the FWHM provided by radioactive sources [2; 33-34], for this it must to be inserted into the input file (INP), the mathematical model of the detector. This step aims to validate the response of the detector quality by means of energy resolution while the order quantity will be achieved by normalize the of the full energy peak from PHD obtained by MCNP-X code. In calculations, it has been considered the radiation background and the contributions due to interactions by Compton Effect.

a) Experimental validation

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the simulation is quite representative of the detector's response, otherwise no match between simulation and experimental would be reached. Nevertheless, a wide variation in the total number of electron at the output of the photomultiplier tube is expected, which is the cause of the high detection resolution. Therefore, the spread in energy response must be considered in the simulations and is accounted for by experimental measurements.

To simulate the response functions for NaI(Tl) detector by the MCNP-X code it should be modeled with the best possible accuracy because variations of the detector's crystal and surrounding materials dimension influence the photon detection efficiency [34]. The detector's physical data for the mathematical model was obtained by gammagraphy technique and the "crystal effective" dimension was obtained by the relation between simulation and measurement of two point sources at several positions around the detector.

Two sources were used to determine the effective dimensions of the detector's crystal, ^{241}Am and ^{137}Cs, low and high energy emitter respectively. The sources were measured with the NaI(Tl) detector under well-defined distances, positioned toward the longitudinal axis and laterally around the detector's crystal. Four measurements were performed with the two sources on the longitudinal axis (source-detector distance: 5.45 cm) of the detector and the other with the sources positioned laterally (source-detector distance: 4.78 cm), to determine experimentally detector's response for a known radiation field. This geometry was reproduced by simulation and the results were compared with the experimental values. The procedure to define the effective volume of crystal was published in previous works Salgado et al. 2008, 2012 [33-34].

b) Resolution energetic curve

In the experimental spectra, the data has a Gaussian distribution shape for the energy lines. However, the MCNPX code does not simulate physical effects leading to the broadening of the spectrum, but it uses a fitting technique to take into account the resolution of the real detector, measured experimentally, and provided in the input file of this code. Thus, for more realistic results obtained by simulation, it is necessary to consider the spectrum resolution by applying a Gaussian function. The technique consists of using a ‘‘FT8 GEB’’ card and calculating the full width at half maximum (FWHM) of the peak. A set of measurements were performed in order to determine the energy resolution curve of the NaI(Tl) scintillator detector (crystal + housing + photomultiplier tube material equivalent) used for this work, and the curve's coefficients were introduced in the function provided by the MCNP-X code that fits a Gaussian to the spectrum to make the proper corrections [33-34].

c) Efficiency curve

The experimental absolute efficiency measurements for point sources were compared with the simulated results under the same conditions of the experimental setup to validate the simulation of the NaI(Tl) detector [36-37]. In order to avoid the sum effect, improve the counting statistics and consider the source as point, these sources were measured at a well-defined position source-detector distance of 5.45 cm, located on the longitudinal axis of the detector's crystal [33-34]. The PHD estimate (F8 tally), available in the code MCNP-X, was used to obtain the deposited energy distribution per incident photon on the considered detector volume. This tally accumulates, for each individual starting history, the kinetic energy lost by local photon-induced secondary electrons in their multi-step Coulomb interactions with the surrounding atoms.

2.2. Salinity

Measurements using energy gamma-ray attenuation show elevated sensitivity to the salinity of water component, mainly at low energy due to high atomic number of chlorine atoms, which modify the photoelectric absorption [15-16]. The cross section for photoelectric effect at a given photon energy is considerably dependent on the atomic number of the absorbing material, and hence on the salinity of water component. In a multiphase flow with brine and gas, the salt atoms will give a relatively large contribution to the average atomic value of the mixture since the main components of the flow are low atomic number atoms (hydrogen, oxygen, and carbon). Air was considered as gaseous phase with 1.203×10^{-3} g.cm^{-3} density and water phase was assumed as pure water (molecular formula H_2O) with density of the 1.203×10^{-3} g.cm^{-3}. These values were obtained by MCNP-X code using brine consisting of NaCl mixed by weight (w/w) with water.

2.3. Proposed Geometry of the Underwater Detection System

The detection geometry which consists in one NaI(Tl) scintillator detector, positioned at 180º diametrically opposed to source. The proposed geometry uses the transmitted from detector beam measurement from gamma-ray source with a isotropic beam positioned at 50 cm of detector. The NaI detector has been posisioneted in a water tank of 1.0 m^3 volume, with different to investigate to the high attenuation of the gamma-rays in the water. The gamma-rays used to perform the energy calibration of the system were isotropic gamma-ray point sources (40 to 800 keV) simulated in the MCNP-X code. This energy range was choice because the acquired spectra in the water tank the 661 keV of ^{137}Cs peak is the most important in this work.

The measurements reveal different response of the interaction mechanisms of the radiation and the flowing medium. The model developed in the MCNP-X code considers the main effects of interaction of radiation with matter involved and the PHDs from the NaI(Tl) scintillator detectors. The energy resolution, dimensions and characteristics of a real detector are also considered; in general, the model presented tends to approach the realistic case [33-34].

3. DISCUSSIONS AND RESULTS

3.1. Simulation of NaI(Tl) detector

The detector's simulation was validated both qualitatively by the energy resolution curve and quantitatively by the photon detection efficiency. The gammagraphy technique showed to be an important tool to estimate, with some precision, the detector's dimensions used in this work. The aluminum disk at the base of the simulated detector takes into account the effect of all the materials of the photomultiplier tube located under the crystal. The volume sensitive dimensions of the NaI(Tl) detector that showed better agreement was 30.10 ± 1 mm height and 17.20 ± 1 mm diameter. For the process of determining the volume of crystal, the difference between the measured and calculated values for point sources of ^{241}Am and ^{137}Cs were kept as small as possible with an estimated uncertainty smaller than 5%. Fig. 1 shows the schematic representation of the detector used for the simulation [33].

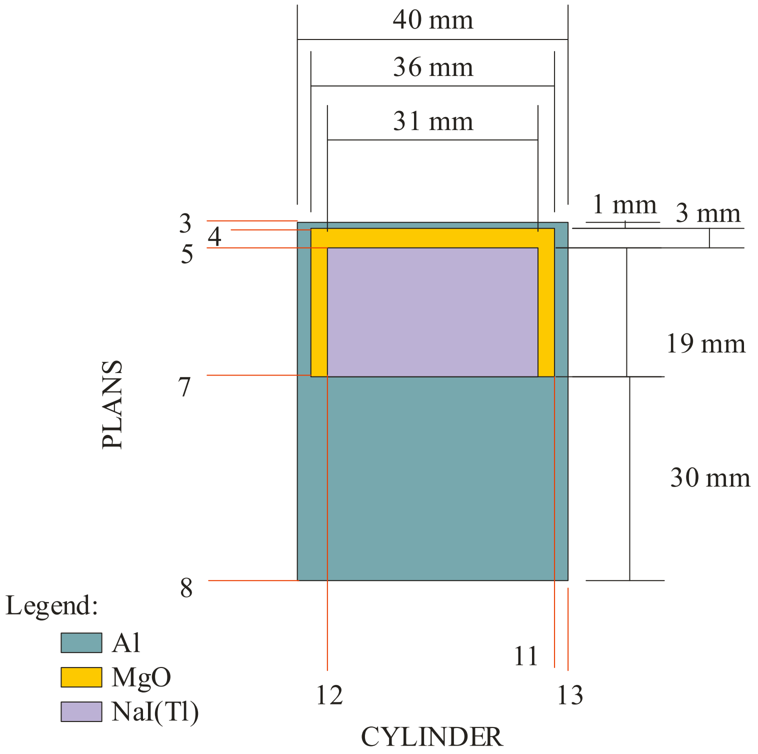
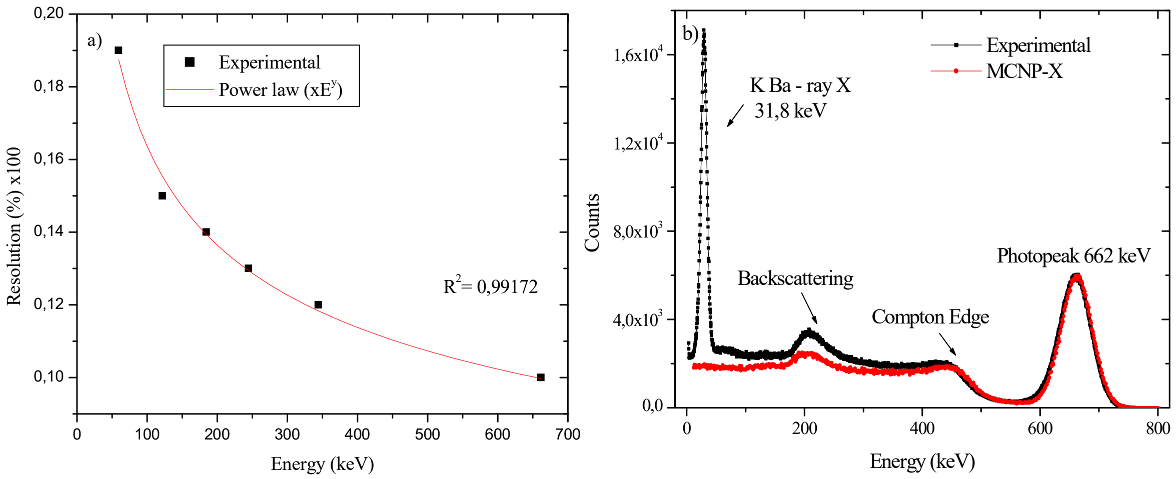


Figure 1: Schematic representation of the NaI(Tl) detector considered in simulation.

Fig. 2a shows the energy resolution curve expressed by a Power law relation (xE^y) [24], the coefficient were calculated by least-squares fitting. Fig. 2b shows the comparison between the experimental and simulated data by MCNP-X code. Fig. 2c shows the experimental absolute photopeak efficiency and the simulation curves as a function of the energy. Energies other than the measured ones were simulated in order to obtain a curve fitted in the energy range from 59.45 to 662 keV, for the same experimental geometry. Fig. 2d shows the linear attenuation coefficients of brine at different salinities [33].



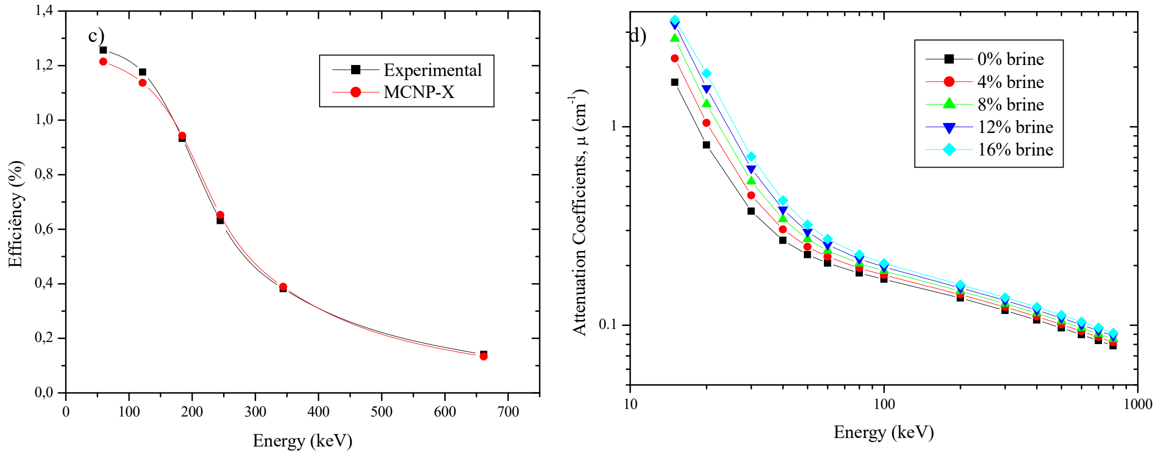


Figure 2: Parameters of NaI(Tl): a) Response curves of energetic resolution; b) Comparison between experimental and simulated pulse height distributions to sources ^{137}Cs; c) NaI(Tl) detector's efficiency curves obtained both by simulation and experimental for radioactive sources; d) The linear attenuation coefficients of brine at different salinities.

It is important to emphasize that the pulse height distribution shown in Fig. 2b represents the radiation sources spectra with background subtraction and, for the simulated spectra, peak broadening treatment was considered. It can be noticed an acceptable agreement on the photopeak, for ^{137}Cs source but the Compton continuum below 400 keV, all the calculated results are a little lower than the experimental data due to the photons scattered on the detector's shielding, support and surrounding materials [32]. The technique of the "shadow shield" can minimize these differences [37-40]. For ^{241}Am, in comparison with the experimental data, a small discrepancy can be seen because the effect of the scintillation efficiency increase in low-energy region where the scintillation efficiency is non-linear and is not considered in MCNP-X code.

All the calculated pulse height distributions were normalized to the experimental data by the counting number in the maximum height of the photopeak and also that the X-ray K-shell of ^{137m}Ba$_{56}$ (BaKα_1) [40] was not simulated and, for this reason, does not appear in pulse height distribution obtained by the MCNP-X code.

The photopeak efficiency values obtained by Monte Carlo calculation were compared with experimental data aiming to validate the detector's simulation. The estimated uncertainties, given the fluctuating counts, the source activity and the emission probability remained below 5% (for a confidence interval of 95.45% k = 2). The experimental and simulated data showed good agreement. The largest discrepancy of 5.54% was found to be for the energy of 59.45 keV from ^{241}Am. The adjustment coefficients of the efficiency curve for the Na(Tl) detector were obtained by the method of least squares and the correlation coefficient (r^2) was 0.9987 [33].

The attenuation coefficients of water with different salinities present major diffences in low energy range as show in Fig. 2d. It is possible to see a clear difference between the lower and the higher salinity more intensely in the low-energy range [38].

This fact is more evident in the PHDs calculated water mixed with salt in four different values of the salinity index (0%, 4%, 8% and 16%) as shown in Fig. 3.

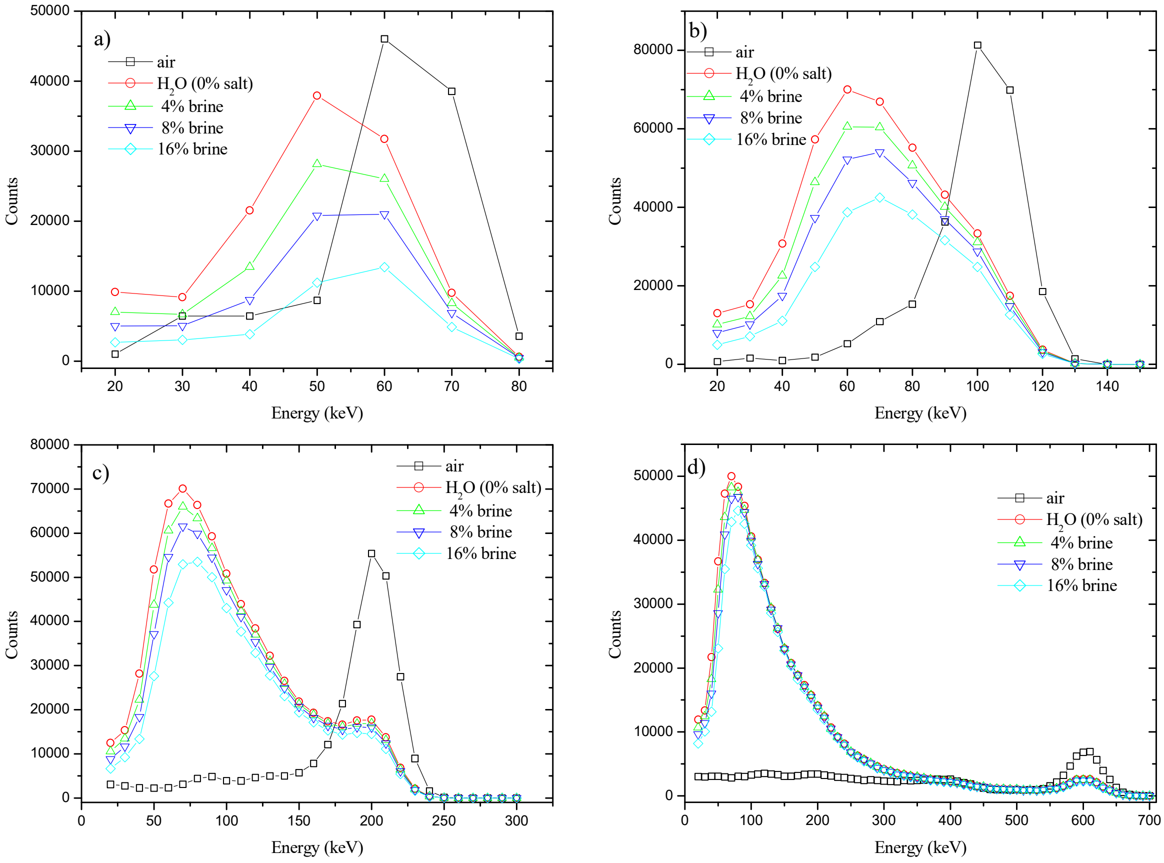


Figure 3: PHDs obtained by detector at different salinities and materials for energy of: a) 60 keV; b) 100 keV; c) 200 keV; d) 600keV.

It is possible to see a clear difference between the lower and the higher salinity. It is possible due the attenuation coefficients for water with different salinity, are shown in the Fig. 2d.

The values of the photopeak efficiencies for 200, 400, 600 and 800 keV for the gas and water with different salinities are shown in Table 1.

Table 1: Photopic efficency values.

Energy (keV)	Photopic Efficiency				
	Gas	Pure Water	Brine 4%	Brine 8%	Brine 16%
200	8.90E-04	2.29E-04	2.17E-04	2.03E-04	1.83E-04
400	3.69E-04	1.15E-04	1.09E-04	1.04E-04	9.52E-05
600	2.09E-04	7.58E-05	7.28E-05	6.98E-05	6.48E-05
800	1.45E-04	5.88E-05	5.67E-05	5.46E-05	5.09E-05

The relative errors are shown in Table 2. It can be noted that the photopic efficiency at 600 keV is decreased by 63.78% when it changes the material around the source and detector, gas to pure water (0% salinity), for a layer of material 50 cm. At 600 keV the photopic efficiency is decresaed by 14.48% when it changes the pure water (0% salt) to brine 16%.

Table 2: Relative Error.

Energy	Relative Error (%)		
(keV)	gas/water	gas/brine 16%	water/brine 16%
200	74.28	79.50	20.27
400	68.95	74.22	16.99
600	63.78	69.03	14.48
800	59.34	64.81	13.45

This detector shows a non-linear drift in the produced spectra, changing the exact position of the photopeaks and altering the full-width at half-maximum. This may probably be caused by the scattering of low energy gamma- rays by water, decreased the number of photoelectric events and Compton scattering increases. The energy spread of the photons in seawater, the energy drift and the poor energy resolution of the NaI detector can reduce the capability of the this detector for underwater use.

4. CONCLUSIONS

A procedure for simulation of a NaI(Tl) detector has been presented in this work with the MCNP-X code. The photon detection efficiency curve was used for the validation of the mathematical model developed in the code. The efficiency curve was obtained both experimentally and by simulation; the results were compared and showed a good agreement. The methodology using two radioactive sources, a low and one high energy photon, was found to be satisfactory to fit the dimensions of the crystal to be used in the detector's simulation.

For applications in marine waters, the study shows that to identify radionuclide is essential to know the characterisics of water to calculate of the exact position of two photopeaks and perform a energy calibration. It can be noted that the photopic efficiency at 600 keV is decreased by 14.48% when it changes the pure water (0% salt) to brine 16% for a layer of material 50 cm. It is essential to consider the effect of water and the amount of salt present, as these factors strongly influence the determination of counting efficiency and obviously to calculate the activity.

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