2013 International Nuclear Atlantic Conference - INAC 2013
Recife, PE, Brazil, November 24-29, 2013
ASSOCIAÇÃO BRASILEIRA DE ENERGIA NUCLEAR - ABEN
ISBN: **978-85-99141-05-2**

DESIGNING A WATER CALORIMETER AS PRIMARY STANDARD OF GAMMA RAYS AT IPEN/CNEN

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ABSTRACT

This work aims to describe the present stage and the next steps of the development of a water calorimeter of the Calibration Laboratory of IPEN/CNEN. This calorimeter will be used as a primary standard of gamma ray sources at the laboratory. Between the design and the construction step it will be shown how this model was chosen and how it is modeled virtually with computer simulation. The two main codes used, MCNP and Fluent, to characterize the prototype before its construction are presented.

1. INTRODUCTION

There is consensus that any country that values the quality of the uses of ionizing radiation needs metrological laboratories that keep the standards updated, execute the calibrations and guarantee the devices that employ ionizing radiation. The ideal is that each country invests in its own metrological laboratories. The primary standard dosimetry laboratories are the American NIST, the French BIPM, the English NPL and the German PTB. In Brazil, the Brazilian Commission of Nuclear Energy (CNEN) and their laboratories maintain the standards and keep the national uses of nuclear radiation updated.

The Calibration Laboratory at IPEN/CNEN provides calibration services and characterizes equipment as ionization chambers, dosimeters and radiation beams. To complete the laboratory set-ups, a calorimetric based device was designed and will be constructed as an improvement, with all its advantages.

Calorimetric devices are far from new, but their versatility and simplicity are highly valued specifically for nuclear technology [1, 2, 3, 4]. These devices have some disadvantages, for example the relative poor sensibility for low dose rates, constraining the calorimeter as a dosimeter for only high dose rates and therefore usually for characterization of gamma rays from sealed sources or industrial irradiators [5].

Water is a medium with high thermal capacity, so this choice for a calorimeter nucleus contributes to the general low sensibility, added the fact that this material stays in a liquid phase at room temperature with all convective currents, therefore it is not trivial to employ a water calorimeter as a dosimeter device (to solve the convection problem, an ice calorimeter was already tried [6]).

Differently from an ionization chamber that relies in air kerma and a well-known source, the water calorimeter may ignore any source information to measure the absorbed dose. The idea is simple; the radiation energy is deposited in the medium in heat form. There are no conversion factors as air-water kerma or something like that. Thereby the measurement is direct and simple despite some heat defect or noise readings in temperature [1, 7].

The designing of a water calorimeter for gamma rays, as any other dosimeter, needs a careful planning to avoid wasting resources in a so relevant radiation detector. Two solid and known computational softwares were utilized to optimize the assembling of the device.

2. MATERIALS AND METHODOLOGY

This work is divided in four parts; first, it will be presented how the best project was chosen. Secondly, the modeling of the thermodynamic behavior of the calorimeter as a whole will be presented, especially from the refrigerator fluid fluxes around the nucleus. The question is how and when this flux can drop the temperature until the expected degree and maintain this state with the proposed geometry and with a commercial water chiller (PolyScience model EW-12930-32). Thirdly, the general radiation transport code MCNP5 was utilized to simulate a typical irradiation of the calorimeter in a collimated ^{60}Co beam and the energy distribution profile in the water nucleus; finally the temperature variation from ^{60}Co sources with several activities was simulated. The operational range of the calorimeter was therefore estimated with the available technology and resources.

2.1. Choosing the model

A calorimeter for dosimetry purposes can essentially be quasi-adiabatic (or adiabatic), isothermal, or differential [8]. The quasi-adiabatic equipment presupposes an isolated nucleus of some composition; this nucleus is sealed and cannot exchange either matter or energy with the external medium, so the temperature change is related to the incident radiant energy. The isothermal one measures the heat flux that moves in or out from the nucleus so that the temperature remains unchanged. The differential equipment is similar to the heat flux equipment, but in this case the heat flux that transits from one nucleus to another is measured.

Based on other calorimeter results around the world, the best procedure is to measure the temperature with good precision. As it is cheap and simple to isolate a water bulk from typical room temperatures, a quasi-adiabatic calorimeter was chosen for this work.

The model uses a cylindrical symmetry. Basically there is a water nucleus with 30cm x 30cm of height and diameter. The nucleus is sealed with glass and filled with ultra-pure water (UPW) with conductivity above 5.4×10^{-6}S/m or 18.2MΩ.cm; this inorganic degree of purity is also referred as Type I water. Around this vessel there is an active heat barrier that is composed by circulating water and an antifreeze additive that guarantees that the nucleus is at right temperature and is ready for irradiation. This second wall is made of acrylic, and is surrounded by a passive vacuum barrier and is sealed by another acrylic wall. All layers are grounded and with a minimum contact with the outer layer. To avoid the known heat defect that introduces noise in the temperature measurements, it was decided to purge the oxygen and saturate the nucleus medium with N_2. Both glass and acrylic walls were designed to have 0.5cm of width. When completed in a prototype stage, probably it will have an additional structure of wood, aluminum and styrofoam. The nucleus operates at a reference temperature of 277K so the water has its maximum density, and therefore the convective forces that shift the temperature measurements at one specific point of interest are minimized.

Figure 1 shows the scheme of all subsystems that compose the primary standard calorimeter. This work focuses only the calorimeter design.

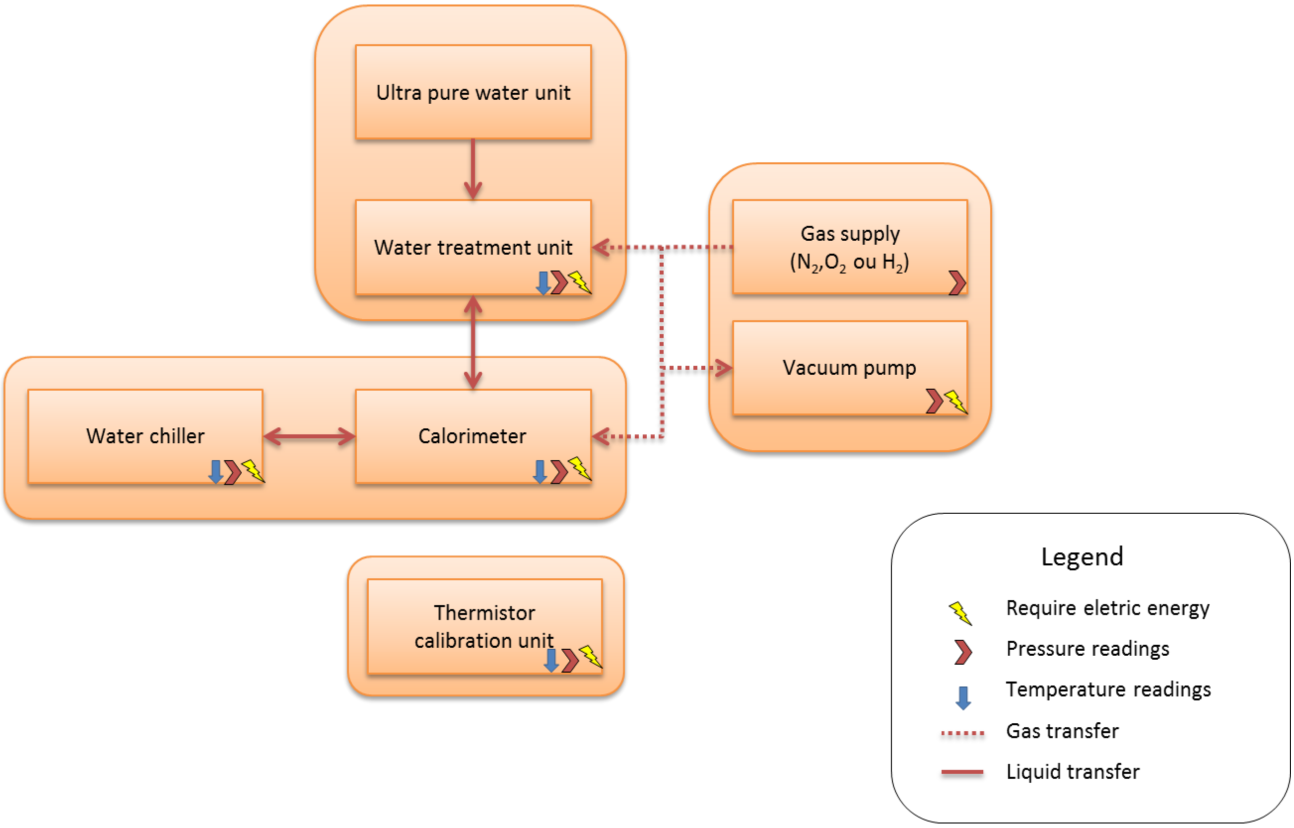

Figure 1: Scheme of all systems used in a calorimetric primary standard for gamma rays.

Figure 2 shows the calorimeter as designed in Ansys Design Modeler. This software view was chosen because the CAD representation is very rich in details.

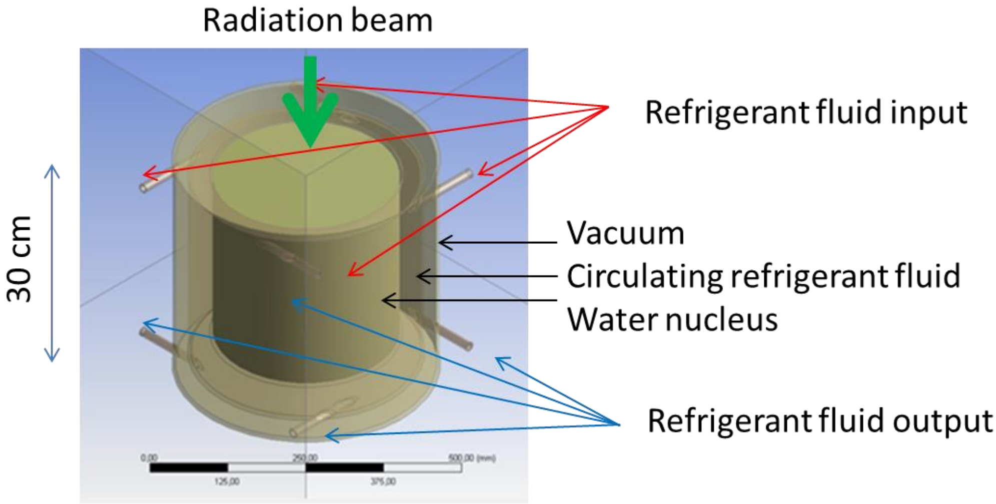

Figure 2: Water calorimeter designed in Ansys Design Modeler. The acrylic and glass parts are translucent.

Note that there are four entrances for the refrigerator fluid (upper conduits) and four outputs for the refrigerant fluid (bottom conduits) All ducts have 0.5cm of radius and flow of 0.77m/s at 263K (initial temperature). These parameters were chosen based on a commercial water chiller, so these data are based in real parameters.

All simulations (with Fluent and MCNP5 codes) used an Intel i7 2600K at 3.4GHz with a RAM memory of 16GB with running Windows 7.

2.2. The Ansys Fluent simulations

Fluent is an Ansys solution to simulate the fluid flow and its thermal changes in any geometry [9]. As the other fluid dynamic code, CFX, this software is very powerful and requires a great dose of knowledge to be used. It is able to simulate heat transfer, turbulence and even chemical reactions as combustion. A very simple model was assumed where the entire assemble used 4.5 x 10^6 elements divided into tetrahedrons of 0.5cm and no inflation technique was used.

A transient type simulation was performed with a total time of 60min and timesteps of 30s. The simulation was restricted to a maximum of 10 loops and a RMS of 10^{-4} in the convergence criteria. The whole set was at an initial temperature of 300K and the refrigerant fluid at a temperature of 266K with 0.77m/s in all 4 upper entrances. Buoyancy forces were present, a $(k-\varepsilon)$ turbulence model (intensity of 5%) was used, and the entire reference frame was stationary. The pressure was 10^5Pa (1atm) and the radiation heat transfer was disregarded. The output tallied the smallest size file every timestep.

2.3. The MCNP5 simulations

The **M**onte **C**arlo for **N-P**article code is a transport radiation code used to simulate the origin, transfer, interaction and death of a radiation particle; it is a versatile software with already about 60 years in use at the scientific community. As the name suggests, it is based in a stochastic approach to simulate radiation transport through a medium with a random number generator and sampling preset probability density functions [10].

In this simulation the objectives were to answer two questions: First, to acquire the energy deposited at some positions inside the water nucleus and to estimate the radioactivity range of a typical gamma radiation source (as for example a ^{60}Co source) to induce a temperature change meaningful enough to be reasonable detectable in a water medium with these proportions; second, if saturated the nucleus with a gas (N_2 for example in this case) it can change the energy deposition profile compared to a simple water nucleus.

Thus the same geometry presented in Figure 1 was modeled in MCNP5, and the same materials, densities and dimensions were utilized in both codes. The difference is the presence of an array of artificial surfaces inside the nucleus to estimate the dose in some chosen positions. Figure 3 shows the calorimeter design in MCNP5 and the tallied cells. These cells have 0.25cm in diameter spaced 1.5cm, each one disposed in a matrix of (11 x 10) units covering almost all the sensitivity volume of interest. The composition and density of all spheres are exactly the same of the external medium of the nucleus. The cards “mode p e”, the tally “*f8” to sample the energy deposition in each cell, the “dbcn 17j 1” to select the ITS indexing algorithm were utilized, and all the cell “importances” were settled to unit. The source was assumed to be a square (14.5cm x 14.5cm) plane beam positioned at a distance of 117cm from the water nucleus surface to the normal vector. Both source and calorimeter irradiation side were aligned. This source emits with a maximum cosine direction of 0.999 between the symmetry axis and the emission direction. The energy spectrum is only composed by photons of a ^{60}Co source (it was specified “par 2” in the “sdef” card), see Table 1 for more details. 6.6 x 10^6 histories were generated. The 10 statistical tests were satisfactory and a maximum uncertainty of 10% at farther spheres was achieved.

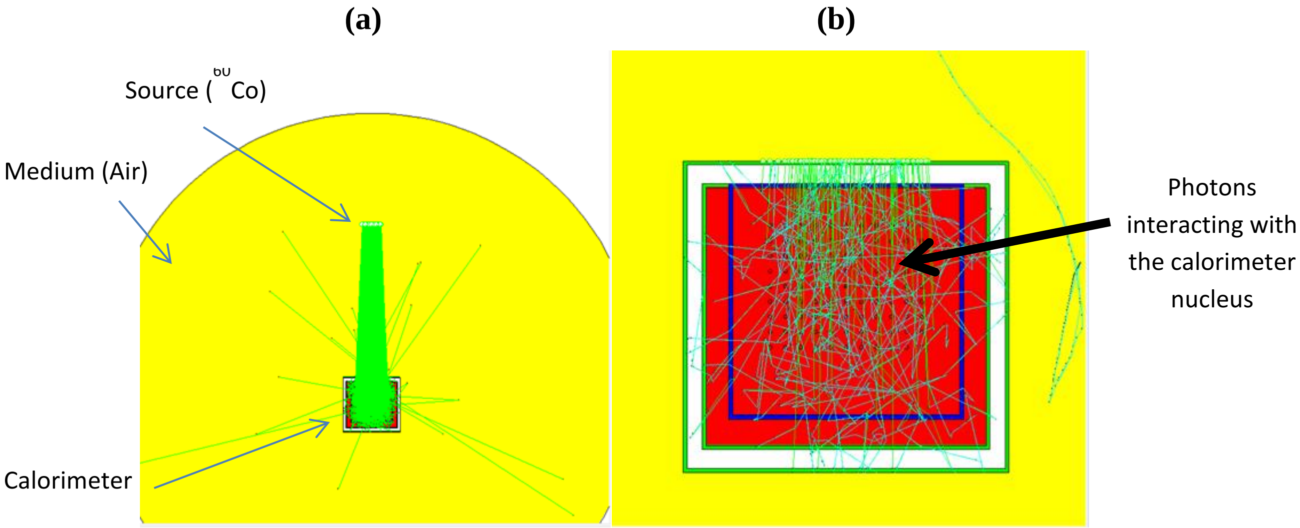

Figure 3: Water calorimeter modeled in MCNP5 (obtained with Vised X_22). (a) View of source and photon tracks (in green) and (b) closer view of calorimeter with inner structures and the spheres tallied (11 x 10 matrix).

Table 1: Gamma spectrum of a ^{60}Co source [12].

Energy (MeV)	Intensity
0.347	0.008
0.826	0.008
1.173	99.974
1.333	99.986
2.159	0.001
2.505	2×10^{-6}

2.4. Estimating the temperature change

The relation between the deposited energy by the beam radiation and the temperature increase in the water medium is a hard issue, because the heat defect inserted to the radiochemical reactions in water is mainly due to the presence of oxygen. So a widely solution is to purge the water with oxygen and to saturate it with another gas that introduces a stable and predictable heat defect. Viable options are the N_2 and the H_2 gases; in this work the presence only of nitrogen was considered.

The heat defect h is defined as

$$h = \frac{E_{dep} - Q}{E_{dep}} \tag{1}$$

where E_{dep} is the deposited energy portion in matter and Q is the energy that actually contributes to the temperature raise. Several studies aimed the evaluation of the heat defect in water. If negative, the reaction is exothermic, and if positive, the heat defect is endothermic. In this study a heat defect of -3% was assumed based on work of Ross and Klassen [2] and Seuntjens and Palmans [11]. The change in the temperature distribution is given by the fundamental equation of calorimetry, adapted to count the heat defect in water:

$$\Delta T = \frac{E_{dep}(1-h)}{mc} \tag{2}$$

where ΔT is the temperature change (the reference temperature is $T_0 = 277K$), m is the mass of the water cell, and c is the specific heat in cal/g^{o}C (for water this parameter is one). Using spheres of 0.25cm in diameter the mass is 0.06545g. Using equation (2) the tallies given by MCNP (the $E_{dep}\, per\, particle$) could be adapted to estimate the effective temperature rise, if there is any information about the source radioactivity and the exposure time. An uninterrupted irradiation time of 45 min was considered.

3. RESULTS AND DISCUSSION

Figure (4) shows the results of the Fluent software, specially the radial thermal profile at the beginning and at the end of the simulation time. It can be seen that after approximately 30min the system reaches its operation temperature of 277K. Inside the water nucleus the temperature is homogenous and in principle the temperature readings can already be taken from a thermistor. Figure (5) shows similar information: an axial profile of the temperature after the same period of time (30min).

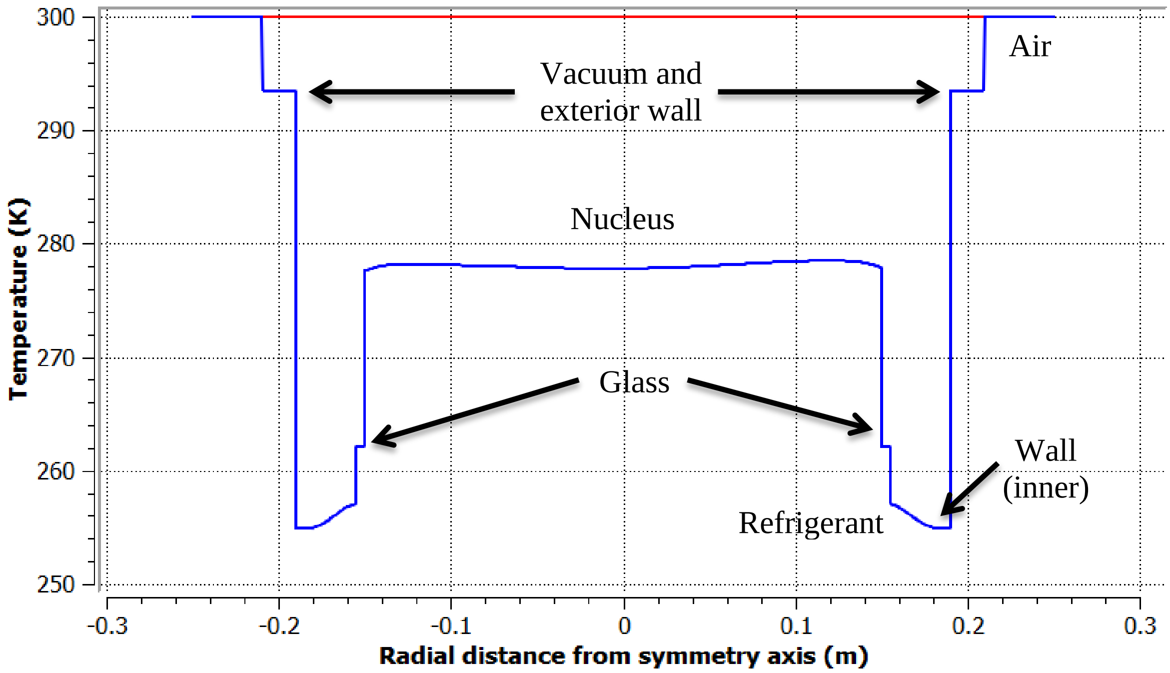

Figure 4: Radial temperature profile in the core, at half of the height of the calorimeter at time t=0s (red line) and at time t=1830s (blue line).

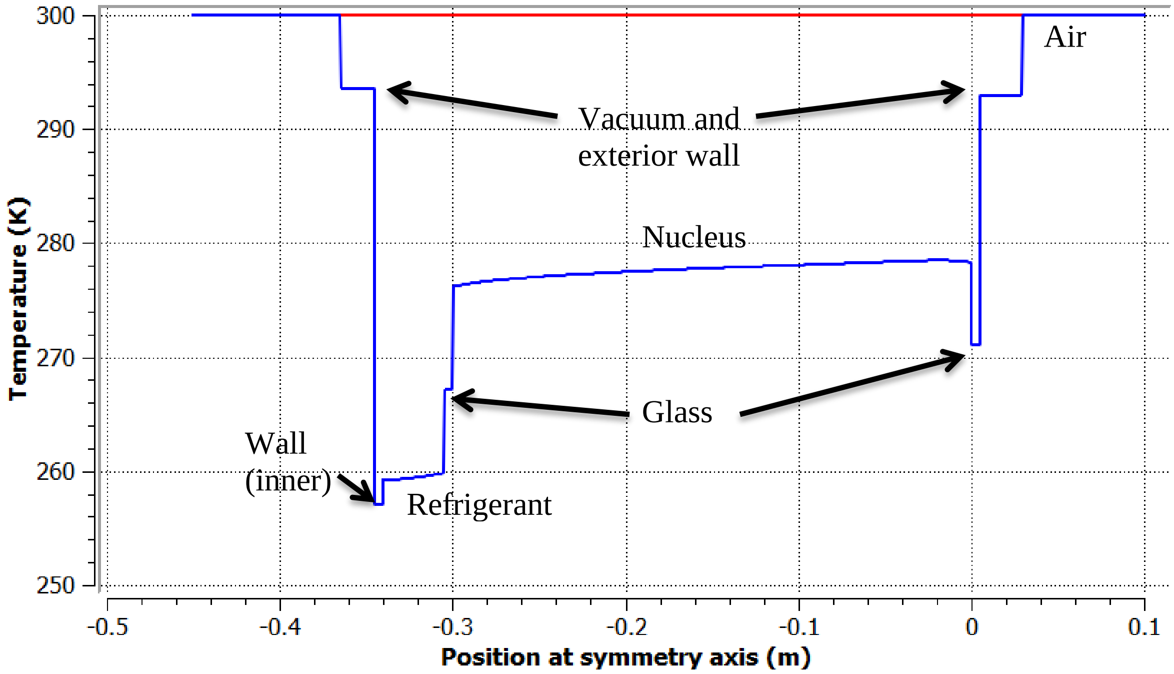

Figure 5: Axial temperature profile in the symmetry axis of the calorimeter at time t=0s (red line) and at time t=1830s (blue line).

The water nucleus is positioned between -0.15 and +0.15m in Figure (4), exactly the core radius. It can be seen that inside the calorimeter the temperature distribution is homogenous and stable at 277K. Outside the nucleus are the structural components and the refrigerant; their temperature profiles are presented too. So, in order of appearance, starting from the symmetry axis are the water core, glass, refrigerant, polyethylene inner wall, vacuum, polyethylene exterior wall and outside air (room).

Figure (5) highlights the need for cooling in two stages: first with the water chiller at full power (operating at 263K), second, after the core reaches an average temperature of 277K it starts a temperature maintenance cycle, with the chiller operating at 277K for 30min. This ensures the best temperature stability especially at higher depths, dismissing additional correction factors in temperature measurements. Another idea is insert a stirrer to help homogenize the water nucleus; this can streamline the cooling process and avoid temperature gradients.

The simulation results obtained with the MCNP5 code are presented in Figures (6) and (7) where the deposited energy is shown according to the depth and distance related to the symmetry axis.

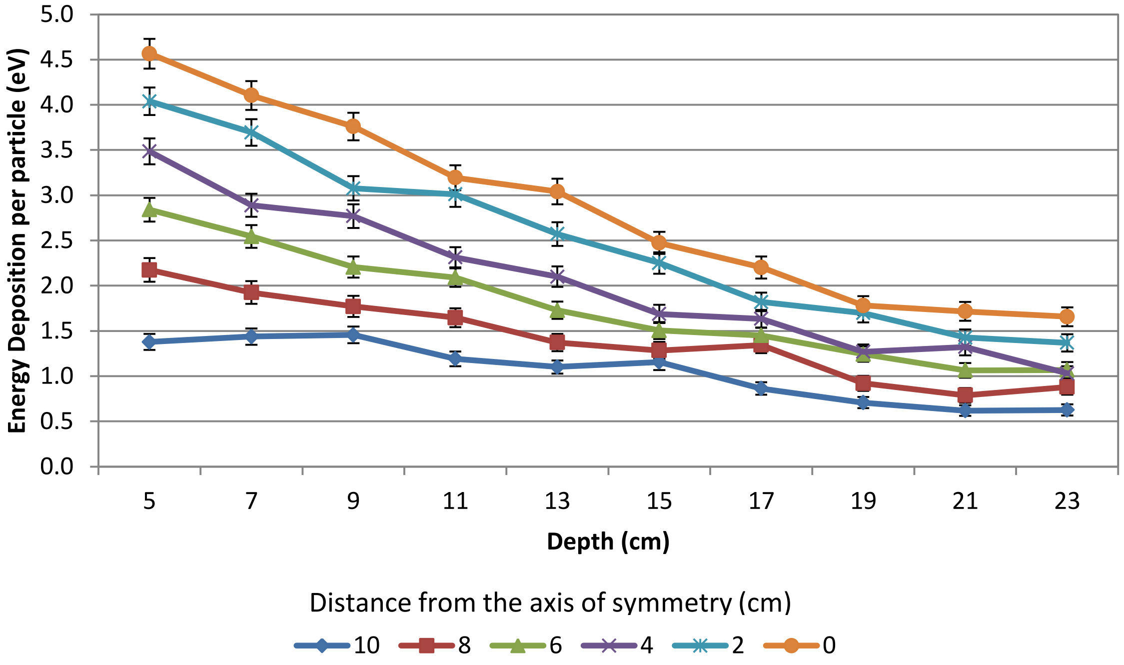

Figure 6: Energy deposited per simulated particle relative to the depth in the water bulk for several distances from the symmetry axis of irradiation.

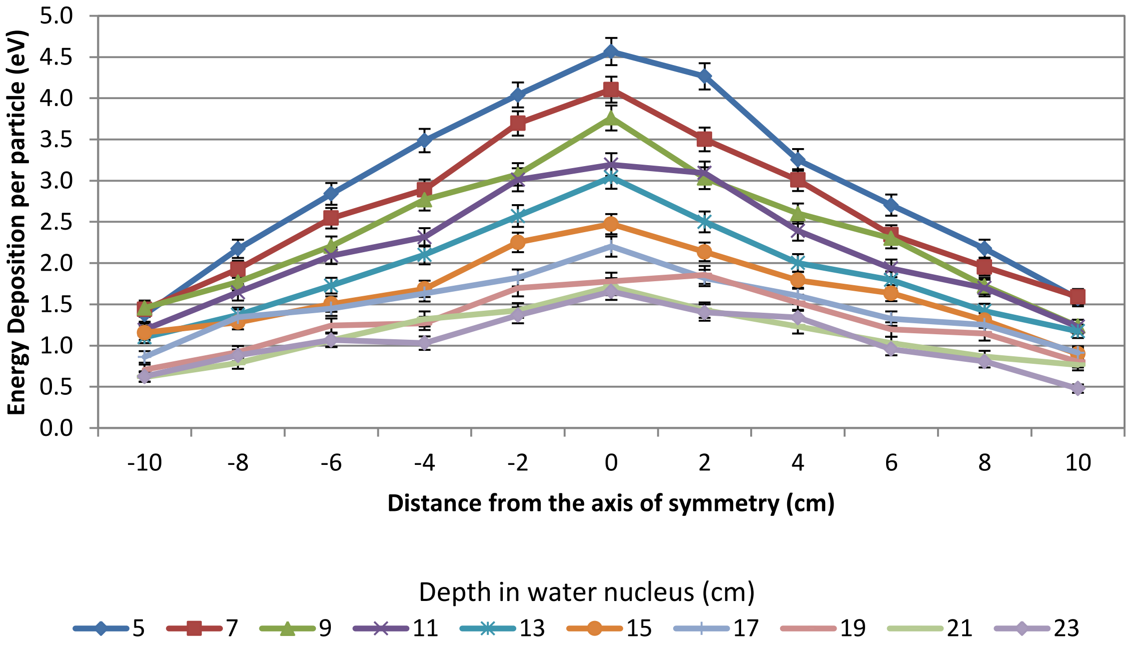

Figure 7: Energy deposited per simulated particle relative to the axial distance for several depths in the water nucleus (reference for depth distance: surface faced directly to the beam).

It is important to remember that the uncertainty may be dropped to lower levels, but requiring even more processing time. These data can feed the Equation (2) and result in a temperature variation instead of energy deposition data. A heat defect of -3% was chosen, so the chemical reaction in water is exothermic. This fact implies a N_2 saturated nucleus with less oxygen as possible. Figure (8) presents the temperature profiles calculated for two source radioactivities, 18.5GBq (500Ci) and 7.4GBq (200Ci). The irradiation time was 45min in both cases.

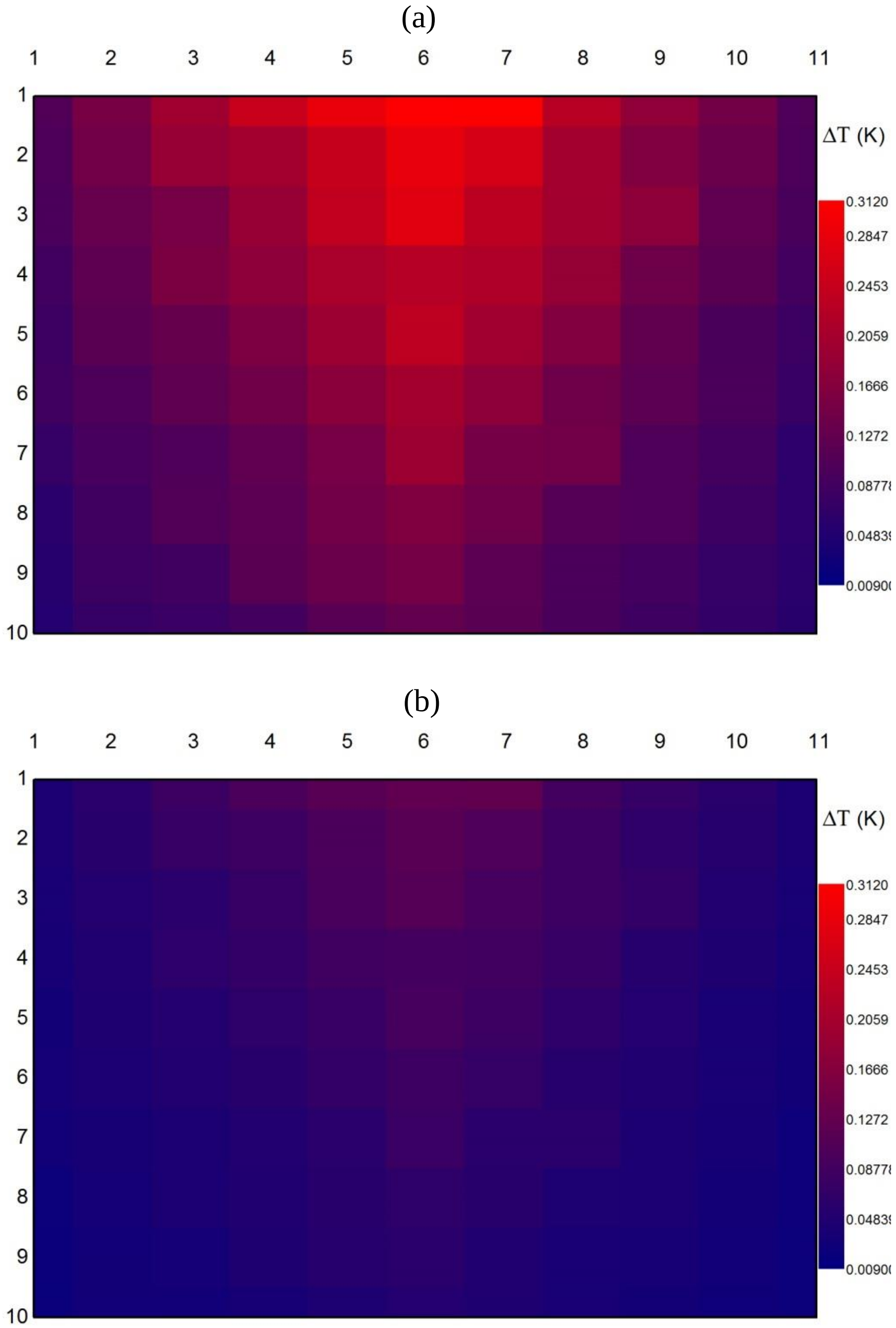

Figure (8): Temperature profiles in the (11 x 10) matrix spheres using ^{60}Co sources with: (a) 18.5GBq (500Ci) and (b) 7.4GBq (200Ci).

4. CONCLUSIONS

The designing of a water calorimeter using both the Fluent and MCNP5 codes was presented in this work. This calorimeter will be used at the Calibration Laboratory of IPEN as a primary standard for dosimetry of gamma rays of ^{60}Co and ^{137}Cs sources. The behavior of this model in two distinctive approaches, the thermodynamics and the radiation transport analyses, was studied. Using the Fluent code it could be seen how the configuration and initial parameters can drop down the temperature to start a measurement and how this flow will evolve in the internal structure of the calorimeter. The simulation showed that approximately 30min is enough to successfully reach and stabilize reliably the operation temperature. From Figures (4) and (5) it could be concluded that the horizontal and vertical profiles are suitable for measurements at 277K.

The MCNP5 simulation provided the energy deposition profile along the entire volume and showed that sources from 7.4GBq (200Ci) already can be used with commercial thermistors, because their typical sensibility edge is 0.01K for this temperature range. Figures (6) and (7) show how the energy was deposited in all spheres and Figure (8) provided with Equation (2) the expected temperature differences relative to 277K.

This work shows that this model is reasonable for a calorimeter as a dosimeter since the radioactivity exceeds approximately 7.4GBq (200Ci) for a standard ^{60}Co square beam assuming an exposure time of 45min. Further work will be toward the construction of a prototype and a future comparison between simulated and experimental data.

ACKNOWLEDGMENTS

The authors thank the Brazilian agencies CNEN, CNPq, FAPESP, MCTI and CAPES for partial financial support. The ESSS company is acknowledged for the training and the support using the Ansys Fluent software.

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