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OPTIMIZATION OF THE METHOD FOR PREPARING WATER-EQUIVALENT SOLID SOURCES

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ABSTRACT

The Nuclear Metrology Laboratory at IPEN has been involved on developing radioactive water-equivalent solid sources prepared from an aqueous solution of acrylamide polymerized by high dose ^{60}Co irradiation. The sources have been prepared with ^{57}Co, ^{137}Cs and ^{133}Ba radioactive solutions. In this paper, the study of the distribution of radioactive material in the resin is presented. The uniformity obtained was around 1 %, in agreement with the literature.

1. INTRODUCTION

Ionization chambers for activity measurements have cylindrical geometry with a re-entrant well [1] and are used to measure liquid radioactive sources in ampoules. In these sources the radioactivity is homogeneously distributed in an aqueous solution with density of around 1 g cm^{-3}. The ionization chambers used in Medical Services, generally called “dose calibrators” or “activity calibrators”, are calibrated by the manufacturer using conventional standard solutions. Nevertheless, for quality assurance, a periodical control of the response of these calibrators is required.

In general, the sources used for this control are, in general, gamma reference sources with energy in the range from 100 keV up to 700 keV with long half-life. The radionuclides recommended are ^{57}Co, ^{133}Ba and ^{137}Cs, which are characterized in terms of activity, can be used for testing the constancy of the calibrator daily, a procedure advisable to be done. To avoid contamination and handling this source type with safety it was made of resin with the radionuclide uniformly distributed.

To supply this type of sources for Medicine Services in Brazil, the Nuclear Metrology Laboratory at IPEN developed the method of preparing radioactive water-equivalent solid sources using an aqueous solution of acrylamide polymerized by a high dose of ^{60}Co irradiation.

The sources have been prepared with ^{57}Co, ^{137}Cs and ^{133}Ba radioactive solutions. The final activity of the sources is: 185 MBq, 9.3 MBq and 5.4 MBq, respectively, Fig. 1. These sources are suitable for performing constancy and accuracy tests, mainly for Medical Radionuclide Calibrators. The sources have density similar to water and good uniformity. In this paper, the study of parameters involved in the procedure to improve the preparation method is presented.

Figure 1. Water equivalent sources of ^{57}Co, ^{137}Cs and ^{133}Ba.

2. EXPERIMENTAL METHOD

The solid sources were prepared on the basis of the method developed by Sahagia and Grigorescu [2], which consists of dispensing the radioactive solution into a container with distilled water and a colour agent. After 24 hours, an aqueous acrylamide solution mixed with small quantities of EDTA was added to the previous solution and exposed to an high dose of ^{60}Co gamma radiation. The concentration of EDTA and the water aliquot used in the final solution were set in order to aid the polymerization and to obtain a good uniformity in the distribution of the radioactive material inside the polymer, respectively.

In the present study the activity of the sources was around 300 kBq, differently from the activity of the sources for the medical services, for this reason an aliquot of carrier was added to the distilled water with the radioactive solution in order to simulate the final volume of radioactive material.

The sources were irradiated in a Gammacell-220 manufactured by the Atomic Energy of Canada Ltd., with 4.53 kGy h^{-1} for 1:10 h, a dose defined previously [3]. Initially one sample was irradiated at a time. Fig. 2a shows the container for one sample (C1). As can be seen, ground ice is placed around the sample to avoid boiling the acrylamide radioactive solution, overflow and the formation of air bubbles during the polymerization. To allow irradiation of more than one sample in a single run, containers for two (C2) and for four samples (C4) were also tested by checking the sources uniformity, Fig. 2b and 2c.

a – C1 **b – C2** **c – C4**

Figure 2. Containers used for polymerization in the Gamma cell.

3. RESULTS

The uniformity of the resin was obtained measuring the activity of pieces of polymerized resin in an HPGe spectrometer, previously calibrated with IAEA standard sources of ^{152}Eu, ^{133}Ba and ^{137}Cs. Fig. 3 shows the source obtained in polymerized acrylamide resin before the cutting process. It has a soft and rubbery texture. The density value was determined by measuring mass and volume of each resin and the average density obtained was (1.06±0.02) g cm^{-3}.

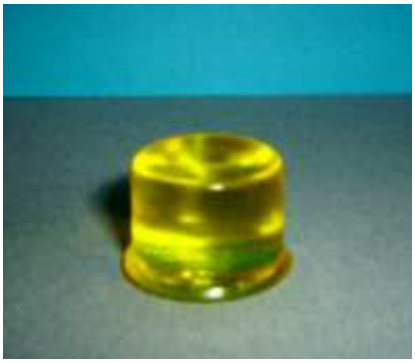

Figure 3. Polymerized acrylamide resin source used for the determination of the uniformity and of the density

The first tests for obtaining the acrylamide resin polymerization were carried out by changing the concentration of EDTA to be added in the solution. Several quantities of EDTA were tested initially without radioactive material. After establishing the best concentration of EDTA to obtain the best polymerization, tests with radioactive solutions were performed in order to verify the uniformity. Table 1 shows the results of activity obtained for ^{133}Ba sources

(sample 1, 2, 3 and 4), irradiated in the container for one sample (C1). After the irradiations, the samples 1 and 2 were left with their polyethylene vial caps opened to harden the source a little more by water evaporation from the resin. Afterwards, they were cut in three parts to measure the activity of each piece and just for comparison; the samples 3 and 4 were kept closed.

Table 1. Activity in kBq g^{-1} of the pieces of ^{133}Ba sources for studying the uniformity by leaving the polyethylene vial caps opened (samples 1 and 2) and closed (samples 3 and 4).

Sample	Bottom piece	Middle piece	Top piece	Average
1	18.85(46)	17.879(45)	17.77(46)	18.14(62)
2	19.15(49)	18.23(46)	18.48(47)	18.62(47)
3	18.07(41)	17.90(51)	17.71(45)	17.89(18)
4	17.36(42)	17.07(45)	17.16(43)	17.20(15)

The relative deviation to the average of the measurements of each sample cut in three pieces is presented in Fig. 4. As it can be seen, the maximum difference from the average was 3.9% for the sample 1 and 2 and 0.97% for the sample 3 and 4. This information led us to keep the polyethylene vial caps closed for some period before cutting the polymerized resin in parts to study its uniformity. The reason for higher uniformity variation in the open polyethylene vial (sample 1 and 2) when compared to the closed ones (sample 3 and 4) was because the top pieces of the resins hardened more rapidly than the bottom pieces due to water evaporation.

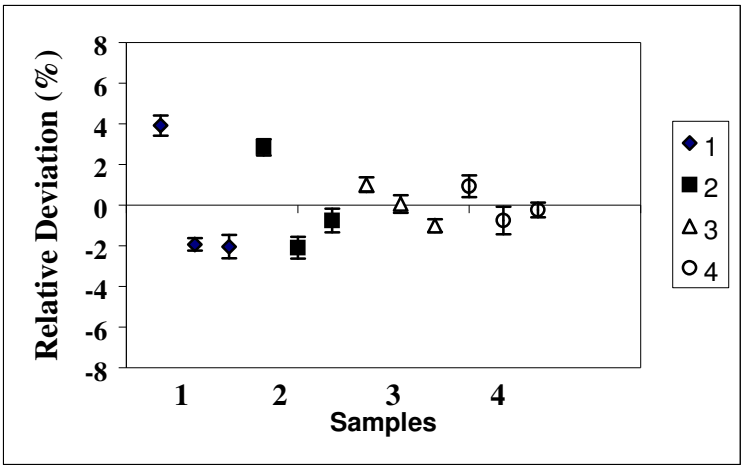

Figure 4. Relative deviation from the average activity of ^{133}Ba samples.

Table 2 shows the activity obtained for sources of ^{137}Cs, ^{57}Co and ^{133}Ba irradiated in the containers for one, two and four samples. These sources were cut in two pieces each. The containers, which have presented best uniformity, were C1 and C2, as it can be seen in the Fig.5, where the relative deviation from the average is presented. In this figure, the results of the measurements with container C4 have presented variations between 1 % and 7 %, showing that samples cast shadows on each other, affecting the distribution of activity in the samples, therefore this container was rejected.

Table 2. Activity in kBq g^{-1} of the pieces of ^{137}Cs, ^{57}Co and ^{133}Ba sources for studying the uniformity by changing the containers

C1 (container for one sample at a time)			
Radionuclide	Bottom piece	Top piece	Average
^{137}Cs	34.97(24)	35.13(31)	35.05(11)
^{57}Co	10.71(12)	10.61(5)	10.66(8)
C2 (container for two samples at a time)			
^{57}Co	10.63(5)	10.68(5)	10.66(4)
^{57}Co	10.62(7)	10.67(2)	10.65(3)
C4 (container for four samples at a time)			
^{133}Ba	35.29(17)	39.24(42)	37.3(28)
^{133}Ba	36.40(23)	35.70(16)	36.05(49)
^{133}Ba	35.74(14)	36.81(29)	36.28(76)
^{133}Ba	34.21(35)	39.04(78)	36.6(34)

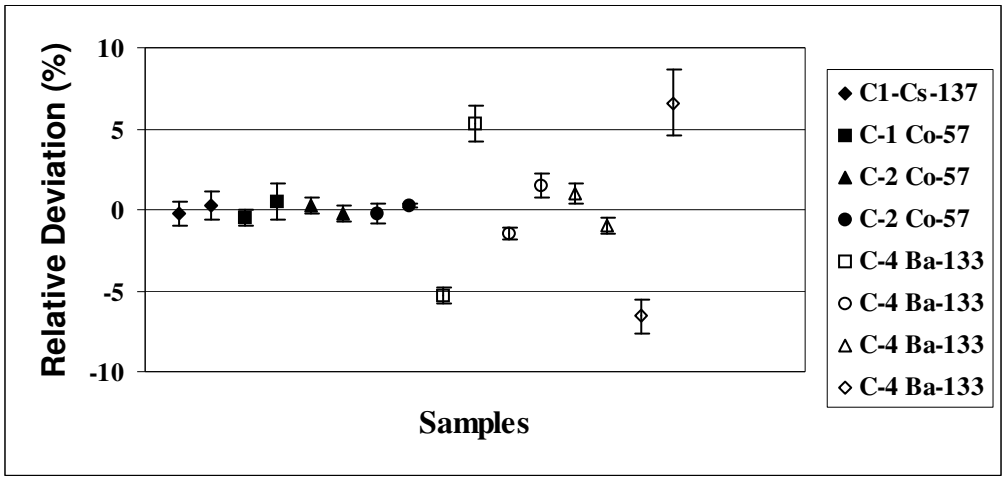

Figure 5. Relative deviation from the average activity of samples irradiated in different containers

4. CONCLUSIONS

The results presented in this paper showed that the method for preparing water-equivalent solid sources developed by the LMN was improved concerning to the uniformity by using appropriate containers for irradiations as well as by leaving the polyethylene vial caps with the polymerized resins closed for some period before cutting them in small pieces. The solid sources prepared have good uniformity, around 1%, in good agreement with the values presented in the literature [2,4].

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