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Characterization of Ceramic Seeds with Samarium-153 for Use in Brachytherapy

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Ceramic seeds were synthesized by the sol-gel technique with Si:Sm:Ca. One sample was irradiated in the TRIGA nuclear reactor IPR-R1. After irradiation, the seeds were submitted to instrumental neutron activation analysis to determine the ^{153}Sm concentration in weight. The same irradiated seed sample was submitted to gamma spectrometry analysis to determine all existing radionuclides as well as its individual activities. A second sample was submitted to ICP-AES atomic emission spectrometry. A third sample was submitted to X-ray fluorescence spectrometry to determine qualitative chemical composition. The measured activity was due to ^{153}Sm with a well-characterized gamma spectrum. The X-ray fluorescence spectrum demonstrates that there is no discrepancy in seed composition. Maximum range of beta particles from ^{153}Sm were evaluated, as well as the total dose and dose rate on its range´s volume. The results are relevant for investigation of the viability of producing ^{153}Sm radioactive seeds for use in brachytherapy.

Keywords: *ceramic seed, radioactive seed, samarium, radiotherapy*

1. Introduction

Cancer affects great part of the world population. Therefore, any techniques for controlling neoplastic tissues must be carefully investigated. One of the efficient methods of treatment is brachytherapy[1] which implants radioactive seeds into the tumor giving a high local radiation dose, capable of eliminating tumoral cells and preserving the surrounding healthy tissue.

The radioactive seeds for brachytherapy currently used on clinical applications are made of a 0.8 mm diameter, and 5 mm length titanium pipe filled with 59.4 d half-live I-125, decaying by electron capture, followed by X-ray and 35.5 keV gamma photon emissions. The required number of seeds, their dimension, and the permanence into the tumor complicates its implantation. Therefore, the development of new ways of tumor implants applying radioactive devices is justified. Attention shall be paid to β radiation emitters that, besides having a higher linear transference of energy have limited range. Most of the energy is, then, delivered into the tumor[2].

The isotope Sm-152 incorporated in ceramics synthesized through sol-gel route, can improve brachytherapy, since the seeds can be produced in suitable dimensions[3,4]. After neutron activation, the produced radionuclides, Sm-153, will decay by high energy β particle emission coupled with γ radiation of 103 keV. These suitable properties suggest that those seeds shall be investigated deeply since they may provide an efficient brachytherapy for various types of tumor in close future.

2. Materials and Methods

A set of seeds synthesized by the sol-gel technique were produced with Sm incorporated, composed of SiO_2, Sm_2O_3 and CaO. The nuclide precursor Sm-152 was provided by $Sm(NO_3)_3$, supplied by Alfa Aesar (Johnson Mattehy Company) with 99.8% purity. Those seeds were chemically characterized by X-ray fluorescence technique in a JEOL-JXA-8900RL spectrometer. A crystallographic analysis was made by X-rays difratometry, as well. Afterwards, samples were irradiated in the research reactor IPR1-100 kW. Induced activity, gamma spectrometry and Sm concentration were performed. Also, the activity of all possible radionuclides on the seed has been investigated. Dimensional characterization and electronic microscopy image was taken from the set of seeds.

In the preparation of ceramic seeds by sol-gel technique[5], natural Samarium was added in order to achieve concentration of 20% in weight. As the sol-gel technique involves physical transformations in the material it is important to verify the final weight concentration of Sm in the seeds.

Three seeds containing Sm-152 were irradiated in the TRIGA type 100 kW research reactor, IPR-R1 in an irradiation position with neutron fluxes of 2.8×10^{12} n/(cm^2.s) (thermal) and 2.6×10^{11} n/(cm^2.s) (epithermal)[6]. It is important to consider the epithermal neutron flux in induced activity calculation because the neutron absorption cross section for both elements in this range of energy is higher than that for thermal neutrons. The induced activity was, therefore, obtained to a large extent through the epithermal neutron flux. The induced seed activity is given by the expression[7]:

$$A_0 = \left[\frac{(m.\Theta.N_A)}{A}\right].\left[\left(\sigma_y.\Phi_{th}\right)+\left(I_y.\Phi_{epi}\right)\right].\left(1-e^{(-\lambda.t)}\right).e^{(-\lambda.d)}[Bq] \quad (1)$$

where *m* is the mass of the element to be irradiated in grams, the factor Θ is isotopic abundance of target nucleus, *A* is the atomic mass of the element, N_A is the Avogadro's number, Φ_{th} is the thermal neutron flux, Φ_{epi} is epithermal neutron flux, σ_γ is the neutron capture cross section of the target nucleus in cm^2, I_γ is the resonance integral for the epithermal neutron flux of the target nucleus in cm^2, λ is the decay constant of the produced radionuclide, *t* is the time of irradiation in seconds and *d* is the time to decay after irradiation in seconds.

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Another set of seeds was immersed in 15 mL vials filled with SBF (simulated body fluid) and left for 20 days to verify its solubility in such fluid. The vials were kept at room temperature (25 °C) and were shaken once a day.

3. Results

3.1. The sol-gel route for ceramic seeds synthesis

The seeds were produced by the sol-gel route described in details in Hench and West[5] composed by the substances and amounts presented in Table 1.

3.2. Dimensional characterization

A set of 20 seeds with Samarium was prepared. The average dimensions obtained are:

- 0.75 mm diameter; and
- 1.6 mm length.

One can observe the smaller diameter and length of the seeds compared to the I-125 seeds. The implantation tool for these smaller seeds may be reduced on diameter minimizing the effect of needling the tumor avoiding the spread of cancerous cells to the blood vessels and production of metastasis.

Figure 1 presents the seeds, after synthesis.

3.3 Electronic microscopy image

Electronic microscopy image of cross-section of the seed was taken using the X-ray diffractometry spectrometer JEOL-JXA-8900RL, in which the physical aspect of the seeds can be observed (Figure 2).

Figure 3 shows an enlarged detail of the seed.

Table 1. Substances used in sol-gel route for ceramic seeds synthesis.

Substance	Weight (g)
Tetraetil Ortosilicate – $Si(OC_2H_5)_4$	1.49
Hydrated Calcium Nitrate – $Ca(NO_3)_2.4H_2O$	0.73
Deionized water – H_2O	1.03
Nitric Acid solution – 2N – HNO_3	0.17
Samarium Nitrate	0.65

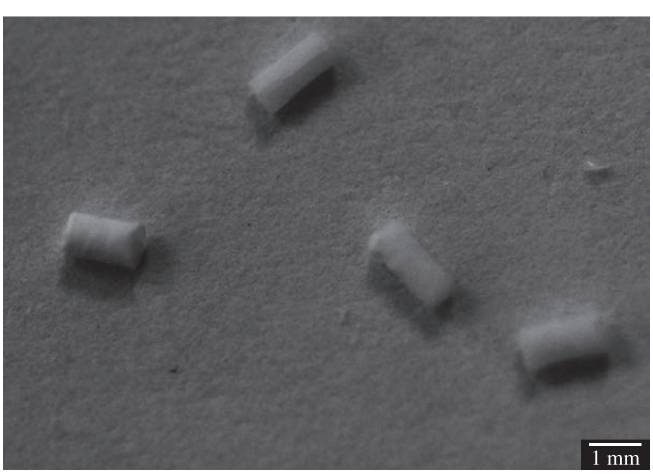

Figure 1. Ceramic seeds with samarium after synthesized by sol-gel route.

3.4. Chemical characterization of the seeds

The chemical characterization of the seeds was made using the X-ray fluorescence spectrometry in JEOL-JXA-8900RL spectrometer. The presence of the chemical elements Samarium, Calcium, Silicon and Oxygen was evident, uniformly distributed throughout the seed as shown in the spectra collected in different points of the sample, distributed on the cross-section surface of the seed. The amplitude of

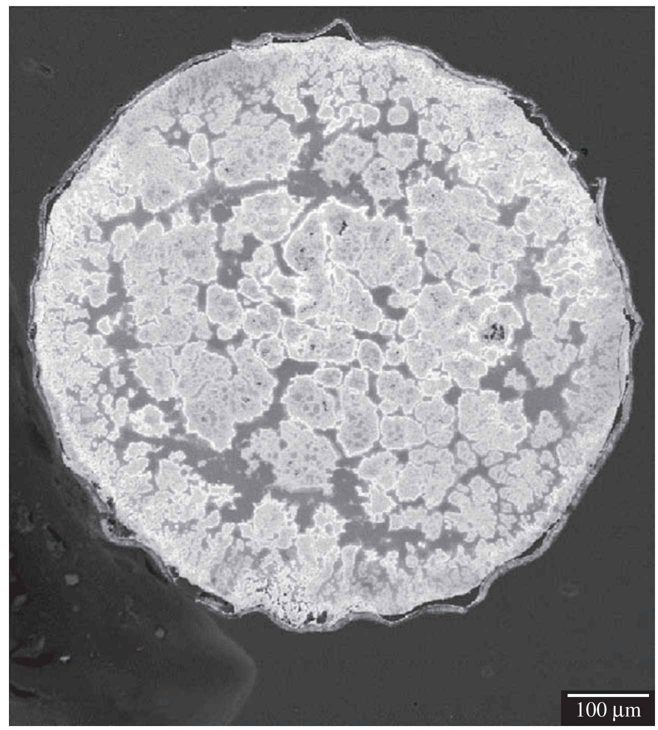

Figure 2. Electronic microscopy of the cross-section of the ceramic seed.

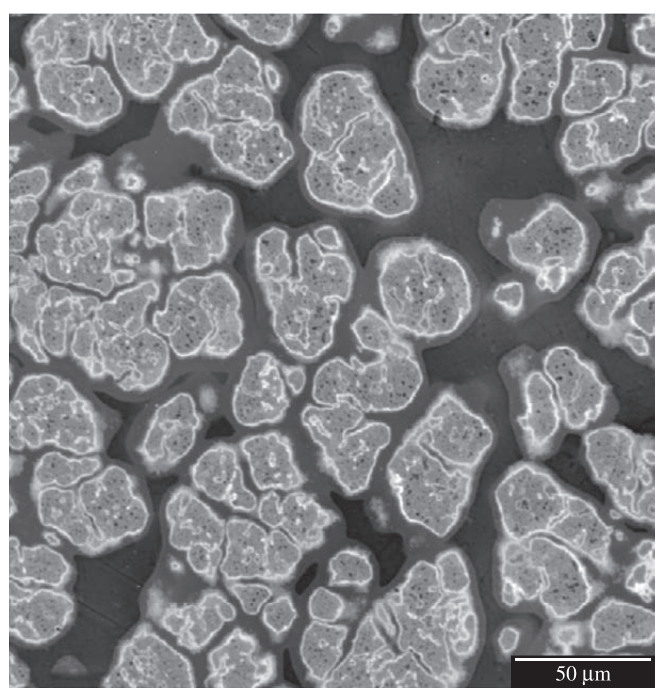

Figure 3. Enlarged electronic microscopy of the cross-section o the ceramic seed.

the corresponding peaks of each element points out, comparatively, the concentration of the element in the analyzed point. The elements Oxygen, Samarium, Calcium and Silicon keep constant peak amplitude throughout the seed, on all points randomly selected on the cross section surface. Figure 4 shows one of the X-ray emission spectra obtained for the seeds. The other spectra were so similar to each other that its presentation is unnecessary. The Gold element only appears as an artifact due to the conducting film laid over the sample to make it thermally conductive.

3.5. Crystallographic characterization of the seeds

Crystallographic analysis of ceramic seeds made by X-ray diffraction revealed an amorphous structure with some vestiges of crystalline phase. The spectrum obtained in this analysis is shown in Figure 5, in which one can observe the peak in the angle of 31°, indicating the possible presence of samarium crystalline phase. In the rest of the spectrum, nothing is evidenced but amorphism.

3.6. Determination of Samarium concentration in the seeds

In the preparation of the ceramic seeds by sol-gel technique, natural Samarium in 20% elemental weight concentration was added following stoichiometric balance. As the sol-gel technique involves physical transformations in the material it is important to verify the final amount of Sm in each seed. For this purpose neutron activation analysis (INAA) and atomic emission spectrometry (ICP-AES) had been used. The results of the analysis are shown in Table 2.

3.7. Induced activation and gamma spectrometry

The activity related to Sm-153 in each seed calculated by the expression (1) in comparison to the measured value are shown in Table 3.

As expected, the gamma spectrometry analysis demonstrates that the highest activities in the seeds are due to Sm-153 radionuclide.

The gamma radiation spectrum of seed sample is shown in Figure 6 where peaks of higher activity correspond to the energies of gamma radiation emitted by Sm-153. Other expected radionuclides, such as ^{31}Si, ^{145}Sm, ^{155}Sm, ^{41}Ca, ^{49}Ca, have short half-life or their precursor nuclides have low neutron capture cross section.

The two first peaks on Samarium spectrum are X-rays emitted by the element Eu-153 to witch Sm-153 decays after β emission.

The spectrum above 120 keV is intentionally suppressed because all measurable peaks were detected bellow that region.

Table 4 shows the calculated activity of the expected nuclides present on the seeds where it can be observed that they are very small compared with those of the desired nuclides.

4. Discussion

The use of Sm-153 in brachytherapy is very attractive due to its nuclear characteristics that are: β particle emission with maximum energy of 803 keV, 103 keV gamma radiation emission and half-life of 46.2 hour[6]. Activation can be achieved in low neutron flux reactor if enriched Sm-152 is used.

Physic and chemical characteristics of the ceramic seeds produced by the sol-gel route are suitable for use in brachytherapy: small dimensions, chemical and mechanical stability. Solubility experiments in SBF (simulated body fluid) demonstrated that they are not soluble in the first 20 days of immersion. This characteristic assures that it will not spread radioactive material in the body when implanted. Its amorphous characteristics points out to a possible solubility in long term immersion in SBF. This is an advantage as it can be completely absorbed after radioactivity has decayed.

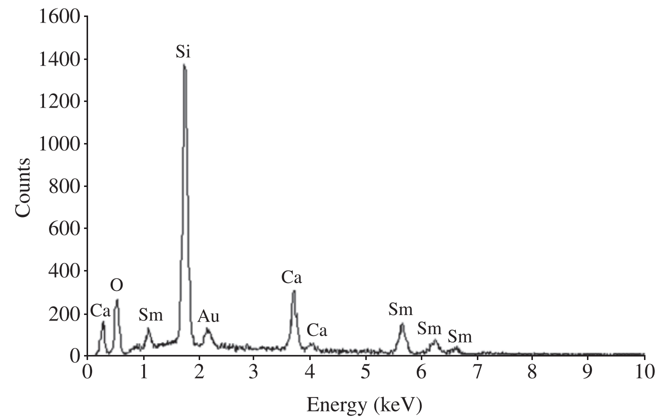

Figure 4. X-ray fluorescence spectrum of the ceramic seed.

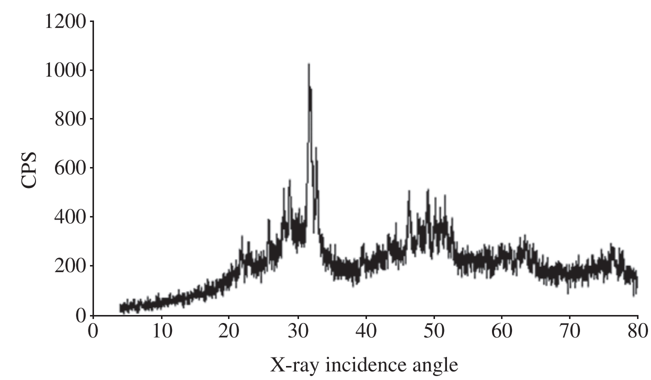

Figure 5. X-ray diffractometry spectrum of the ceramic seed.

Table 2. Samarium concentration in weight in the seeds.

Sample	Concentration (%) – INAA	Sample	Concentration (%) – ICP-AES
Sm-1A	19 ± 1	Sm-1	18.6 ± 0.4
Sm-2A	19 ± 1	Sm-2	17.5 ± 0.4
Sm-3A	18 ± 1	Sm-3	20.2 ± 0.4

Table 3. Experimental and calculated activities of the seeds.

	Activity per seed MBq (mCi)	Number of seeds
Sm-153	14.6 ± 0.74 (0.39 ± 0.02) - calculated	6
Sm-153	39.6 ± 0.8 (1.07 ± 0.02) - measured	6

The difference between the theoretical and measured values for the seed's activities is due to position of the seeds in the reactor core as well as to the difference in the value of the neutron flux adopted in calculations and the real value.

The sol-gel route showed to be suitable for producing such seeds as it involves low temperatures and simple handling when compared with other ceramic producing process.

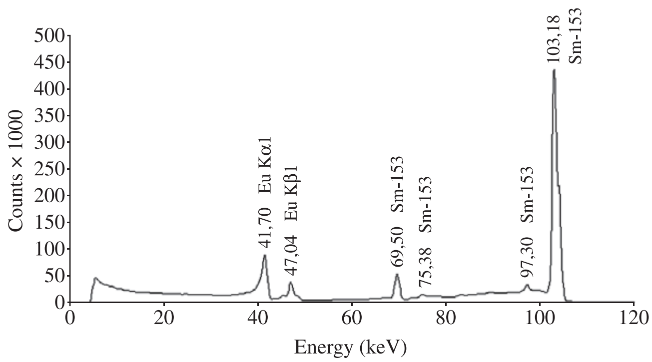

Figure 6. Gamma radiation spectrum of Sm-153.

Table 4. Calculated activity of other expected radionuclides present on the seeds.

Nuclide	Half-life	Activity after 2 hours MBq/seed
Si-31	157.3 minutes	0.004997
Sm-145	340 days	0.000003
Sm-153	46.27 hours	0.890886
Sm-155	22.3 minutes	0.009618
Ca-41	103000 years	0
Ca-45	162.61 days	0.000014
Ca-47	4.536 days	0.000001
Ca-49	8.718 minutes	0.001388

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