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A-TYPE APATITE/ESR DOSIMETRY IN THE THERAPY DOSE LEVEL TO PHOTONS

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ABSTRACT

In this report is evaluated the response of the apatite powder ESR dosimeter irradiated with ^{60}Co photons in the 0.2-100 Gy range. The specific material was developed in a previous systematic study, which established the correlation between synthesis process, structural and dosimetric properties of the A-type synthetic apatite. An excellent linear fit was obtained to the dose response in the therapy level. Sources of the uncertainty intrinsic to the dosimeter and to the ESR technique was evaluated showing the excellent precision level achievable with the apatite powder dosimeter. Improvements can be performed to increase dosimeter accuracy.

Keywords: A-type apatite, dosimetry, ESR, radiotherapy

I. INTRODUCTION

The ESR dosimetry with biological apatites has been applied worldwide in the last two decades to accidents, food irradiation and archaeological dating [1-5]. The method is based on the measurement of radiation-induced radicals in hydroxyapatite of calcified tissues. These radicals are produced from carbonate impurity substituting for phosphate (synthetic B-type apatite) and hydroxyl (synthetic A-type apatite) ion in the crystalline lattice. Several studies have been reported on the analysis of the ESR spectrum of biological and synthetic apatites. However few efforts have been made to investigate the dosimetric properties of these synthetic materials. Special features are observed in biological apatites such as lifetime of the dose marker (10^7 years), threshold dose (100-200 mGy), sensitivity to different types of ionizing radiation (alpha and beta particles, X and γ-rays, heavy ions), linear dose response in wide range. On the other hand, the increasing application of ESR dosimetry requires alternative materials to alanine dosimeter. Thus, observing the ESR spectra of synthetic apatites, a previous systematic study was developed to establish the correlation between synthesis process, structural and dosimetric properties of the A-type synthetic apatite [6,7]. A controlled synthesis process yielded a promising dosimetric material with special properties to ESR dosimetry particularly in the therapy dose level.

For this purpose, the dosimetric system should present some basic requirements, such as linear dose response, energy independence, lower limit of detection, high reproducibility, small size and high level of accuracy. In radiation therapy the deviation of the absorbed dose delivered to the target volume from the prescribed value plays an important role. An accuracy of better than 5% in the delivered dose is required for a clinical control factor (ICRU, 1976; ICRP, 1985)[8,9]. Absorbed dose measurements at the reference point in a phantom should be performed with an uncertainty of about 3% [10]. Therefore, a detailed investigation on the sources of uncertainty in the dose evaluation with apatite/ESR dosimetry is needed in the therapy dose range.

This study presents the dose response of the apatite powder dosimeter in the therapy level and uncertainty sources in the evaluation of the ESR signal amplitude to the dose assessment with the apatite/ESR system.

II. EXPERIMENTAL PROCEDURE

Samples preparation Synthetic apatite was prepared by mixing 0.5 M solution of $Ca(NO_3)24H_2O$ and 0.3 M solution of $(NH_4)HPO_4$ at 80°C. The hydroxyapatite precipitate was dried at 100°C for 24 hours. A-type carbonated apatite, $Ca_{10}(PO_4)_6(CO_3)_x(OH)_{2(1-x)}$, was produced by heating hydroxyapatite in ultra-dry CO_2 flux at 900°C, as described by Bonel, 1972 [11]. The synthesis process was established with extensive characterization of the samples. X-ray diffraction pointed out the production of

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an A-type carbonated apatite through structural unit cell refinements (a=b=9.4854; c= 6.8748), which confirmed the expansion of the parameter *a* and contraction of the parameter *c* with respect to hydroxyapatite structure, $Ca_{10}(PO_4)_6(OH)_2$. Infrared absorptions at 1534 cm^{-1} and 1465 cm^{-1} indicated that CO_3 groups occupied OH^- sites. The carbonate ion concentration in material was determined using a Multiphase Carbon Analyzer with a RC-412 Leco infrared detector [6]. The sample stoichiometry was obtained by Induced Couple Plasma. Thermal dessorption spectroscopy showed that the CO_3 decomposition is started above 800°C and reaches the maximum at about 930°C, suggesting the stability of carbonate ions in this structure.

Irradiationn The samples were irradiated in electron equilibrium conditions at room temperature (~ 21°C) with ^{60}Co Picker V4M/60 source in the Laboratory for Metrology of Ionizing Radiation/IRD/CNEN, Brazil. The calibration of the source was performed using a secondary standard ion chamber. The uncertainty in the evaluation of air kerma was ± 0.84% at 95% confidence level. The absorbed dose rate to the material was 0.4370 Gy/min.

ESR measurement ESR spectra were recorded at room temperature in a Bruker EMX spectrometer equipped with standard rectangular cavity operating in the X-band microwave range and modulation frequency of 100 kHz. The spectrometer setting were: microwave power 50 mW, sweep width 5 mT, time constant 41 ms to all spectra. The modulation amplitude, gain and number of spectral scans were changed according to the absorbed dose in the samples (0.2–100 Gy). Values of modulation amplitude from 0.15 to 0.27 mT with number of scans between 10 and 120, respectively, were used to optimize the signal-to-noise ratio. The results were normalized for modulation amplitude and gain. The mean of summed scans number was taken without applying mathematical signal treatment.

Measurements were carried out with 50 mg of powdered samples in quartz tubes of 2.8 mm outer diameter. The tubes were marked to ensure reproducible positioning inside the cavity and placed such that the centers of cavity and dosimeters were correspondent. A secondary standard was used in order to reduce the error due to small differences in measuring conditions during the experiments.

The irradiated samples were annealed at 100°C for 24 hours. After the thermal treatment the dosimeters were maintained at ambient temperature until the ESR measurements.

III. RESULTS AND DISCUSSION

The X-band ESR spectrum (50 scans) of A-type carbonated apatite irradiated to an absorbed dose of 2 Gy and annealing at 100°C is shown in Figure 1a. The lines at $g=g_{\perp}=2.0028$ and $g=g_{//}=1.9973$ are associated to CO_2^- species with axial symmetry. After the annealing the signal amplitude did not present temperature dependence up to 100 °C [12]. This apatite powder samples did not show any ESR signal before irradiation (Figure 1b).

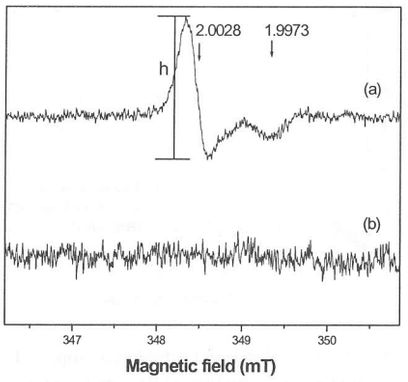

Figure 1. (a) A-type apatite ESR spectrum irradiated to an absorbed dose of 2 Gy and (b) unirradiated.

The absorbed dose evaluation was based on the peak-to-peak amplitude (h) of the line at $g=g_{\perp}$ from the radiation induced apatite signal. Spectrometer parameter setting that affect the signal height was investigated in order to optimized the signal-to-noise ratio and the detection threshold. Hence, the microwave power was selected below the saturation (above 65 mW) and the modulation amplitude (Fig. 2) smaller than the line width (0.3 mT) for obtaining a linear ESR response of the apatite over a wide dose range.

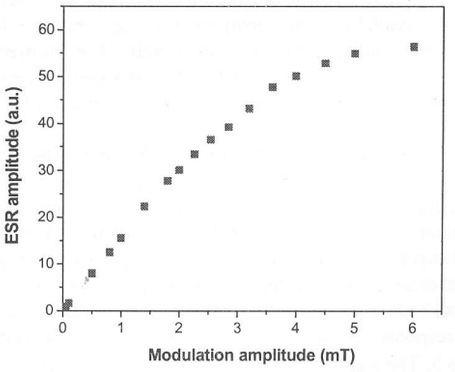

Figure 2. Variation of ESR signal amplitude with the field magnetic modulation amplitude.

In previous studies the saturation of the dose indicator signal was observed above 3 kGy [13]. Therefore, a detailed investigation was needed to evaluate the growth of the signal amplitude in the therapy dose range. In Figure 3 is shown the dose response of the apatite dosimeter from 0.2 Gy to 100 Gy. A linear fit with correlation-coefficient of 0.99951 was obtained for the dose response of the apatite dosimeter up to 100 Gy, although it can still be used up to 500 Gy in very good approximation.

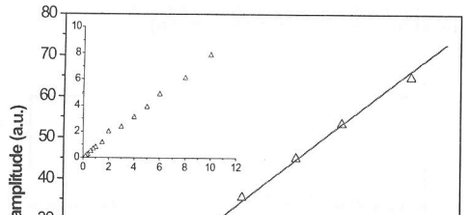

Figure 3. ESR signal normalized amplitude of apatite samples as a function of the absorbed dose.

The least squares fit to the data yielded an "initial dose" to the dosimeter in maximum of 0.7 Gy. However the unexposed material did not present ESR signal. This false "pre-dose" can be attributed to the background signal measured with the empty ESR cavity in the same parameters setting used to the dosimeter. This background signal was observed in three cavities of the same model (rectangular standard cavity). The background was the lowest at measurements performed with a new cavity and the same was observed to the "pre-dose" values. The dose response was investigated in the therapy dose level with six sample batches.

The Figure 4 presents the apatite spectrum irradiated to 0.2 Gy. A lower limit of the detection can be obtained using the available high sensitivity ESR devices, to which the signal-to-noise ratio was increased. The numerical signal treatment can still be applied filtering simultaneously background and noise in the frequency domain of ESR spectra.

A preliminary evaluation of the precision level achievable with the apatite powder dosimeter was obtained by quadratic combination of uncertainty sources intrinsic to the dosimeter and to the ESR technique. The reproducibility of the dosimeter production process was investigated with eight batches. The samples were irradiated to an absorbed dose of 2 Gy and the mean of 30 scans was taken for the dose response of each batch. The results are showed in Figure 5. The signal amplitude was reproduced within ± 1% at a 95% confidence level for the several batches of apatite dosimeter.

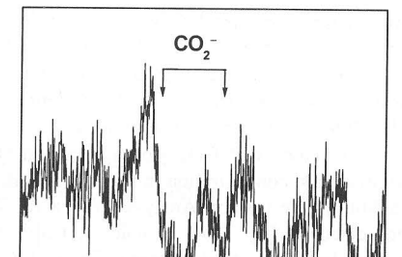

Figure 4. A-type apatite ESR spectrum irradiated to an absorbed dose of 0.2 Gy.

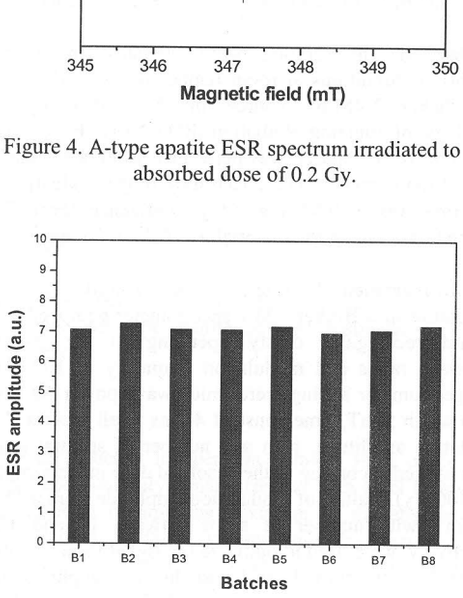

Figure 5. Reproducibility of the dosimeter production process to an absorbed dose of 2 Gy.

The repeatability test of the measurement for the apatite dosimeter of a batch was also performed at an absorbed dose of 2 Gy and 30 scans for each spectrum. In the first procedure the sample was inserted into the cavity and maintained in a fixed position. ESR signal was recorded ten times and for each one the spectrometer parameters were reset. In this conditions the dose response was reproduced within ± 0.84% at a 95% confidence level. In the second procedure, the ESR spectrum of dosimeter was measured; the sample was removed and replaced at the same height in the cavity. The spectrometer parameters were reset for each time and the spectrum was obtained for ten times. The ESR amplitude was reproduced within ± 0.88%.

The results revealed that an accuracy of at least ± 1.8% at a 95% confidence level has been achieved with apatite/ESR dosimetry system on a therapy level reference dose (2 Gy).

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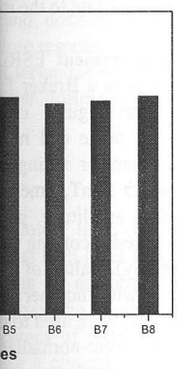

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IV. CONCLUSION

Due to the high accuracy required to clinical dosimetry, a close investigation on the each source of system uncertainty in the evaluation of dose in the therapy range is need. A first evaluation on overall uncertainty of the system showed that the apatite dosimetry is already suitable for applications in the radiation therapy level. However an improvement in the signal manipulation procedures can increase the accuracy of signal amplitude measurement to doses ≤ 2 Gy. The production quality of apatite samples plays an important role for improving the precision in the dose evaluation and it can also be obtained. The numerical signal treatment filtering simultaneously background and noise will yield better precision in the calibration curve for dose response of apatite samples.

For *in vivo* dosimetry measurements, the apatite ESR dosimetry can be used in integrative mode by using the same apatite sample positioned at the same place for each step in the fractionated treatment. In this case the higher doses increase the precision of ESR amplitude. The system can be also used to relative dose measurements in phantom, which require a precision < 2%, or to quality control of therapy beam. The apatite ESR dosimeter still offers a long-term dosimetric signal stability with a low cost material. The dose information can be readout repeatedly at different times and places, since the readout process does not affect the signal.

In conclusion, the results of the present study showed that the apatite dosimeter can be applied in the radiation therapy dose range and improvements can be performed to further increase of accuracy.

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