ESR Studies of Low Energy Irradiated Tooth Enamel

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ABSTRACT

Hydroxyapatite is one of the most promising material for retrospective and accident dosimetry. In this paper we provide an analysis of the ESR spectra of X and γ-irradiated synthetic B-type carbonated hydroxyapatite, compared with biological hydroxyapatite (tooth enamel). The energy dependence of the synthetic and biological hydroxyapatite ESR signal irradiated with low energy photons (14.3 – 21.2 keV) in the dose range between 0.2 and 5 Gy was studied aiming to span the energy response curve for energies lower than 30keV, extensively studied in the range between 30 keV, ^{60}Co until 2.5MeV. The samples were irradiated at room temperature and the measurements were carried out 48h after irradiation.

INTRODUCTION

Measurements of the concentration of stable free radicals radiation induced in calcified tissues can be associated with the absorbed dose and has been used for dose assessment in retrospective dosimetry [1-4] .

Electron Spin Resonance (ESR) dosimetry using tooth enamel is a powerful tool for the evaluation of absorbed doses for individuals exposed to ionising radiation [5]. The ESR radiation induced signals in tooth enamel are derived from interactions with carbonates ions, producing CO_2^-, CO_3^-, CO and CO^{3-} radicals.

The retrospective dosimetry method using teeth samples is based on the measurement of radiation induced CO_2^- radicals in the biological hydroxyapatite, $Ca_{10}(PO_4)_6(OH)_2$, that is the mineral phase of teeth and bones. The ESR response of tooth enamel for X-rays and gamma radiation at diagnostic and therapy energies and dose levels, as well as, for neutron therapy has been studied aiming the practical application of this technique to accident personal dosimetry [6-12].

The analysis of the ESR signal is complex, the ESR spectrum of the tooth enamel is composed of multiple elements divided in two categories: radiation induced signal with $g_{//} = 1.9973$ and $g_{\perp} = 2.0022$ and radiation insensitive signal with $g = 2.0039 \pm 0.0003$, that is thermally stable, has no fading and is not affected by radiation dose, but its amplitude depends on the grain size [9,13,14] .

The B-type carbonated hydroxyapatite ESR spectra consist mainly of the lines originated by CO_2^- and CO_3^- radicals[15], similar to CO_2^- radicals produced in tooth enamel [9]. The ESR spectrum can be predominantly due to either isotropic and orthorhombic CO_2^- species. The spectra can vary substantially with the method of sample preparation [15]. As a consequence, the choice of the synthesis parameters and their sample preparation are essential, if these materials are to be used in ESR dosimetry.

The ESR signal energy dependence of the synthetic and biological hydroxyapatite, irradiated with photons with energies between 30 keV, ^{60}Co until 2.5MeV, in the diagnostic and radiotherapy dose range, are being studied by experimental verifications and Monte Carlo calculations [16-26] . In this work we provide an analysis of the ESR spectra of X and γ-irradiated synthetic B-type carbonated hydroxyapatite,

compared with biological hydroxyapatite (tooth enamel) and the energy response for photon energies lower than 30keV aiming to contribute to the dose evaluation using this material.

MATERIALS AND METHODS

Sample preparation

a) Synthetic B-type carbonated hydroxyapatite: The samples, in powder form, with grain sizes between 80-200μm, were obtained from Odontology Faculty of the University of Sao Paulo.

b) Biological hydroxyapatite - Tooth enamel : The teeth were obtained from a private clinical and only teeth coming from exodontias were chosen. Samples of tooth enamel were obtained from non-carious molar teeth. There are different methods to separate the tooth enamel of the tooth, in this work was applied the MRRC [6] method. The roots of the teeth were cut off using a diamond saw with water irrigation;

Enamel was mechanically removed carefully from the dentin with a dental drill with diamond microsaw 0.5mm thick with water irrigation. UV light, 365nm [6], was used to identify the dentine from tooth enamel during the mechanical separation. The tooth enamel pieces were washed in distillate water in an ultra-sonic bath , 50W, during 3h [4]. After that, the samples were dried at room temperature during 4 h. and crushed using a porcelain mortar to obtain powder with grain sizes between 85 – 185 μm.

The powder samples (120mg) were encapsulated in polyethylene tubes with dimensions: 30mm length, 2.0mm internal diameter and 2mm wall thickness, specially developed for alanine/ESR dosimetry [27], and the extremities of the tubes were sealed with paraffin.

Irradiations

The samples were irradiated with gamma radiation using a ^{60}Co Panoramic source (50TBq) installed at the Radiation Technology Centre of Ipen - CTR, and a ^{137}Cs source (600Bq), installed at the Instruments Calibration Laboratory of Ipen - LCI, at room temperature, in air, using a cylindrical detector holder of polyethylene with dimensions: 15 mm external diameter, 7 mm internal diameter, 50 mm long and 4 mm

wall thickness, that guarantees electronic equilibrium for ^{60}Co[27] energy. The irradiations were carried out with source-sample distance 40cm and dose rate 5.4 Gy/h.

For X-rays irradiations was used a Rigaku X-rays machine model Geigerflex installed at the LCI. The irradiations were performed at room temperature, in air, using only the polyethylene tubes, with source-sample distance 50cm, dose rate 8.7mGy/h and effective energies of 14.3; 17.7 and 21.2keV. The irradiation parameters of low energy NPL X Ray qualities are presented at Table 1 and the X- rays spectra at Figure 1.

After irradiation the powder samples were placed in quartz ESR sample tubes and stored in controlled environment with low relative humidity during 48h, aiming to eliminate transient signals.

ESR measurements

The ESR measurements were performed using a conventional X band spectrometer Bruker-EMX installed at Physical Institute of the University of Sao Paulo. The ESR spectrometer parameters settings selected for tooth enamel measurements are show in Table 2.

The first derivative of the absorption spectrum is displayed and the peak-to-peak amplitude values was recorded. Each presented point is the average of 3 measurements, and the error bars the standard deviation of the mean (1σ).

RESULTS AND DISCUSSIONS

Using the parameters selected for tooth enamel measurements, ESR measurements of both type of studied samples were carried out 48h after irradiation. Figure 2 shows the ESR spectra of the B-type carbonated hydroxyapatite irradiated with ^{60}Co gamma radiation and with X-rays with effective energy of 17.7keV, and absorbed dose of 5 Gy. The obtained ESR spectra are similar to that reported by Schramm [15] . Fig. 3 shows the ESR spectra of tooth enamel irradiated ^{60}Co gamma radiation and with X-rays with effective energy of 17.7keV, and absorbed dose of 5 Gy. The obtained ESR spectra are similar to that reported by

Schramm [9].The ESR signal of the synthetic hydroxyapatite for ^{60}Co energy is approximately 25 % higher than the tooth enamel, indicating more sensitivity of the synthetic material.

The ^{60}Co relative energy dependence of the ESR signal of the synthetic hydroxyapatite and enamel in the low energy range between 14.3 and 21.2 keV, and to 660 and 1.25keV is presented at Table 3. The strong dependence of the ESR signal as a function of photon energy, in the region of photoelectric effect, is due to the high effective atomic number of the tooth enamel.

Figure 4 shows the dose response curve for tooth enamel irradiated with X-rays, 17.7keV, with absorbed doses between 0.2 and 5 Gy. A good correlation was found between the strength of the ESR signal and the absorbed dose range studied.

CONCLUSIONS

These studies show that the obtained ESR signals of B-type carbonated hydroxyapatite and tooth enamel agree with the published results.

Strong energy dependence of the ESR signal was verified for B-type carbonated hydroxyapatite and tooth enamel in the energy range between 14.3 and 21.2keV compared to ^{60}Co response. For 660keV (^{137}Cs) the energy factor found was 1.4 and 1.2 for B-type carbonated hydroxyapatite and tooth enamel respectively. The obtained results can contribute to the effort to standardise ESR tooth dosimetry.

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Table Captions

Table 1 – Low Energy NPL X Ray Qualities

Table 2 - ESR spectrometer parameters settings used to tooth enamel measurements.

Table 3 - ESR signal energy dependence of the synthetic hydroxyapatite and tooth enamel irradiated with ^{137}Cs, $^{60}C0$ gamma radiation and X rays with effective energies between 14.3 and 21.2keV, and absorbed dose of 5 Gy.

l Current (mA)	Voltage (kV)	Filtration (mmAl)	Eff. Energy (keV)	1º HVL (mmAl)	2º HVL (mmAl)	Coef. Homog.
30	25	0,44	14,3	0,25	0,34	0,68
30	40	0,68	17,7	0,53	0,81	0,59
25	50	1,02	21,2	0,89	1,40	0,58

Table 1.

Parameter	Value
Center Field	**348 mT**
Microwave Power	**10,12 mW**
Microwave Frequency	**~ 9,8 GHz**
Modulation Amplitude	**0,3 mT**
Modulation Frequency	**100 kHz**
Conversion Time	**163,8 ms**
Time Constant	**168,3 ms**
Nº of scans	**5**
Resolution	**1024 points**

Table 2.

Radiation Energy keV	B-type carbonated Hydroxiapatite u. a.	Tooth Enamel u. a.
14.3	3,600	3,300
17.7	4,600	4,500
21.2	6,100	5,900
660	1,400	1,200
1,250	1,000	800

Table 3.

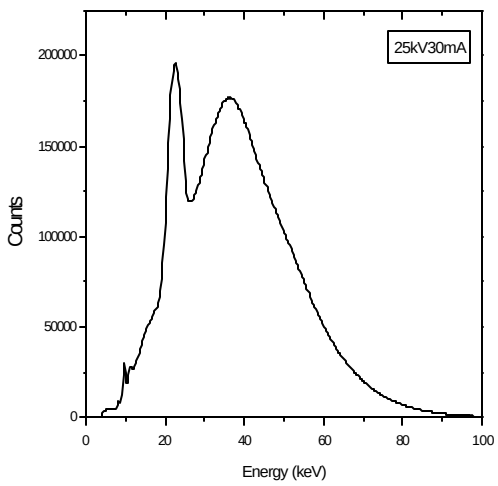
Figure Captions

Fig. 1- Spectra of low energy NPL X ray qualities with effective energies of 14.3 , 17.7 and 21.2keV.

Fig.2 - ESR spectra of synthetic B-type carbonated hydroxyapatite irradiated with ^{60}Co gamma radiation and X rays with effective energy of 17.7keV, absorbed dose of 5 Gy.

Fig.3 - ESR spectra of tooth enamel irradiated with ^{60}Co gamma radiation and X rays with effective energy of 17.7keV, absorbed dose of 5 Gy.

Fig. 4- ESR dose response of tooth enamel irradiated with X-rays: effective energy 17.7keV.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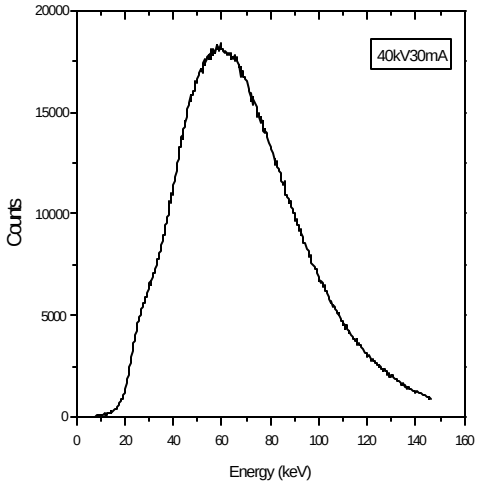
100000
150000
200000
0
20
40
60
80
100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Energy (keV)
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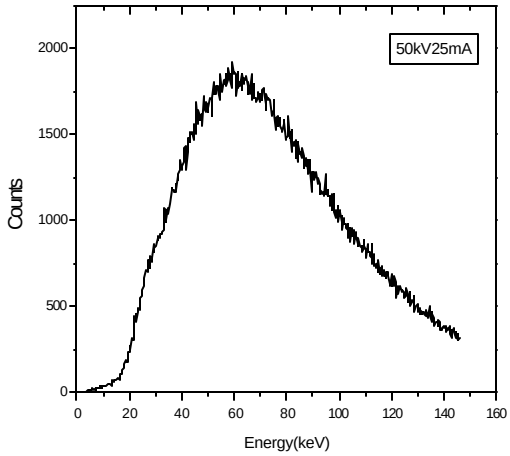
10000
15000
20000
0
20
40
60
80
100
120
140
160


50kV25mA
Counts
Energy(keV)
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500
1000
1500
2000
0
20
40
60
80
100
120
140
160

Fig.1

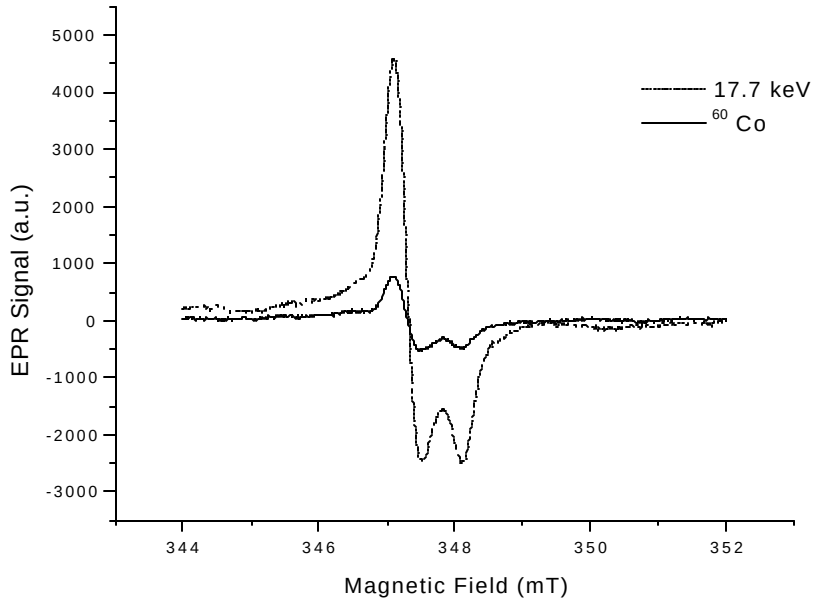
5000
4000
3000
2000
1000
0
-1000
-2000
-3000
EPR Signal (a.u.)
344
346
348
350
352
Magnetic Field (mT)
17.7 keV
60 Co

Figure 2.

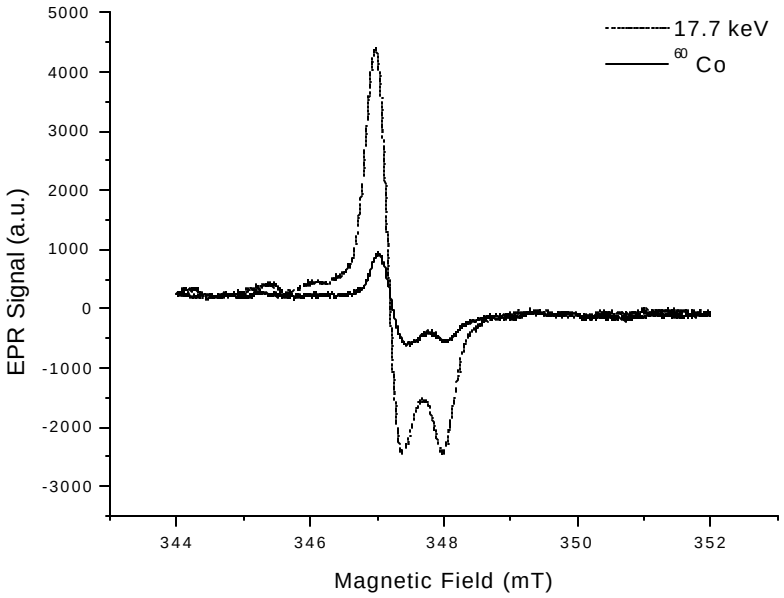
17.7 keV
60 Co
EPR Signal (a.u.)
5000
4000
3000
2000
1000
0
-1000
-2000
-3000
344
346
348
350
352
Magnetic Field (mT)

Figure 3.

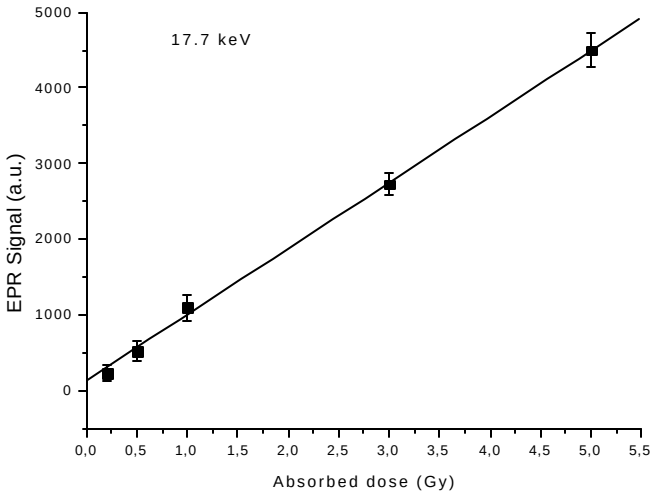
17.7 keV
EPR Signal (a.u.)
5000
4000
3000
2000
1000
0
0,0
0,5
1,0
1,5
2,0
2,5
3,0
3,5
4,0
4,5
5,0
5,5
Absorbed dose (Gy)

Figure 4.