Sintered Al_2O_3 based Materials for Radiotherapy Thermoluminescent Dosimeter Applications

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INTRODUCTION

The thermoluminescent (TL) properties of various materials have been studied since a long time but many of these developments never reach the thermoluminescent dosimeter (TLD) users. α-Al_2O_3:C TL detectors were first developed and produced at Urals Polytechnical Institute (UPI) (1) in the form of a single crystal. These TLD (model 2600 – 80) are now supplied by Victoreen to many countries, and have been used for personnel and environmental dosimetry.

Improvement in the TL and TSEE characteristics of Al_2O_3 could be achieved by the intentional introduction of lattice defects such as oxygen vacancies and impurity centers formed by carbon (2, 3).

The objective of the present work is to develop an alumina based material for application as a thermoluminiscent dosimeters (TLDs) for patients submitted to radiotherapy sessions, and also to establish the improvement on TL properties related to the microstructure, composition and samples process production. The TLDs are commonly used to determine the patient absorbed doses of radiation (4, 5) thus providing an opportunity to estimate the radiation dose received by a patient during a radiotherapy session, and to detect errors that can occur in dose delivery for a patient.

SAMPLES PREPARATION AND EXPERIMENTAL METHODS

Samples were prepared using Al_2O_3 powder supplied by Alcoa – Poços de Caldas, MG, Brazil (alumina A16 SG). To this powder it was added 1% in weight of PVA (Polivynilacetate) aglomerating additive was added. The powders were uniaxially pressed in a steel dye under 100 MPa into pellets of 7mm in diameter and 1mm thick. The green density varied from 47 to 54 % of the theoretical density. These samples were sintered at 1650^o C in air during one (batch1) and three hours (batch 2) with a heating and cooling rate of 10^{o}C/min. The sintering during three hours was made to obtain samples with larger size grains. This was done to verify if the TL response was increased. Other samples were prepared using pure Al_2O_3 and graphite powders. The samples were prepared in four differents weight percentages : 0.5 %, 1%, 10% and 20%. The homogeneization of these samples were made in a Turbula mixer for 3 to 12 hours. The powders were dryed at 45^o C during 24 hours and desaglomerated in a ágata mortar. The procedure for pressing the samples was the same used for the Al_2O_3 pure samples. The graphite doped samples were sintered at the same temperature and time schedule in a graphite resistence furnace in argon atmosphere. All the samples were characterized for phase composition and microstructure by the X-ray diffraction and the scanning electron microscopy (SEM) with dispersive X-ray spectroscopy (EDS).

In order to determine the TL characteristics of the sintered materials, such as radiation dose response, fading, lower limit detection, energy dependence and reproducibility they were submitted to ^{60}Co gamma radiation., in a panoramic source beam from Yoshizawa Kiko Co. Ltd., with an exposure rate of 30.8 $C.kg^{-1}.h^{-1}$, at a distance of 5cm. The TL behavior of the samples was also studied using a Varian Clinac 600 linear accelerator (6 MV X-ray beams and 6 to 20 MeV electron beams) for radiotherapy, on a polymethyl methacrylate phantom.

RESULTS AND DISCUSSION

Powders characterization

- **Particle size and distribution**

The method used for determining the particle size and distribution was forward laser light scattering (6,7). The powder is dispersed and fed into the sample cell where scattering takes place. The detector system measures the angular intensity for subsequent computer calculation of the particle size distribution. The particle shape is assumed to be spherical. It was found a 0.56 μm medium diameter for Al_2O_3 powder and a 20.57 μm for graphite powder were determined. The results obtained can be seen in Figure 1.

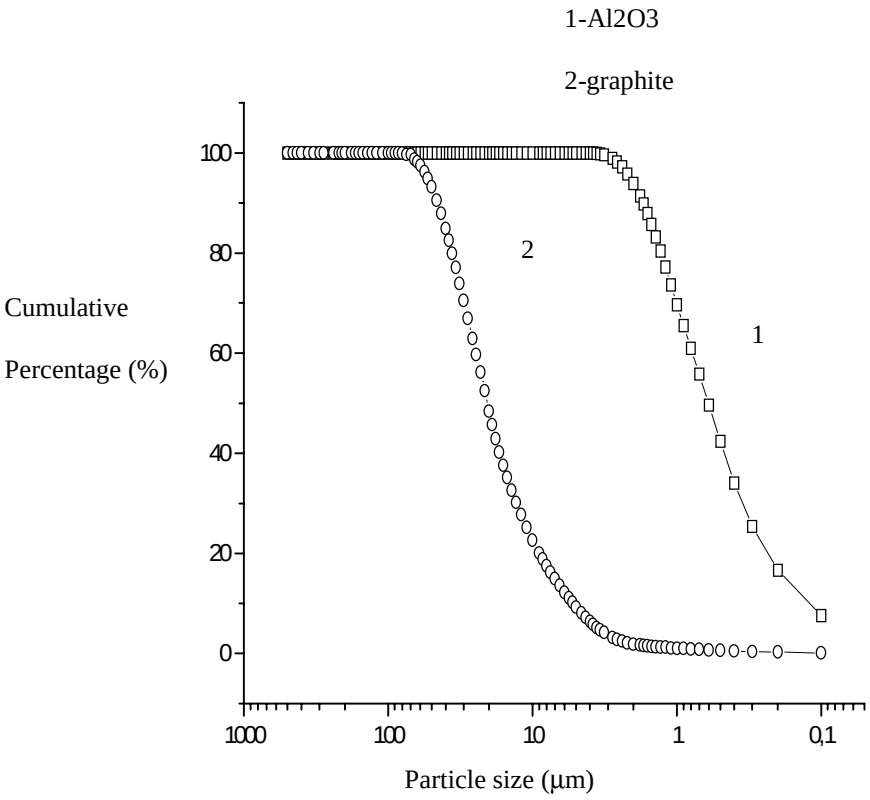

Figure 1. The cumulative particle size distribution plot.

- **Surface area analysis (particle shape)**

The surface area was obtained by a nitrogen BET specific surface area analyzer. A clean powder sample is chilled to a temperature where adsorption will occur (liquid nitrogen temperature in this case). The helium and nitrogen flows are adjusted for a specific partial pressure ratio. The gas sample is exposed to the gas mixture, resulting in a change in gas thermal conductivity as nitrogen is selectively absorbed. The area of the adsorption peak provides a means of measuring the surface area. The specific area obtained for Al_2O_3 was 8.5928 m^2/g.

- **Chemical analysis**

In relatively high-purity materials, the chemical analysis focuses on the impurity concentration. A semi-quantitative analysis was made by IPEN (Instituto de Pesquisas Energéticas e Nucleares – Diretoria de Materiais) to determine the impurity concentration in the Al_2O_3 powder. The results obtained are shown in Table 1.

Table 1. Semi-quantitative spectroscopy for Al_2O_3.

ELEMENT	B	Si	Fe	Ga	Mg	Mn	Ca	Na
PPM	4	120	30	30	300	5	350	100

- **Phase analysis**

Crystalline phases diffract x-rays according to the Bragg law, $n.\lambda = 2.d.\sin\theta$, where θ is the diffraction angle for a lattice spacing d, λ is the wavelength of the x-rays, and n is an integer. Powder or polished polycrystalline specimens are used, and the diffraction 2θ angles was recorded. The identification of a phase is accomplished by comparing the d spacing and relative intensities of the sample material with reference data for known materials. Figure 2 shows the results obtained for the α-Al_2O_3 powder, and it can be seen that this

material presents only the alpha phase.

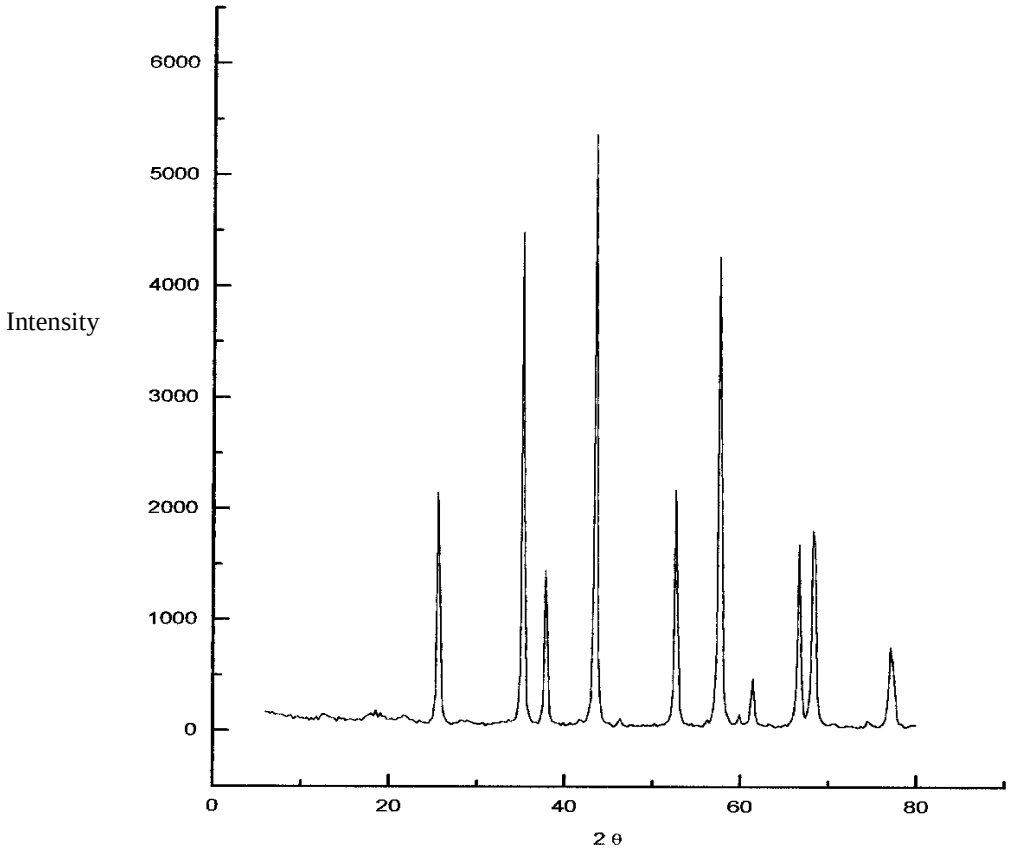

Figure 2. α-Al_2O_3 X-ray diffraction (DRX).

Sintered pellets characterization

- In each bath a DRX analysis was carried out to verify the formation of new phases, with pure and doped Al_2O_3 samples. None new phases were found in any mixture. Figures 3, 4, 5 and 6 show the results. These photos were made of a cracked pellet or polished surface, with the aim to localize the carbon in the structure of the sintered pellets. The method utilized was the scanning electron microscopy (SEM) with dispersive x-ray spectroscopy (EDS). In Figure 7, the micrography of a fratured pellet is shown. In this case, the ampliation used was 500 times, and it can be observed that the carbon was not incorporated to the alumina microstructure.

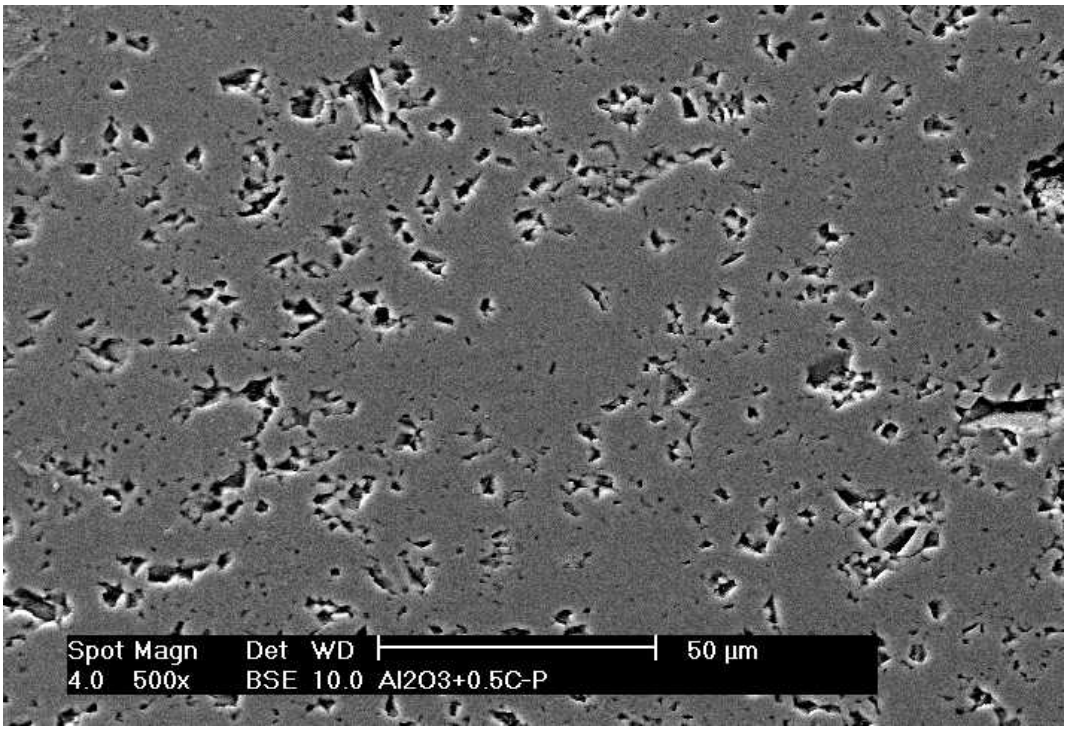

Figure 3. Al_2O_3 + 0.5% C sintered pellet polished surface. Some pores can be seen, but no different phases were observed, confirmed by the EDS analysis of this surface.

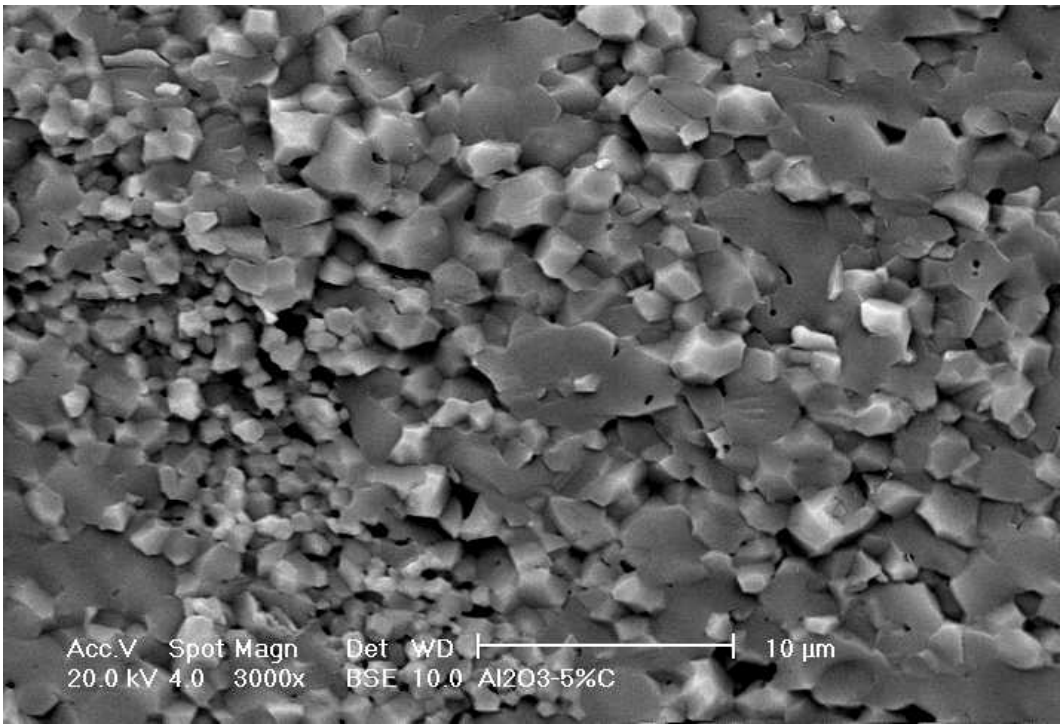

Figure 4. Microstructure of fractured pellet (Al_2O_3 + 5 % C). No carbon or new phases were observed.

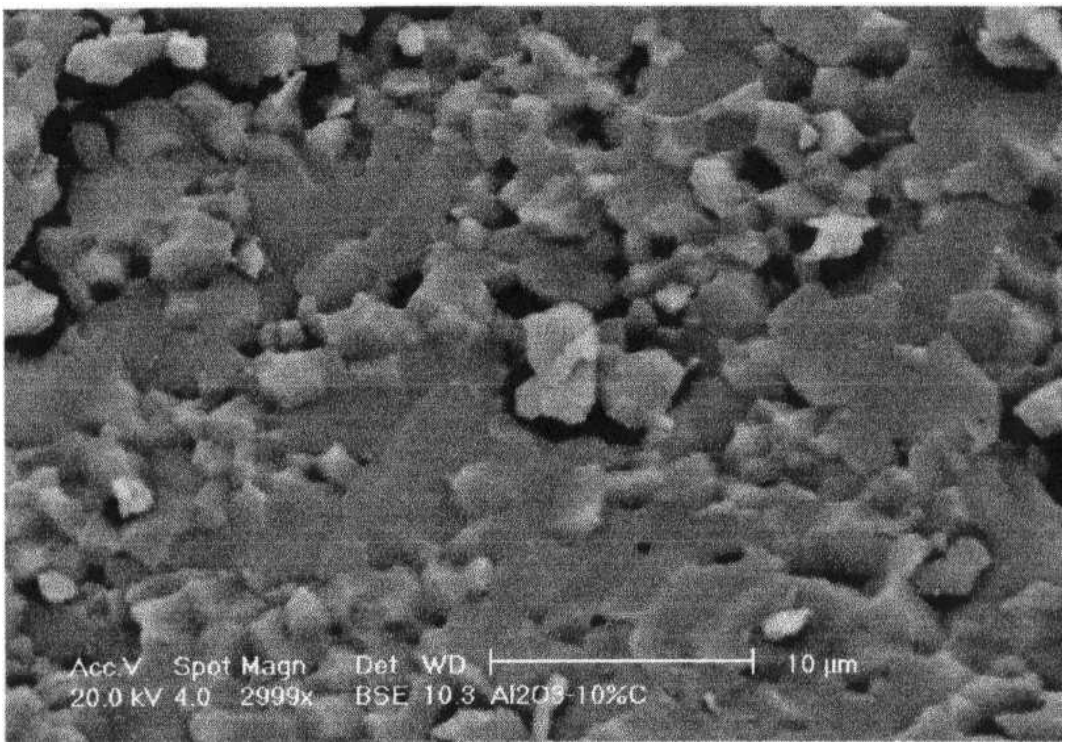

Figure 5. Microstructure of fractured pellet (Al_2O_3 + 10% C), only Al_2O_3 was observed.

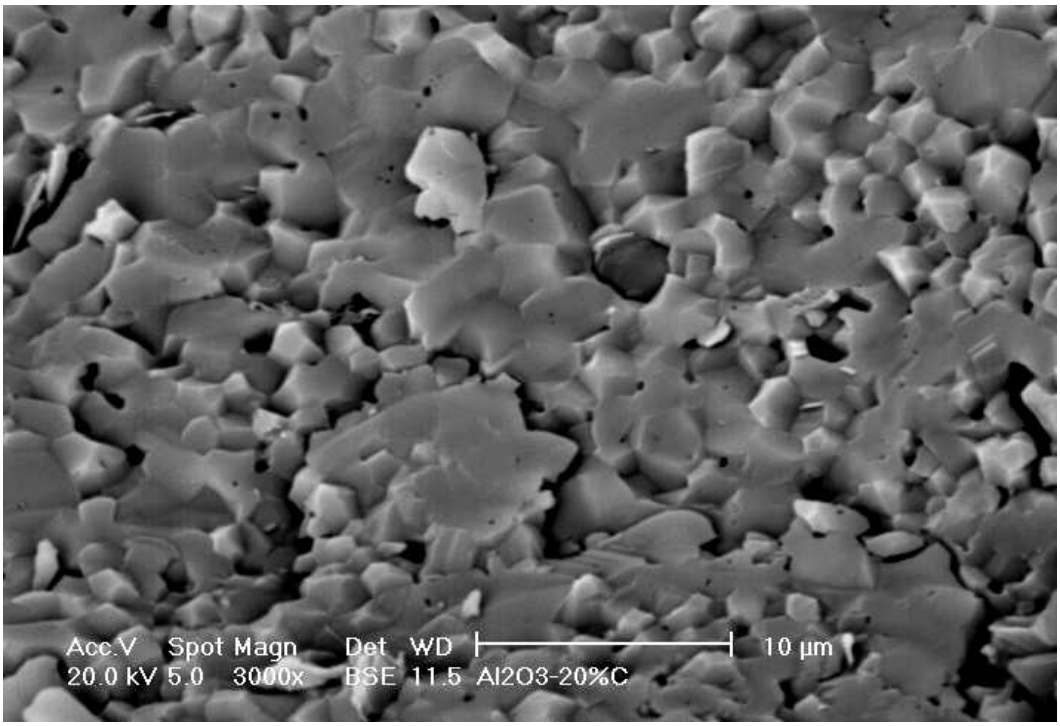

Figure 6. Microstructure of fractured pellet (Al_2O_3 + 20 % C), only Al_2O_3 was obseved .

Figure 7. Microstructure of a fractured pellet with 20% of graphite in weight.

TL Characterization

The TL glow curve for a Al_2O_3 sintered pellet shows a main glow peak at about 200°C. The reproducibility of the TL response of the pure Al_2O_3 sintered pellets and others with graphite were obtained, and after 10 readout cycles, the spread of the TL response was 3.5% for batch 1 and 3.0% for batch 2 respectively. The reproducibility of the graphite mixed Al_2O_3 sintered pellets showed no good results. This can be explained by the fact that the graphite was not incorporated to the mixture. The TL response as a function of dose was also determined a a linear region was observed in the interval of 0.01 to 100 Gy. The Al_2O_3 pellets were subjected to an absorbed dose of 50 mGy of the ^{60}Co source, and then the fading at ambient temperature was studied up to 60 days. It showed a rapid decrease (6.0% after 3 hours) reaching a relative stability (1.5%) after two days of irradiation. The energy response of the pure Al_2O_3 sintered pellets was studied in a range of electron energies between 6 and 20 MeV and X-rays beams of 6 MV. The maximum energy dependence was 48% in the case of electrons and 4.2% for X-rays beams.

CONCLUSION

The Al_2O_3 sintered pellets produced by IPEN have useful dosimetric characteristics. The tests showed that the samples sintered for three hours did not show significantly change in the TL response. The samples doped with a mixture of graphite did not present a good reproducibility and could not be used as dosemeters. All the results obtained with the pure Al_2O_3 sintered pellets are very promising for in vivo dosimetry. Samples with a mixture of graphite are being investigated to improve their properties, and to be used as TL dosemeters.

ACKNOWLEDGEMENTS

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