Development of a Dosimeter for High Doses Assessment Based on Alanine/EPR

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INTRODUCTION

The increasing use of radiation sources of high activity for industrial and medical applications and in the generation of electric energy and the consequent increase in the probability of to occur accidents, becomes important the research and the development of detectors and dosimetry methods for quality control of the applied doses.

The radiation dosimetry is important for all radiation processes, it has as objective the determination of the absorbed dose resulting from the interactions of the ionising radiation with the medium, decisive condition to the effectiveness of the dosimeter (1).

The electron paramagnetic resonance (EPR), also well-known as electron spin resonance (ESR), is a technique commonly used for high doses dosimetry of the gamma, X, neutron and electron radiations, accident dosimetry, archaeological and geological dating, studies of defects, characterization of materials, analysis of radicals, etc.

The EPR technique detects unpaired electrons trapped in the crystalline lattice. The trapped electrons are measured by microwaves absorption spectroscopy, and the intensity of the EPR signal is proportional to the absorbed dose. The non destructive nature of the EPR detection also allows the study of species trapped in biological samples such as bone, quartz, tissues, drug, teeth, hair, fingernails and dry skin. The most used inorganic materials are: $MgO:Mn^{2+}$, stalactites, stalagmites, chorales, shells, bones, minerals, $Al_2O_3:Cr^{3+}$(ruby), and the organic ones are the amino acids (alanine) and sugars (2,3,4).

Some advantages of EPR are: the information is cumulative with the dose, the reading is not destructive, high sensibility, handle easiness, good reproducibility, small fading of the sign, large linearity interval, doesn't present energy dependence and it allows the use of organic materials, once the measurement is made at room temperature or at the liquid nitrogen temperature.

The main disadvantages are the high cost of the EPR equipment and the fact that organic materials could not be reused, once they cannot suffer thermal treatment at high temperatures.

The use of the alanine as a dosimeter happened after the discovery of EPR technique. The alanine presents a very resolved spectrum and a great number of free radicals formed by absorbed dose unit, it is an amino acid with effective atomic number very close of the human tissue, it presents a stable and simple signal, with low background, low cost, easy handling and available universally (5,6,7,8).

The dosimetric system using alanine as detector element can be considered a reliable system for absorbed dose measurements in processes of industrial irradiation. They are also being studied and applied in medical irradiation processes, where lower doses are used (9,10,11,12,13).

MATERIALS AND METHODS

For the preparation of the dosimeters the following materials were used.

Alanine: DL-alanine Merck for biological applications with high purity (>99%).

To facilitate its handling, the alanine should be bound or encapsulated, once, pure alanine, when cold pressed, doesn't present good mechanical resistance.

Binder: As binder material were used the paraffin and an acetate polyvinyl solution with solid content around 60%. These materials don't present interference or noise in the EPR signal, before or after the irradiation.

Encapsulator: As encapsulator was used a polyethylene tube of low density with 3mm of external diameter and 2mm of internal diameter, which doesn't present interference or noise in the EPR signal, before or after the irradiation.

Detector Jacket: It was projected and developed a detector jacket in polyethylene, **(figure 1)**, with 4mm thick wall that assures electronic equilibrium ^{60}Co gamma radiation . The length of the detector jacket was determined in agreement with the length of the detector, 30mm, suitable for the cavity of the EPR spectrometer.

Detector: The detectors were prepared using alanine and binder material, or pure alanine encapsulated in polyethylene tube **(figure 2).**

The **dosimeter** comprises the jacket and detector.

Matrix: To obtain the detectors in the cylindrical form using binder material, a matrix was developed in steel, with 4 cavities with 3mm of diameter and 100mm of length, to be used in cold press.

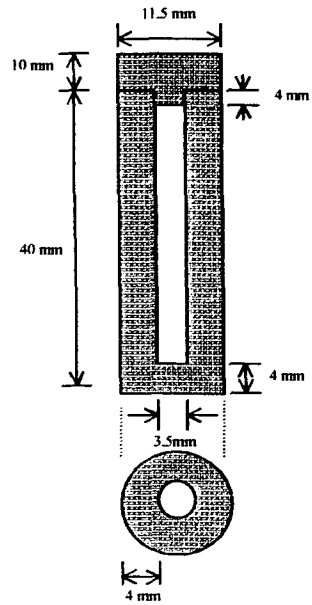

Figure 1: Polyethylene detector jacket.

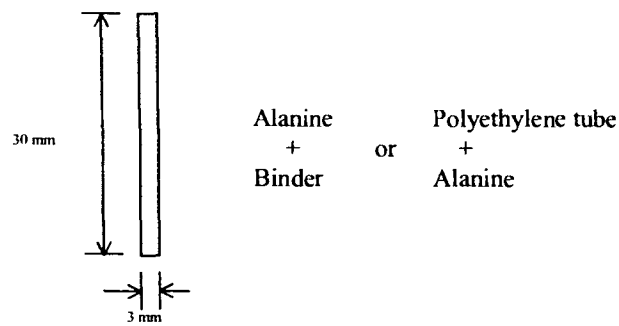

Figure 2: Alanine detector prepared with binder material or polyethylene tube.

IRRADIATION SOURCES

The X ray irradiations were performed with reference beam qualities ISO 4037 for wide spectrum (14,15) generated by one equipment Rigaku Denki 60kV, with effective energies between 14,3 and 21,2keV and a therapy equipment Stabilipan 300, with effective energies between 120 and 250 keV.

The gamma irradiations were carried out using two ^{60}Co irradiation systems, a gammacell 220 source (38,9 x 10^{13}Bq) and a panoramic source (6,94 x 10^{13}Bq).

The alanine detectors were evaluated in an EPR spectrometer Bruker model EMX with rectangular resonant cavity model ER4102ST.

The working parameters are shown in the **table 1**.

Table 1: Working parameters of EPR Bruker system.

Sweep Width	400G
Time Constant	10,240ms
Mod. Amplitude	5G
Gain	Variable
Power	10,13mW
Center Field	3480G
Resolution	1024points
Sweep Time	21s
Mod. Frequency	100kHz
Frequency	9,76GHz
Temperature of reading	20 - 25°C

DETECTORS PREPARATION

To standardize the preparation method, with the purpose of to obtain uniformity of the lot and, at the same time, to avoid problems with humidity, the alanine was always dehydrated before the preparation of any lot of detectors. It was done given a thermal treatment at low temperature: in a weigh-filter, free of humidity and impurity, a certain amount of sample was weighed and it was kept into a stove with temperature controlled at 50°C during a period of 6 hours, time enough so that no variation in the weight was observed.

During the preparation of the detectors the laboratory relative humidity was maintained between 70 and 75% and the temperature between 22 and 25°C.

Detectors in polyethylene tube

The detectors made with polyethylene tube are prepared in a simple and fast way. The tube is cut in the length of the detector (30mm) and one side is sealed with paraffin. The alanine is placed inside the tube that is sealed definitively. The mass of alanine inside of the tube is 100mg.

Alanine/paraffin detectors

The paraffin in the form of block was triturated in an almofariz at liquid nitrogen temperature.

The ratio alanine paraffin is 80% and 20% respectively. To obtain a homogeneous blend, the mixture is heated until the melt point of the paraffin (~ 60°C). With the matrix free of impurity, the mixture was placed in the cavity and cold pressed. The applied pressure was 1 ton. After this procedure, the detector is extracted of the cavity and held in dissecator. The detectors with mass of 100mg present length of 15mm, that ones with 200mg, 30mm, the diameter is the same of the cavity of the matrix, 3mm.

Alanine/pva detectors

For preparation of this type of detector a pva solution was used with 60% of solids. The ratio alanine pva solution was 70% and 30% respectively.

The mixture was homogenized and cold pressed. In this case a pressure of 50kg was enough. After to extract the detector of the cavity, it was submitted a heat treatment of 50°C, until reaches constant weight. This weight control was carried out each 2 hours; after 6 hours of heat treatment the detector was totally dry. After this procedure the detectors were maintained in dissecator. The detectors with mass of 100mg presented length of 15mm, that ones with 200mg, 30mm, the diameter is 3mm.

RESULTS AND DISCUSSIONS

The results obtained with respect to no dimension changes, no mass loss, no contamination, not to suffer humidity and handling influence, mechanical resistance and construction easiness showed that the polyethylene tube presents the best characteristic for obtaining a good detector.

Obtained the detector considered the most appropriate (tube), were performed tests to verify its dosimetric properties.

Fading

To study the fading of the EPR signal as a function of the time after the irradiation, it was considered 3 conditions:

- **ideal conditions**, the detector is maintained in dissecator and free of light.
- **natural light exposure condition**, the detector is maintained at room temperature, pressure and humidity.
- **fluorescent light exposure condition**, in a dark room the detectors were exposed to a fluorescent light at

room temperature, pressure and humidity during a period of 67 days.

The first reading, denominated zero day, was performed immediately after the irradiation with 500Gy.

After 67 days or the tenth reading, the detectors exposed to the natural light and the that exposed to fluorescent light were placed together and stored in ideal conditions. After 60 days, at ideal conditions, it was carried out the eleventh reading.

After 67 days the fading of the EPR signal of the detectors exposed to the natural light was of 45%. After 60 days stored in ideal conditions the EPR signal stays unaffected.

The detectors exposed to the fluorescent light during 67 days and stored during 60 days at ideal conditions presented fading of approximately 2%. The obtained results can be seen in the **(figure 3)**

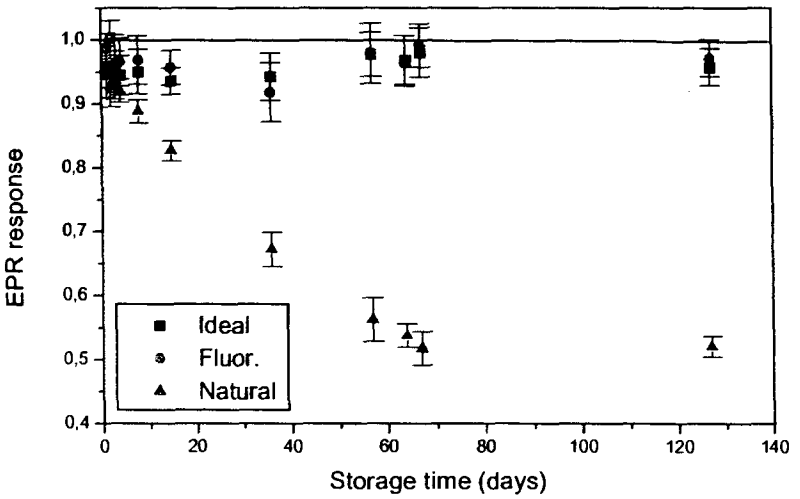

Figure 3: Fading of the EPR signal in function of the condition and of the storage time.

Reproducibility

Batch Reproducibility

To verify the reproducibility of the dosimeter a batch of 30 detectors was prepared and irradiated with 200Gy of ^{60}Co gamma radiation and measured. The observed data indicate a standard deviation of 3% (1σ).

Reproducibility of the Reading System

For study the reproducibility of the reading system a batch of 6 detectors was prepared, 3 of them were irradiated with 2Gy and the other 3 were irradiated with 500Gy, in the same conditions. With the reading parameters adjusted, each detector was measured 10 times. The analysis of the data show a standard deviation of 4% (1σ) for that with dose of 2Gy, and 3% for that with dose of 500Gy.

Dose-response Curve

To obtain the dose-response curve, 3 dosimeters were irradiated for each dose with gamma radiation of the ^{60}Co, with doses between 0,217Gy to 2.10^5Gy. The results present a linear region between 10Gy to approximately 10^5Gy, where start the beginning of the saturation of the response.

The results can be seen in the **figure 4**.

Lower Detection Limit

The lower detection limit was taken as 3 times the standard deviation (3σ) of the smallest EPR signal, and the obtained value dose was 0,3Gy.

Energy Dependence

The dosimeters were irradiated with energies between 14,3 and 1250keV, with doses of 10Gy. The obtained results were normalized for the ^{60}Co response, and they are presented in the **figure 5.**

The energy dependence of the EPR signal was observed in the range between 14,3 and 120keV. These

values were not correct for absorption in air or jacket wall.

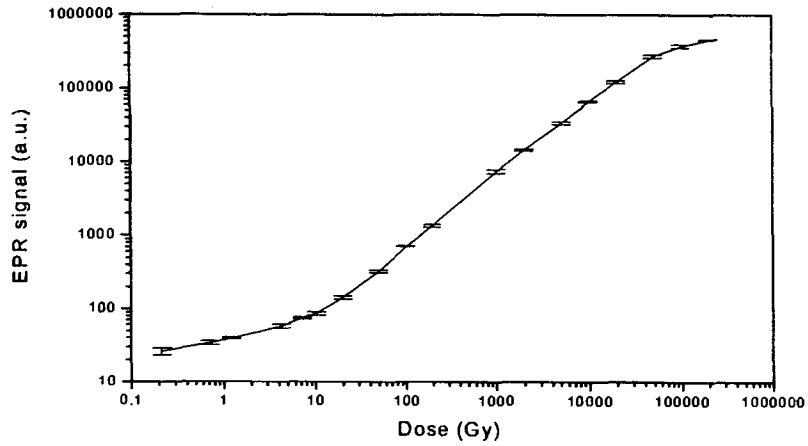

Figure 4: Dose-response curve for alanine dosimeters.

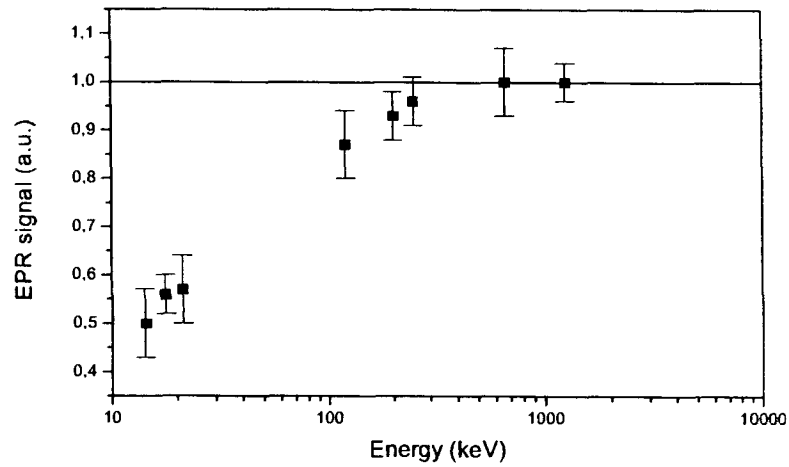

Figure 5: Relative energy response of the alanine dosimeters.

CONCLUSIONS

The reproducibility, the wide linearity interval, the stability of the sign, the low energy dependence, the handling easiness and the fact of the polyethylene doesn't present EPR signal, even for high doses, indicate that the proposed dosimeter, that is, alanine encapsulated in polyethylene tube, is suitable for application in the high doses dosimetry the EPR technique.

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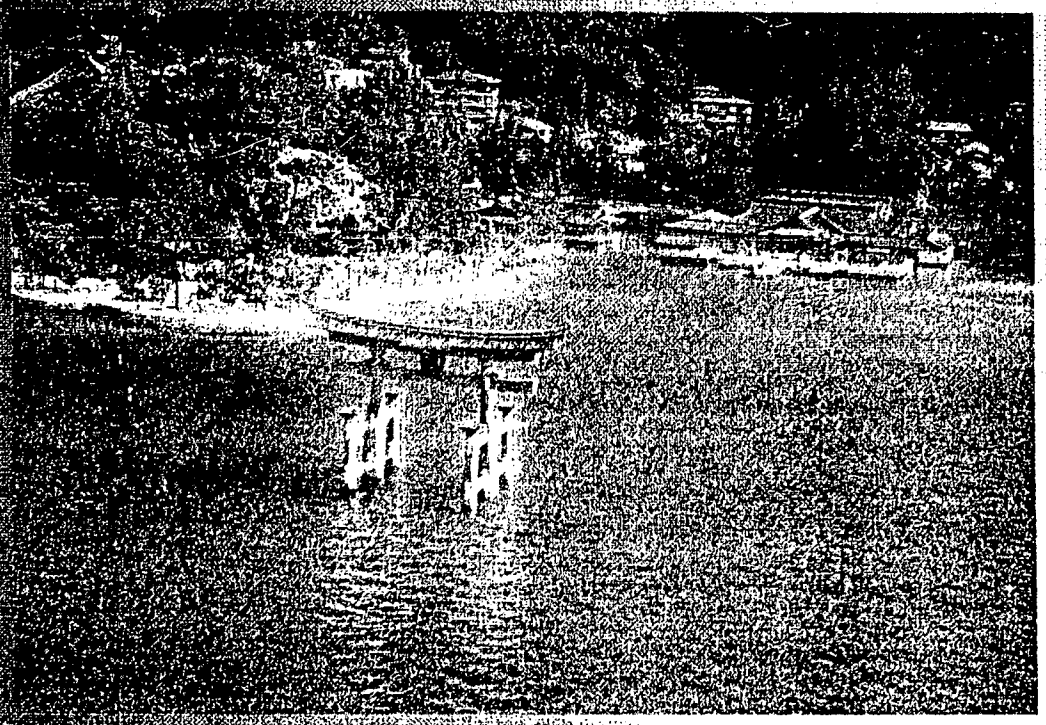

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