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:Mn²⁺, stalactites, stalagmites, chorales, shells, bones, minerals, Al₂O₃:Cr³⁺(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 ⁶⁰Co 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 ($38.9 \times 10^{13}$Bq) and a panoramic source ($6.94 \times 10^{13}$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.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sweep Width</td>
<td>400G</td>
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<tr>
<td>Time Constant</td>
<td>10.240ms</td>
</tr>
<tr>
<td>Mod. Amplitude</td>
<td>5G</td>
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<tr>
<td>Gain</td>
<td>Variable</td>
</tr>
<tr>
<td>Power</td>
<td>10.13mW</td>
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<td>Center Field</td>
<td>3480G</td>
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<tr>
<td>Resolution</td>
<td>1024points</td>
</tr>
<tr>
<td>Sweep Time</td>
<td>21s</td>
</tr>
<tr>
<td>Mod. Frequency</td>
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</tr>
<tr>
<td>Frequency</td>
<td>9.76GHz</td>
</tr>
<tr>
<td>Temperature of reading</td>
<td>20 - 25ºC</td>
</tr>
</tbody>
</table>

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 50ºC 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 25ºC.

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 ºC). 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 ºC, 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
The project aimed to analyze the behavior of EPR dosimeters under various conditions. The temperature, pressure, and humidity were monitored over a period of 67 days. The first measurement, referred to as Day Zero, was conducted immediately after the 500 Gy irradiation. After 67 days or the 10th reading, detectors exposed to natural light and those exposed to fluorescent light were placed together and stored under ideal conditions. An additional measurement, the 11th reading, was performed after 60 days, under ideal conditions.

After 67 days, the natural light detectors showed a 45% fading of the EPR signal. The detectors stored in ideal conditions for 60 days did not show any fading. The detectors exposed to fluorescent light for 67 days and stored for 60 days in ideal conditions exhibited a fading of approximately 2%. The results can be visualized in Figure 3.

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

### Reproducibility

**Batch Reproducibility**
A batch of 30 dosimeters was prepared and irradiated with 200 Gy of 60Co gamma radiation. The measured data showed a standard deviation of 3% (1σ).

**Reproducibility of the Reading System**
For the reading system, a batch of 6 dosimeters was prepared, 3 of which were irradiated with 2 Gy and the other 3 were irradiated with 500 Gy, under identical conditions. Each detector was measured 10 times with the reading parameters adjusted. The analysis showed a standard deviation of 4% (1σ) for the 2 Gy group and 3% for the 500 Gy group.

### Dose-response Curve

To obtain the dose-response curve, 3 dosimeters were irradiated for each dose level with gamma radiation from 0.217 Gy to 2.10^5 Gy of 60Co. The results indicate a linear response between 10 Gy and 10^5 Gy, starting at the beginning of the saturation of the response. The findings are presented in Figure 4.

### Lower Detection Limit

The detection limit was defined as 3 times the standard deviation (3σ) of the smallest EPR signal, and the detected value was 0.3 Gy.

### Energy Dependence

The dosimeters were irradiated with energies between 14.3 and 1250 keV, with doses of 10 Gy. The obtained results were normalized for the 60Co response and are displayed in Figure 5.

The energy dependence of the EPR signal was observed in the range between 14.3 and 120 keV. 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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REFERENCES


