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Calcium carbonate as a possible dosimeter for high irradiation doses



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HIGHLIGHTS

- This work analyzed the interactions of 5 MeV electron beam radiation and a 290 MeV/u Carbon beam with calcium carbonate (powder) at 298 K and at different irradiation doses.
- The main goal is to search the potential use of calcium carbonate as a high-dose dosimeter.
- High-energy radiation induces the formation of free radicals in solid calcium carbonate that can be detected and measured by electron paramagnetic resonance (EPR).
- An increase of the EPR response for some of the free radicals produced in the sample was observed as a function of the irradiation dose.
- We propose this chemical compound as a high-dose dosimeter, mainly for electron irradiation.

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ABSTRACT

The aim of this work is to analyze the interactions of 5 MeV electron beam radiation and a 290 MeV/u Carbon beam with calcium carbonate (powder) at 298 K and at different irradiation doses, for the potential use of calcium carbonate as a high-dose dosimeter. The irradiation doses with the electron beam were from 0.015 to 9 MGy, and with Carbon beam from 1.5 kGy to 8 kGy.

High-energy radiation induces the formation of free radicals in solid calcium carbonate that can be detected and measured by electron paramagnetic resonance (EPR).

An increase of the EPR response for some of the free radicals produced in the sample was observed as a function of the irradiation dose. These measurements are reproducible; the preparation of the sample is simple and inexpensive; and the signal is stable for several months. The response curves show that the dosimeter tends to saturate at 10 MGy. Based on these properties, we propose this chemical compound as a high-dose dosimeter, mainly for electron irradiation.

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1. Introduction

Research on the interaction between solids and radiation has helped to complement the knowledge of solid structures and reactions, making it possible to expand the uses and applications of radiation. As result of such interactions, free radicals can be produced.

The large variety of compounds that retain free radicals for long periods after they were produced by irradiation includes carbonates, which are among the most abundant minerals in the Earth's crust. Due to their ubiquity—they occur from limestone and

marble to invertebrate exoskeletons—these compounds play a significant role in the natural sciences, and important information about paleoclimatology, paleogeography and archeological chronology can be obtained from their analysis (Bahain et al., 1994; Gilinskaya, 2005; Nokhrin et al., 2006). The analysis of such samples can be performed by EPR of the free radicals produced in the carbonate fraction. The use of carbonates for dating, retrospective dosimetry and other applications has been intensively studied (Murata et al., 1996; Murali et al., 2001; Herrera et al., 2005; Nokhrin et al., 2006; Ureña-Núñez and Davila-Ballesteros, 2009; Romanyukha and Trompier, 2011; Vorona et al., 2013). These studies were performed with gamma radiation in the low doses range of 1–1500 Gy. To our knowledge, no prior reports are at high doses of irradiation with electron and heavy ions, as in this report.

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Several industrial or commercial applications require the use of high-radiation doses. Some of the most common applications for radiation are the sterilization of single-use medical products, food irradiation to reduce pathogens, and modifications of materials to improve their properties. For all of these applications, researchers need to know the doses these items have received.

Dosimeters used for different applications should be calibrated using irradiation conditions as close as possible to the conditions used when the irradiation dose is to be measured (Miller et al., 2009). Most dosimeters are calibrated at room temperature using well-characterized gamma ray sources.

A variety of dosimetry devices have been used for high-dose measurements in various environments—dye and chemical systems, both liquid and solid—but for some of these devices, modest alterations in the environment could change the chemical transformation rate, and corrections might be required. The choice of the particular dosimeters best suited for a given application depends on the specific conditions under which the measurements must be made.

The aim of this work is to test the applicability of commercial calcium carbonate for potential use as an inorganic dosimeter for high-irradiation doses, based on the formation of free radicals by high-energy radiation (5 MeV electron beam and 290 MeV/u Carbon beam) and using EPR (electron paramagnetic resonance) for its detection.

2. Experimental procedures

2.1. Samples

Sample: calcium carbonate (Sigma-Aldrich, Co. USA), of the highest purity available. Some samples were from J. T. Baker®, USA.

Sample preparation. Solid carbonate, as a powder, was put in glass tubes. One set of samples was kept in air, and the other set in evacuated glass tubes.

2.2. Irradiation

The irradiation was conducted using two sources. The first was an accelerator producing a 5 MeV electron beam at the NEO Beam facility in Northeast Ohio, USA. The doses were from 0.015 to 9 MGy. The second was a 290 MeV/u Carbon ion beam at the heavy ion irradiation source “HIMAC” (Heavy Ion Medical Accelerator) at NIRS (National Institute of Radiological Sciences) in Chiba, Japan. The irradiation doses were 1.5, 4 and 8 kGy. The experiments were performed using the biology beam line. Some samples were irradiated with gamma rays at ICN-UNAM in the Gammabeam 651 PT unit at 0.800 kGy, 1.5 kGy, 1.5 MGy, and 5.25 MGy at room temperature.

2.3. Electron paramagnetic resonance (EPR) measurements

The EPR analysis was conducted at the Instituto de Química-UNAM on 30 ± 0.1 mg of sample in a quartz tube at room temperature with a JEOL JES-TE300 spectrometer operating X-Band fashions at a 100 kHz modulation frequency and a cylindrical cavity in the mode TE₀₁₁. The external calibration of the magnetic field was made with a precision gauss meter, JEOL ES-FC5. The spectrometer settings for all spectra were as follows: center field, 335.5 ± 4 mT; microwave power, 8 mW; microwave frequency, 9.43 GHz; modulation width, 0.032 mT; time constant, 0.1 s; amplitude, 32; sweep time, 120 s; 1 scans. The readings were taken at the vertical peak height. The sample irradiated at 6.5 MGy with the electron beam, a sample irradiated at 5.25 MGy with gamma

radiation and the sample irradiated with the 290 MeV/u Carbon beam (1.5 kGy) were measured, changing the microwave power to observe the behavior of the peaks at $g=2.00034$ and 2.0031 . The experimental EPR spectra were analyzed and simulated using the Jeol software ES-IPRITS/TE and the isotropic simulation V. 1.11 Jeol LTD.

3. Results and discussion

The EPR spectra of the irradiated calcium carbonate consisted primarily of a group of six lines, in the center field attributable to coupling with an $I=5/2$ assigned to Mn^{2+} , obtained as an impurity in the sample. Additional free radical peaks identified as radiation-induced defects were observed at the center of this group (332.670–340 mT). The sample without irradiation showed no signal. The EPR spectra consisted of narrow lines. The signals present in our samples were as follows: (1) $g=2.019$, (2) $g=2.007$, (3) $g=2.0058$, (4) $g=2.0031$ (5) $g=2.0034$, (6) $g=2.0005$, and (7) $g=1.9939$. For dosimetric purposes, we followed the evolution of peak 5. The signal with $g=2.0034$ (5) was better resolved and easier to assign.

Other authors report these signals for low-dose gamma irradiation, in the so-called asymmetric EPR signal near $g=2$ (Callens et al., 1998). Although there have been many efforts to study these signals, little is known about their origin, due to the composite character of the signals and controversies remain in their assignment. (Barabas et al., 1989; Callens et al., 1998; Seletchi and Duliu, 2007; Rudko et al., 2010). For a detail review, see Callens et al. (1998).

It is beyond the scope of this paper to assign the nature of the radicals derived from irradiation of our experiments; also, our samples were irradiated at high doses, and the same species do not necessarily dominate at high and low doses. However, some data in the literature for the analysis of carbonate samples of different origins show various radicals and their assignment.

Several paramagnetic species such as CO_2^- (surface and bulk), CO_3^{3-} , CO_3^- , CO^- (surface and bulk) and some unknown radicals in the range of 2.00010–2.01 have been proposed, and for some of these molecular ions, more than one different type may occur. Romanyukha and Trompier (2011), reported that population of different radical species dominate the spectra depending on the radiation source used (X-ray, UV and gamma radiation). Most of these reports are performed with gamma radiation, only two with heavy ions (^{238}U and ^{197}Au) to study crystal lattice damage (Nagabhushana et al., 2008; Pabst et al., 2010). The radical with $g=2.0006$ is the most important signal for EPR-dating natural carbonates, such as shells or stalactites (Barabas et al., 1989, 1992; Debuyst et al., 1993 and Callens et al., 1998; Nokhrin et al., 2006). It has been unambiguously associated with the isotropic CO_2^- radical and identified in various carbonate samples (Barabas et al., 1992; Debuyst et al., 1993). The signal at $g=1.997$ is indicative of the presence of this radical. In general, CO_2^- centers are very stable at room temperature and can be detected for years.

It was speculated that the signal at $g=2.0058$ (peak 3 in Fig. 1) was due to organic radicals. However, this signal appeared in very pure synthetic crystals, and some authors attributed to assigned it as CO_3^{3-} (McMillan and Marshall, 1968; Serway and Marshall, 1967). In addition, it has been observed in X- or gamma-irradiated calcite, and it is also present in irradiated bicarbonate samples at doses about 50 kV for X-rays (Serway and Marshall, 1967). However, because of its composite character, other authors (Barabas et al., 1989; Callens et al., 1998) proposed that it is more probably due to CO^- , and by adsorption experiments on ^{13}C samples, Callens et al. (1998) assigned it as surface CO^- (Callens et al., 1998). They also observed that this radical is not stable at high temperatures.

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