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The Low-Radiation Dosimetry Application of "tris" Lyoluminescence using Electron Paramagnetic Resonance at Low Temperature

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We present a method for detecting very weak radiation by analyzing the inner structure of irradiated tris (lyoluminescence) materials using electron paramagnetic resonance (EPR) at low temperature. Organic materials have been looked into for use in emergency dosimetry of inhabitants around radiation accidents. However, this technology has never been applied to imperceptible radiation doses (< 0.5 Gy) because there is no proper method for detecting the change of inner structure of the subject bombed by very weak radiation at room temperature. Our results show that tris materials can be applied as a radiation detectors of very small radiation doses below 0.05 Gray, if EPR is used at low temperature (130 K ≤ T ≤ 270 K). The EPR signal intensity from the irradiated-tris sample had barely faded at all after 1 year.

Keywords : electron paramagnetic resonance, tris, radiation, low temperature, fading value, emergency dosimetric material

1. Introduction

In 2011, the Fukushima daiichi nuclear disaster was caused by the Tohoku earthquake in the Japan. Another major accident at a nuclear power plant occurred at Chernobyl in the USSR in 1986. The radiation caused not only the death of many people but also severe damage to the environment. Although people have taken an increasing interest in the radiation effects around the plant, the absorbed dose of each exposed inhabitant has not been directly measured. In general, when organic materials is exposed to a radiation accident, free radicals are created, as such an electron paramagnetic resonance (EPR) signal can be obtained from the organic material with these free radicals because of the paramagnetic properties of free radicals [1, 2].

In the 1990s, it was discovered that the peak-to-peak amplitude of the first derivative of the EPR signal for sugar was proportional to the free radical concentration of the dose, the intensity of the EPR signal had a linear response to doses up to 10^5 Gy [3], the EPR signal had a low fading value [4, 5], and the absorbed dose was independent of the photon energy (above 100 keV) [6-8]. Furthermore, organic materials have the singular advantage in that they can be found in virtually every household throughout the world. Thus, organic materials may be one of the best dosimetric materials. Typical organic materials include sugar, L-alanine, and etc [1, 2].

However, domestic organic materials are usually used to measure the highest doses, but show poor performance at very small does (< 0.5 Gy) for dosimetry. In this research, we investigated the linearity of tris materials bombarded with radiation as a function of dose in a very weak radiation region. Especially, we showed that free radicals of tris can be measured below 0.05 Gray using EPR at low temperatures (130 K ≤ T ≤ 270 K).

2. Experiments

We chose tris (sigma) as our granular sample. The granular size of the sample was about 300 ìm. The tris

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was irradiated with a continuous energy spectrum of X-rays (average energy: 50 keV, oxford instruments, Model: XTF5011) at a dose rate of 0.1 Gy/s. We used X-ray equipment that can irradiate the sample with doses from 0.0001 to 40 Gy, at room temperature, as a radiation source. A quartz tube with a 100 mg sample was put into the cavity of the EPR equipment. We used Bruker EMX-300 system as the EPR spectrometer. The EPR spectrometer was operated using the X-band, which means about a 9.7 GHz microwave frequency for the EPR resonance, with a 100 kHz modulation. The Bruker EMX-300 system is equipped with a low temperature system (130-270 K).

3. Results and Discussion

Figure 1 shows the EPR spectra of the tris according to radiation dose at room temperature. We found that irrespective of the radiation dose, the line-width of the signal was constant at 3.2 mT and the g-factor was 2.015. In general, the g-factor can give information about a paramagnetic center's electronic structure. The g-factor was found in the EPR experiments by measuring the magnetic field and the frequency at which resonance occurs. The line-width is defined in terms of the magnetic induction, and its corresponding units, and is measured along the *x* axis of the EPR spectrum, from the line's center to a chosen reference point of the line. The fading value is I/I_o (where, I_o was the intensity measured immediately following exposure to radiation and I was the intensity measured after a certain period of time.) The EPR intensity of tris increased with increasing radiation dose. The spectrum of tris was complex because the free radicals interacted with two neighboring protons [9]. The molecular structure of tris is shown in Fig. 2. An unpaired

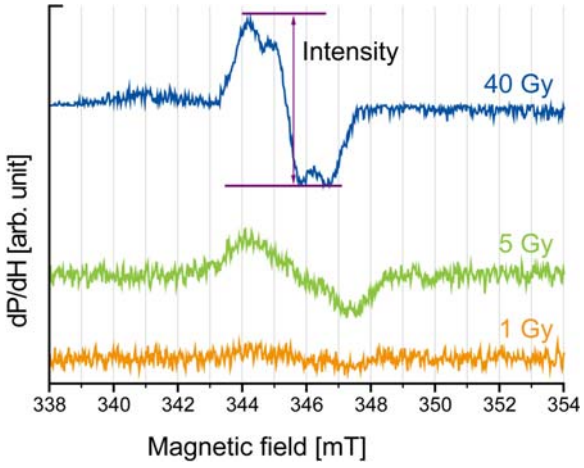

Fig. 1. (Color online) EPR spectra at room temperature of three tris samples irradiated by three different radiation doses.

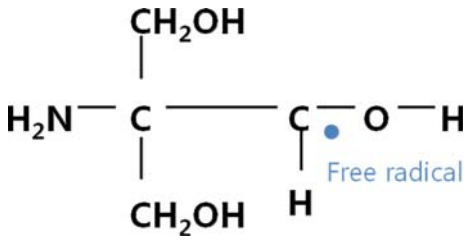

Fig. 2. (Color online) The molecular structure and electron free radical of irradiated-tris.

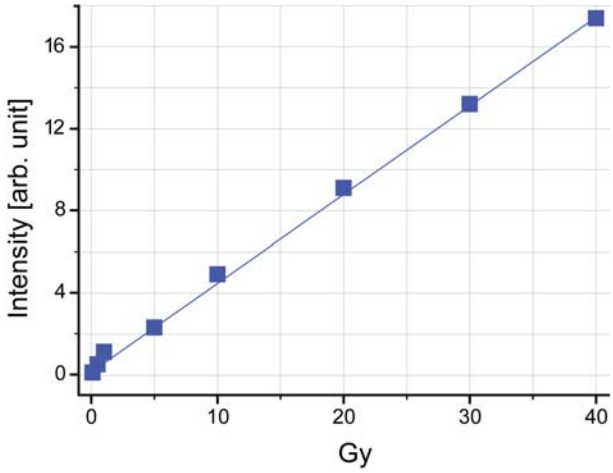

Fig. 3. (Color online) EPR signal intensities of the tris samples measured at room temperature when the tris samples were irradiated with various radiation doses.

electron can be created at free radicals in carbon-bound hydrogen by irradiation. Electron-nuclear double resonance (ENDOR) spectroscopy provided information about protons in the vicinity of the trapped electron [10-12]. In general, ENDOR spectroscopy shows that the unpaired electrons of free radicals were located in carbon-bound hydrogen [13, 14]. Therefore, we can forecast reasonably that the free radicals are created in the carbon-bound hydrogen, as indicated in Fig. 2. The sensitivity (the ratio of the relative EPR signal intensity to the absorbed dose) of the dosimetric material for radiation is one of the most important properties for an emergency dosimeter. Figure 3 shows the EPR signal intensity of tris irradiated at room temperature with respect to the absorbed dose. As the signal intensity is representative of the radical population, the intensity is linearly proportional to the absorbed dose (up to 40 Gy). This linearity helps us to estimate more exactly the radiation dose. But, the EPR signal was not observed from samples irradiated below 0.5 Gy at room temperature.

To examine whether EPR signal of the tris bombarded by very small radiation doses (< 0.5 Gy), is observed at

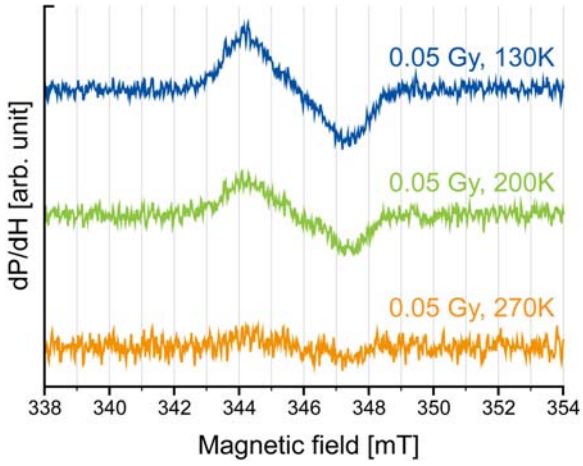

Fig. 4. (Color online) EPR spectra of the 0.05 Gy-irradiated tris sample according to temperature.

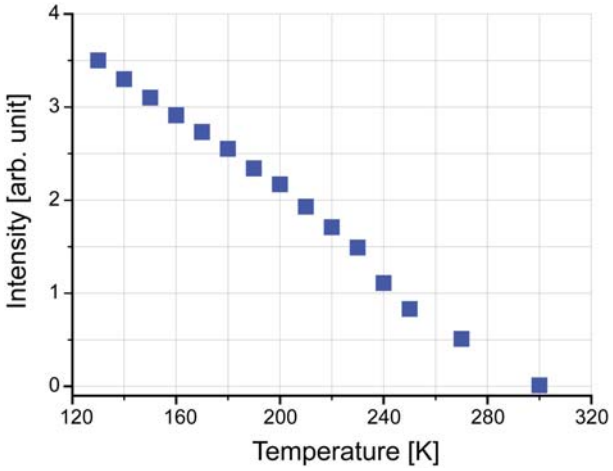

Fig. 5. (Color online) EPR signal intensities according to temperature of the 0.05 Gy-irradiated-tris sample.

low temperature or not, we measured the free radicals of irradiated tris using EPR in a low-temperature region (130 K ≤ T ≤ 270 K). Figure 4 shows the EPR spectra of the 0.05 Gy-irradiated-tris sample according to temperature. We could not observe the EPR signal of the tris sample at room temperature. However, the EPR signal of the sample at 270 K was weak, while at 130 K, the EPR signal of the sample was strong. Consequently, the EPR absorption shape of irradiated tris increased linearly with decreasing temperature. Irrespective of the radiation doses and temperature, the line-width of the signal was still 3.2 mT and the g-factor was also still 2.015. Figure 5 shows the EPR signal intensities of the tris according to temperature in the low temperature region for a low-absorbed dose (0.05 Gy). The intensity is inversely proportional to temperature. However, an EPR signal was not observed from samples irradiated below 0.001 Gy even at low temperatures. This

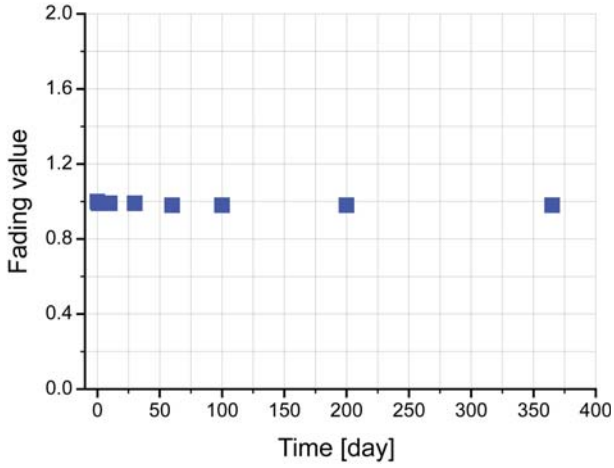

Fig. 6. (Color online) Fading of the EPR signal intensity measured at temperature 130 K according to time of 0.05 Gy-irradiated-tris sample.

signal may be detected by a lower temperature EPR system.

The second important dosimetric property of a dosimeter is "fading value" because the absorbed dose of the exposed organic material was measured and estimated some days after the accident. All samples were kept in temperature and humidity conditions of 300 K and 85% for about a year, respectively. Figure 6 shows the fading value of the EPR signal intensity measured at 130 K for 0.05 Gy-irradiated tris sample. We found that the intensity of the 0.05 Gy-irradiated-tris sample decreased slightly, about 3%, in the year following irradiation at 130 K. The fading value measured by Nakajima is somewhat different from one measured by us [4]. We also measured the fading value at various other temperatures (130 K ≤ T ≤ 270 K). The EPR signal intensity for the irradiated-tris sample hardly faded at all, at most 2-3%, after 1 year. This indicates that the tris is one of best materials for dosimetry applications.

4. Conclusions

We presented a method for detecting very weak radiation by analyzing the inner structure of irradiated tris (lyoluminescence) materials using electron paramagnetic resonance (EPR) at low temperature. Our results show that tris materials can measure radiation doses below 0.05 Gray using EPR at a low temperature (130 K ≤ T ≤ 270 K). We found that the EPR signal intensity increased linearly with increases of radiation dose, as is proper for an emergency dosimeter. Also the EPR signal intensity for the irradiated-tris sample had barely faded at all after 1 year. Therefore, we think that tris material is a good candidate for use as an emergency dosimeter.

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