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Study of the response of phenol compounds exposed to thermal neutrons beams for Electron Paramagnetic Resonance dosimetry

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HIGHLIGHTS

- Linearity of the EPR response of phenol compound with gadolinium exposed to photons.
- Linearity of the EPR response of phenol compound with and without gadolinium exposed to neutrons.
- Stability of the signal in the first 300 h after irradiations.
- Possibility of using this material for EPR dosimetry.

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ABSTRACT

This paper reports the results regarding a new organic compound (IRGANOX® 1076 phenols) with and without low content (5% by weight) of gadolinium oxide (Gd_2O_3) for Electron Paramagnetic Resonance (EPR) dosimetry of neutron beams. The dependence of EPR signal as function of neutron dose was investigated in the fluence range studied between 10^{11} cm^{-2} to 10^{14} cm^{-2}. We evaluated also the effect of gadolinium on ^{60}Co gamma photon sensitivity of this organic compound. Our analysis showed that a low concentration of gadolinium oxide (of the order of 5% of the total mass of the dosimeter) can enhance the thermal neutron sensitivity more than 10 times with a small reduction of photon tissue equivalence. The free radicals produced after irradiation of photons and neutrons are stable for more than first 300 h after irradiation. The presence of additives does not substantially modify the fading of the EPR signal induced by photons and neutrons in the first 15 days after exposure.

1. Introduction

In the last years, the development of Neutron Capture Therapy (NCT) (Sauerwein et al., 2012; Salt et al., 2004) for cancer treatments has stimulated the research on beam characterization in order to optimize the therapy procedures. The success of radiation therapy in treating cancer depends on the delivery of lethal radiation dose to the tumor, with as little as possible harm to surrounding tissues (Khan, 2010; Shani, 2000).

Among the challenges in using NCT there is the characterization of the features of the mixed radiation field (neutrons and photons) and of its components. Diamond detectors (Vatnitsky and Jarvinen, 1993), ionization chambers (Shani, 2000), semiconductor detectors (Bell and Burger, 2010), thermoluminescent dosimeters (TLDs) (Triolo et al., 2007b,a; Marrale et al., 2012a, 2013b), radiochromic film (Gueli et al., 2011) and gel Fricke dosimetry (Marrale et al., 2014b,a; Schreiner, 2004; Gambarini et al., 1994) can be employed for dose measurements.

Several research laboratories have shown an increasing interest aimed at extending the applicability of Electron Paramagnetic Resonance (EPR) dosimetry to radiotherapy with different types of

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radiation beams. In particular, EPR spectrometry provides absorbed dose measurements through the detection of the stable free radicals produced by ionizing radiations. The EPR dosimetric method has many advantages such as simple and rapid dose evaluation, the readout procedure is non-destructive, linear response of many organic and inorganic compounds (Regulla, 2005; Ciesielski et al., 2011; Gustafsson et al., 2005; Wieser, 2012; Longo et al., 2010; Fattibene et al., 2011, 2014; Lund et al., 2002; Guidelli and Baffa, 2014; Parlato et al., 2007; Marrale et al., 2007a,b, 2008b, 2009b,a, 2011b,c,a, 2012b, 2014d,c; 2013a). EPR detectors show a behavior that suggests possible applications for various kinds of beams used for radiation therapy (Herrmann et al., 2011). Nowadays, the most widely used organic compound as a dosimeter is the alanine (Baffa and Kinoshita, 2014; Marrale et al., 2007a,b, 2008b,a; Trompier et al., 2009). However, many investigations are in progress with the aim at improving sensitivity of EPR dosimetry for doses smaller than 1 Gy. These works are focused on the investigations of new materials or new mixtures of organic and/or inorganic compounds with suitable features, such as high efficiency of radiation-matter energy transfer and radical stability at room temperature (Lund et al., 2002; Marrale et al., 2009b).

Recently, our research group has started an investigation of the EPR response of some phenols compounds for possible dosimetric applications. The aim of this work is to investigate the dosimetric features of some phenols for applications in EPR dosimetry. Phenols are compounds possessing a benzene ring attached to a OH group (Fig. 1a). After irradiation the final product is a stable phenoxy radical (Fig. 1a) (Pshezhetskii et al., 1972; Marrale et al., 2014e). The stability of such radical can be improved by adding other alkyl chains which can be attached to the benzene ring (especially in the two positions closer to the OH). Alkyl chains increase the steric hindrance thus lowering the reactivity. We chose IRGANOX since it is a very interesting phenolic compound because of the production of stable radicals, its high molecular weight and it can be mixed with other constituents. His cost is very low because of its production as antioxidant agent. Moreover, the compound is stable to light and, last but not least, it is odorless.

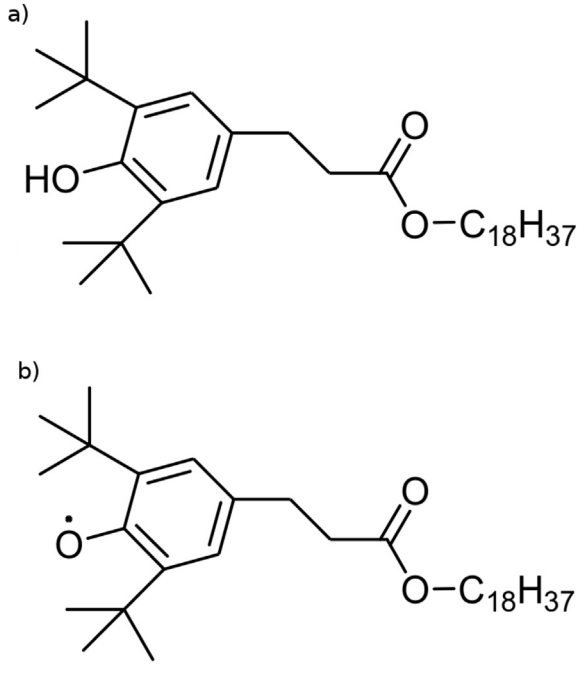

Fig. 1. a) Structure of IRGANOX phenol molecule used. b) Free radical induced after ionizing radiation exposure.

In this work we report the EPR investigation of IRGANOX® 1076 phenols with and without gadolinium oxide (Gd_2O_3) (content about 5% by weight) exposed to neutron beam. The choice of gadolinium as the additive nucleus has been made because we are interested in applications for mixed field (neutrons/photons) EPR dosimetry (Marrale et al., 2008a, 2014c, 2013a, 2011a). Gadolinium has a high neutron capture cross section and, furthermore, the high Linear Energy Transfer (LET) secondary particles (in particular, the Auger electrons) release their energy entirely inside the dosimeter volume (Salt et al., 2004; Marrale et al., 2007b). The low content of gadolinium guarantees a good trade-off between the sensitivity to thermal neutrons and the reduction of tissue equivalence.

2. Materials and methods

2.1. Sample preparation

Solid state dosimeters (diameter ≈4.0 mm, thickness ≈7.0 mm) were produced by University of Pavia using a blend of paraffin (10% by weight) as binder, and IRGANOX® 1076 produced by Sigma Aldrich (99% by weight, full chemical name: octadecyl 3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionate). The molar mass of this phenol is 531 g/mol. The average sample mass is about 120 mg. Two type of phenol pellets were producted: without and with gadolinium oxide Gd_2O_3 (5% by weight in total mixtures). In the IRGANOX® 1076 molecule, the OH of the phenol is protected by ramified alkyl chains; furthermore, the phenol ring is linked also to a long linear chain (Marrale et al., 2014e).

2.2. Irradiations

All two types of dosimeters (phenols and Gd_2O_3-phenols) were exposed to the thermal neutron column at the Triga Mark II reactor of LENA (Laboratorio Energia Nucleare Applicata) of Pavia (Italy). The dosimeters were housed in Teflon holders (height 1.9 cm and diameter 2.1 cm). In order to avoid that the dosimeters shield each other and to guarantee that they receive the same fluence, the holders were set with the axis parallel to the beam direction. The neutron fluence values measured by means of activation methods (Bortolussi and Altieri, 2007; Protti et al., 2007) and provided by the irradiation center are between 10^{11} cm^{-2} and 10^{14} cm^{-2}.

In order to make a comparison with the standard radiation beams, the two types of dosimeters were exposed to ^{60}Co γ-photons beams. The gamma irradiations were performed with ^{60}Co sources, at the IGS irradiator at the Department of Energy, Information Engineering and Mathematical Models of the University of Palermo in dose range from 12 Gy to 60 Gy. The dose rate at the effective dosimeter location was evaluated with an overall uncertainty of 2% (95% confidence level) using the ENEA (Ente per le nuove tecnologie, l'energia e l'ambiente, Italy) secondary standard ionization chamber.

For each kind of beams and for each dose value three different dosimeters were irradiated under the same conditions.

2.3. EPR measurements

The radiation-induced free radicals are detected by the Electron Paramagnetic Resonance (EPR) technique. EPR measurements were performed using a Bruker ECS 106 spectrometer equipped with a TE_{102} rectangular cavity and operating in the X-band at approximately 9.70 GHz. The following parameters were set for spectra acquisition: microwave power 4 mW, modulation amplitude 0.8 mT, modulation frequency 50 kHz (only value allowed by this experimental instrumentation), center field 347.00 mT, 4