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Evaluation of dose uncertainty in radiation processing using EPR spectroscopy and butylated hydroxytoluene rods as dosimetry system

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ABSTRACT

Butylatedhydroxytoluene (BHT) rods represent a potential dosimeter in radiation processing, with readout via electron paramagnetic resonance (EPR) spectroscopy. Among the possible sources of uncertainty are those associated with the performance of the dosimetric medium and the conditions under which measurements are made, including sampling and environmental conditions. Present study makes estimate of the uncertainties, investigating physical response in different resonance regions. BHT, a white crystalline solid with a melting point of between 70–73 °C, was investigated using ^{60}Co gamma irradiation over the dose range 0.1–100 kGy. The intensity of the EPR signal increases linearly in the range 0.1–35 kGy, the uncertainty budget for high doses being 3.3% at the 2σ confidence level. The rod form represents an excellent alternative dosimeter for high level dosimetry, of small uncertainty compared to powder form.

1. Introduction

The physical or chemical changes that ionizing radiations produce in many materials can be measured and related to absorbed dose. It is well known for instance that a number of chemical systems express radiation-induced changes that have already been evaluated for use as dosimeters in radiation processing. Thus said, there remains room in seeking alternative systems that may provide advantageous factors, with users demanding high quality at low-cost, practicality and versatility of use.

Present interest concerns cresols, from the family of methyl-phenols that occur naturally and that are also widely available as manufactured groups of aromatic organic compounds categorized as phenols (sometimes called phenolics). Butylated hydroxytoluene (BHT), 2,6-di-tert-butyl-p-cresol (the molecular structure being depicted in Fig. 1), is a particular fat-soluble organic compound, the primary use for which is as an antioxidant food additive, with further antioxidant applications in cosmetics, pharmaceutical drugs, jet fuel, rubber products and petroleum products (Xing-Tai et al., 2012; Leclercq et al., 2000; Schwarz, 1996; Parke and Lewis, 1992). In reacting with free radicals, BHT reduces the rate of autoxidation, preventing changes in colour, odour and taste (Klein et al., 2003), particularly so in foods containing fats and oils, preventing oxidative rancidity (Ahmad et al., 2015; Takami et al., 1999; Safer and Al-Nughamish, 1999; Siman and Eriksson, 1996; Xiu et al., 1994; Meyer and Hansen, 1980).

Studies of the addition of n-BHT to inhibit oxidation of polyunsaturated fatty acids (Zang et al., 1996) have made use of electron paramagnetic resonance (EPR), investigating the behaviour of different compounds of polypropylene (PP) with stabilizers such as buthyl-hydroxy-toluene (Silva et al., 2007). One important factor in the effectiveness of BHT as a preservative in foods, pharmaceuticals and cosmetics sterilized using radiation is radiostability, studied using gamma radiation on solid BHT, measured using EPR (Tuner and Korkmaz, 2007).

In brief terms, the EPR technique detects unpaired electrons trapped in the crystalline lattice, measured through microwave absorption, offering an EPR signal the intensity of which is typically expected to be proportional to the absorbed dose. To-date, EPR spectrometry has been commonly used for low- and high-dose dosimetry of gamma-rays, x-rays, neutrons and electron radiations, conducted for a wide variety of

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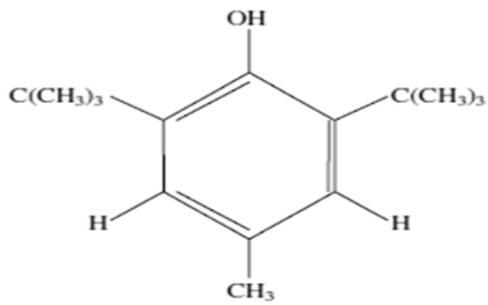

Fig. 1. Molecular structure of BHT.

applications, including accident dosimetry, archaeological- and geological-dating, studies of defects, characterization of materials and analysis of radicals. The non-destructive nature of the detection method also allows the study of species trapped in biological samples, such as bone, tissue, teeth, hair, fingernails and dry skin (Caracelli et al., 1986; Ikeya et al., 1984; Sato, 1979).

Currently, the most well established EPR dosimetry method concerns use of alanine, the latter being a widely available amino acid with an effective atomic number very similar to that of human tissue. At sufficient dose, alanine yields a well-resolved ESR spectrum, exhibiting a stable and simple signal with low background, offering ease of handling in addition to availability (Harbridge et al., 2003; Kőksal et al., 1993; Keizer et al., 1991; Gordy et al., 1955). In comparison, BHT is an odourless, white crystalline substance that is insoluble in water but freely soluble in alcohol, with a melting point of approximately 342–345 K. In present study, supported by use of an EPR spectrometer, examination has been made of the stability of radiation-induced radicals in BHT samples, with further investigation of the possibility of using BHT as a dosimetric material.

Of focal interest is the reality that to varying degree radiation dosimetry is subject to several sources of uncertainty, depending on the performance of particular dosimeters and the conditions under which measurements are made. In the use of radiation processing, recent years have witnessed increasing concern being expressed regarding dose accuracy and the influence of uncertainty on the quality of radiation measurements (Secerov et al., 2016; Anton and Büermann, 2015; IAEA, 2000). As an instance, currently different forms of alanine dosimeter system are being used to accommodate the use of a wide range of energies, dose-rate and dose, in the latter case between 10 Gy up to 100 kGy. In general, all measurements have an unavoidable degree of uncertainty due to systematic and random errors (ASTM ISO, 2015). Recently, using a ^{60}Co source (predominant gamma emissions 1.17 MeV and 1.33 MeV) Secerov et al. (2016) obtained 3% uncertainty for doses in excess of 30 kGy. For low energies (obtained in the kVp accelerating potential range), Anton and Büermann (2015) have reported uncertainty of $\leq 0.4\%$. Therefore, determination of uncertainty, providing statement of the reliability of dose measurement is crucial. To our knowledge, in recent years few studies have tackled this issue despite the widespread use of radiation processing in various fields, not least in regard to the use of the BHT-EPR Spectroscopy dosimetry system (IAEA, 2000; Caldas and Quezada, 2002; Farrar, 1991).

Tuner and Korkmaz (2007) studied EPR spectra using microwave power at two different values of temperature, examining the variations in the assigned intensities, measured with respect to the spectral base line. Building upon this in the present work, EPR spectra are obtained using microwave power and modulation amplitude providing for the greatest signal-to-noise ratio, selecting optimal values for the operating power and modulation amplitude in order to avoid saturation effects, highly important in obtaining a high performance dosimeter. Here it is to be noted that not all EPR spectroscopy studies are performed using the same specifications; thus before any such measurements are carried out and as stressed herein, it is important for study to be made of microwave power and modulation amplitude to ensure choice of the best values. Present study using BHT rods combined with EPR Spectroscopy readout has sought to estimate the dose uncertainty produced for applications in radiation processing, also investigating physical characteristics in different energy regions.

2. Materials and methods

2.1. Materials and Irradiation of BHT

BHT powder with size distribution ranging from 200 to 400 μm was obtained from El Nasr Pharmaceutical Chemicals Co. (Fig. 1). No additional purification of the material over that of the as-supplied material was performed. The BHT rods were obtained by removing the outer polypropylene tube to allow sectioning, obtaining cylindrical rods of dimension 3 × 10 mm and mean mass 84.7 ± 4.1 mg. The rods exhibit favourable mechanical properties, are safe in handling and offer multiple-use. The EVA/paraffin binding-mixture does not give rise to interference or noise in the EPR signal, either prior to or post high dose irradiation.

As a means of measuring absorbed dose, the use of BHT is based on measurement of specific stable free radicals in crystalline BHT. Present work has concerned gamma irradiations made using an Excel ^{60}Co Gammacell-220 facility (AECL, Ottawa, Canada). The absorbed dose-rate over the duration of the experiment was approximately 3.1 kGy/h. Three samples were irradiated together at the central position of the sample chamber using a specially designed holder made from polystyrene, the latter ensuring electronic equilibrium, while the central location of the samples in the chamber ensured uniform dose. Calibration was performed using the NPL standard (National Physical Laboratory, United Kingdom) alanine reference dosimeter. The samples were irradiated at room temperature. To generate stable free-radicals the radiation doses delivered to the samples ranged from 0.1 to 150 kGy. Alanine, which is dose-rate and energy independent, is widely and increasingly used in routine dosimetry for radiation processing, worldwide. It is available in both pellet and film formats based on the application. As with BTH, the response in terms of the creation of stable free-radicals is measured using an ESR spectrometer.

2.2. EPR measurements

EPR spectroscopy, upon application of radio frequency to a paramagnetic substance in the presence of a magnetic field, is based on the measurement of resonant absorption of electromagnetic energy resulting from transitions of unpaired electrons between different energy levels. The EPR signals were recorded at room temperature using an X-band EPR spectrometer (Bruker EMX, Germany). The operating conditions were: microwave power of 1.265 mW; modulation amplitude of 4.00 Gauss; modulation frequency of 100 kHz; sweep width of 200 Gauss; microwave frequency of 9.71 GHz; time constant of 81.92 ms and conversion time of 20.48 ms. The lower part of the EPR tube was fixed to ensure reproducible and accurate positioning of the sample in the sensing zone of the cavity. To take account of and to reduce orientation effects for each sample, EPR spectra were recorded for two orientations in the EPR cavity. The readings were corrected to the peak-to-peak height of the reference standard material DPPH (α, α-diphenyl-β-picrylhydrazyl) under the same operating conditions to obtain the best possible readings. The experimental conditions (microwave power and modulation amplitude) were adjusted to avoid saturation effects in the EPR spectra.

3. Results and discussion

3.1. EPR spectra of irradiated BHT

The EPR spectra of unirradiated BHT powder and BHT powder