Retrospective dosimetry based on Electron Paramagnetic Resonance

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INTRODUCTION

In all events of uncontrolled exposure of individuals to ionizing radiation when data from conventional personal dosimetry do not exist (persons from public exposed to orphan or lost radiation source, absence of wear of dosimeters, radiotherapy accident...) it is desirable to have several complementary methods for reconstructing the dose. For this reason many methods of retrospective dosimetry have been developed based on clinical, biological, physical and radiochemical approaches. Gold method does not exist and the choice of the most appropriate is dependent on case. Dosimetry based on physical methods as Electron Paramagnetic Resonance (EPR) or luminescence either thermally (TL) or optically stimulated (OSL) provide direct measurements of dose in the irradiated material. Electron Paramagnetic Resonance (EPR) spectroscopy is a physical technique for direct detection of paramagnetic molecules in matter. It is a magnetic resonance method that measures the absorption of the microwave energy between the magnetic moment spin states, when the paramagnetic molecule is immersed in a magnetic field. Among the paramagnetic molecules, relevant to dosimetry are the radicals induced by ionizing radiation. The amount of radiation induced stable radicals is proportional to the absorbed dose. EPR is a non destructive method and it has therefore the advantage to allow multiple measurements of the same sample. This work briefly reviews the methodologies and materials for EPR retrospective dosimetry that have been proposed in the scientific literature.

MATERIALS SUITABLE FOR EPR DOSIMETRY

Stable radicals or defects are the final products of the reactions initiated by ionizing radiation in most solid state materials. Though, in principle a large number of materials might be eligible for EPR measurement, suitability of a material for retrospective dosimetry must be evaluated from an analysis of the following properties: - Demonstration of the existence and characterization of a radical species induced by radiation - Discrimination between the radiation induced radical species and other radicals native or induced by other factors - Linearity of the dose response and detection limit - Thermal stability of the EPR radiation induced signal - Effect of light and UV on the EPR spectrum - Invasiveness - Individual variability of radiation sensitivity - Photon energy dependence - Dependence of response to radiation type - Easy availability - Influence of sample preparation, storage (before and after preparation) and environmental parameters

The most established materials in EPR dosimetry are the calcified biological tissues, i.e. bone, dentine and tooth enamel. Other materials that have attracted the interest of scientists are fingernails and hair (as biological materials), sugars, clothings, and building materials.

Tooth enamel The radiation induced radicals are formed is the carbonate present in the hydroxyapatite molecule. Several carbonate radicals have been identified,

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with the most stable and sensitive to radiation being the CO2-, whose EPR signal is then used as radiation indicator. A native signal interferes with the carbonate radical signal. The best property of tooth enamel is its stability in time, that is of the order of decades. For this reason the method is elective for prolonged past exposures that offer sufficient time to collect extracted teeth. Measurements are performed in vitro, i.e. on extracted teeth available in tissue banks. Dose response is linear between 100 mGy and 1 kGy. Detection limit is around 100 mGy, although it strongly depends on the sample preparation method and on the quality of the instrumentation. Effect of UV light, individual sensitivity, dependence of EPR response with radiation type and influence of sample preparation and environmental parameters have been widely investigated [1, 2, 3 and references therein]. In order to bypass the need of tooth extraction, two alternative methods are under analysis. The first is the EPR measurements performed at low microwave frequencies (L-band) that allow use of large samples. With this technology in vivo measurements can be performed. The second method is an in vitro measurement of small biopsies of tooth enamel. Both these methods are attractive for fast and large scale screening, especially the former method. However performances of these two methods are worse than those of the conventional method. In particular detection limit is of the order of a few Gy [4, 5 and references therein].

Fingernails Fingernails and toenails show an EPR signal that appear to be produced in the molecule of α-keratin. Interference signals exist, both native and induced by fingernail cutting. Preliminary results have shown a detection limit of 2 Gy. Because of the low time stability the sample mus be collected within a few hours after the exposure. The EPR measurements can be performed after days if the sample is stored at liquid nitrogen temperature after collection. The main practical advantage is its wide availability and easiness of collection [4, and references therein].

Hair Hair is also an interesting material, but the presence of interfering signals and a low time stability of the EPR signal make it less suitable than fingernails. Fundamental understanding of the EPR spectrum could improve the feasibility of its use as a dosimeter material.

Clothing materials The presence of keratin in wool and silk, although in a form different from that of fingernails, has motivated studies on these materials. Detection limit of 3 Gy has been evaluated in cotton [6]. Wool has been investigated only at doses in the kGy range. Fading is a limitation for both the materials [7].

Sugar Other than in the form of granular table sugar, sucrose is indeed also found in sweeties and pharmaceutical coverings. Because of the many radiation induced radicals, time stability and dose response linearity depend on the dose range. However the detection limit has been calculated around 100 mGy and operative protocols have been developed for a fast and accurate dose estimate. Radiation energy dependence has also been studied [8 and references therein].

Building materials In window glass, the paramagnetic centre induced by radiation is probably an oxygen hole centre. No interfering signals are present. Detection limit is about 5 Gy, above which linearity of dose response holds up to the level of kGy. The signal decays of about 50% in about 50 days. No effect of light and no radiation energy dependence has been detected [9]. Quartz is present in many natural building materials (ceramics, bricks, etc.) and it is one of the top materials for retrospective dosimetry with TL and OSL. EPR response is linear between 1 Gy and 103 Gy. The detection limit of 1 Gy was obtained for 100 mg of pure quartz so it is expected to be higher when quartz is dispersed in the material as happens i.e. in ceramics. [10].

CONCLUSIONS - Tooth enamel has the most appropriate characteristics as a dosimeter (stable, sensitive, etc.), but its use is invasive. Appropriate only for wide populations chronically exposed, not for acute exposures. Development of in vivo EPR measurement of teeth might be advantageous for the acute exposures. - Fingernails are suitable for short delay after the fact. - In most materials, time and thermal instability are the most limiting factors. Protocols for collection and storage of samples must be developed in order to reduce fading. - There is a need for investigation of other materials. The scope is not to find the best material, but rather to have a selection of materials, for a case dependent choice.

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