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Identification of red pepper powder irradiated with different types of radiation using luminescence methods: A comparative study

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ABSTRACT

The purpose of this study was to verify the reliability of photostimulated luminescence (PSL) and thermoluminescence (TL) methods for identifying irradiated foods, described in the European standards EN 13751:2002 and EN 1788:2001, respectively, which were established solely through interlaboratory studies on gamma-irradiated food. Red pepper powder samples irradiated with electron-beams (e-beams), gamma rays and high-energy X-rays were used as model foods. Samples irradiated with each radiation type at ⩾4 kGy could be correctly identified by the PSL method, whereas samples irradiated at ⩾0.5 kGy with each radiation type could be correctly recognized by the TL method when e-beams, gamma rays, or high-energy X-rays were used as normalization sources. However, different TL intensities were observed for minerals separated from red pepper powder for different irradiation sources, which was confirmed using pure quartz and K-feldspar minerals. Further interlaboratory studies are required to verify this phenomenon.

1. Introduction

Red pepper powder (*Capsicum annuum* L.) is one of the most widely used spices in Asian countries. Red pepper powder is generally contaminated with microorganisms, which can cause food spoilage and foodborne illness outbreaks, during the cultivation, drying, grinding, and storage processes (Banerjee & Sarkar, 2003). Decontamination processes, including fumigation with ethylene oxide, steam heat sterilization and irradiation, have been used to eliminate such contaminants. Among these treatment procedures, irradiation is most suitable, because steam heat treatment is limited to decontamination steps prior to packaging and ethylene oxide fumigation is restricted in many countries, owing to the possibility of toxic residues (Diehl, 2002; Farkas, 1998; Farkas & Andrassy, 1998).

Irradiation technology is predominantly used to inactivate microorganisms in dried vegetables and spices (Arvanitoyannis & Tserkezou, 2010). Food can be treated with three types of radiation, namely gamma rays from radioisotopes, machine-generated electron-beams (e-beams; maximum energy of 10 MeV), and X-rays generated by using electrons less than 5 MeV (7.5 MeV in the US). The three types of radiation exhibit different characteristics depending on their energy level and transferring material. Gamma rays and X-rays are types of photon-based radiation in the electromagnetic spectrum (Farkas & Farkas, 2011). Both of these types of radiation are in the short-wavelength, high-energy region of the spectrum and can penetrate foods to depths of several centimeters. However, the energies from gamma rays are 1.17 and 1.33 MeV from ^{60}Co and 0.66 MeV from ^{137}Cs, respectively, whereas X-ray is bremsstrahlung with a wide energy spectrum ranging from several keV up to the electron-beam energy (Cleland & Stichelbaut, 2013; Seltzer, Farrel, & Silverman, 1983). E-beams and X-rays are generated by an electron accelerator, such as a linear accelerator. E-beams with a maximum energy of 10 MeV are particulate radiation and, hence, have a cross-section many times greater than photons, as a result of which they do not penetrate the product beyond a few inches, depending on the product density (Fan & Sommers, 2013). In our previous study, e-beam irradiation was found to be more efficient at inactivating the total aerobic bacteria in red pepper powder compared to gamma and X-ray irradiation. Additionally, none of the tested radiation types changed the quality properties of the red pepper powder (Jung et al.,

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2015). However, the detection properties of red pepper powder irradiated with different types of radiation have not been compared.

To identify irradiated red pepper powder, photostimulated luminescence (PSL) and thermoluminescence (TL) methods described in the European standards EN 13751:2002 and EN 1788:2001 are used internationally as screening and reference methods, respectively (Bayram & Delincee, 2004; Chauhan, Kumar, Nadanasabapathy, & Bawa, 2009; Sanderson, Slater, & Cairns, 1989). However, both of the standards were established solely through interlaboratory studies on gamma-irradiated foods followed by re-irradiation with gamma rays at 1 kGy. Therefore, the acceptability of the TL method for the detection of food samples irradiated with X-rays or e-beams needs to be confirmed. Moreover, the silicate minerals separated from a food sample should be re-irradiated at a defined dose after the TL measurement (TL_1), because the content of compounds in the silicate minerals affecting the TL intensity is immeasurable. This re-irradiation step is called "normalization" and the TL intensity of the normalized minerals is known as the second TL intensity (TL_2). In all the reference studies used in EN 1788:2001, the mineral samples are normalized with gamma rays at 1 kGy except for potatoes, resulting in a TL ratio (TL_1/TL_2) criterion of >0.1 for positive identification of irradiation treatment. However, the use of e-beam and X-ray irradiation for normalization is not mentioned in the standard.

Therefore, in this study, we investigated the detection properties of red pepper powder irradiated with three different types of radiation that can be used commercially for the sterilization of food, by using the PSL and TL detection methods. Moreover, the possibility of using e-beams, high-energy X-rays, and low-energy X-rays, which are more convenient to use than gamma irradiation, in the normalization step was examined.

2. Materials and methods

2.1. Sample preparation and XRD analysis of pure minerals

Red pepper powder was purchased from the local market in Jeongeup, Korea. The samples (200 g) were placed in sterilized oxygen-impermeable nylon polyethylene/polypropylene bags (size: 20 × 30 cm, thickness: 0.07 mm; Sunkyung Co. Ltd., Seoul, Korea) and packaged to a thickness of 2.0 cm to minimize the variation in penetration depth among the radiation sources. Three packages of the sample were prepared for each irradiation dose of a designated radiation type.

Pure quartz and K-feldspar powders were provided by Risø National Laboratory, Technical University of Denmark (Roskilde, Denmark). Each mineral powder sample was sieved using a nylon cloth with a pore size of 125 μm. Aliquots of 2.0 mg were placed on aluminum discs (diameter: 6 mm) and fixed with silicon. Five discs of each pure mineral sample were stuck in a disposable petri dish with a diameter of 90 mm using double-sided tape and irradiated at a designated dose with different types of radiation.

To examine the purity of the minerals, the quartz and K-feldspar powders (0.5 g) were analyzed using a multipurpose X-ray diffractometer (XRD, X'Pert Pro; PANalytical, Almedo, The Netherlands) at the Korea Basic Science Institute (Daegu, Korea). Silicon powder (corundum) was used as a standard reference material to calibrate the X-ray diffractometer. The analysis was performed under the following measurement conditions: detector: X' Celerator (ultrafast); scan angle: 0–60°; scan rate: 11.9 °/s; scan axis: Gonio; scan mode: continuous; radiation: Cu Kα with a wavelength of 1.540598 Å. The XRD spectral data was collected in the 2°θ range of 0°θ and 60°θ and each peak was identified by comparing with reference data.

2.2. Irradiation of the samples

The red pepper powder samples were treated with targeted irradiation doses of 0.5, 1.0, 2.0, 4.0, 6.0, 8.0, or 10.0 kGy, whereas the pure quartz and K-feldspar powders were irradiated at different doses ranging from 0.2 to 1.0 kGy using one of the three radiation types. Gamma irradiation was performed with a ^{60}Co gamma irradiator (point source AECL, IR-79, MDS Nordion International Co. Ltd., Ottawa, ON, Canada) at the Advanced Radiation Technology Institute, Jeongeup, Korea. The source strength was approximately 11.1 PBq and the photon energies of gamma rays from ^{60}Co were 1.17 and 1.33 MeV. E-beam and high-energy X-ray irradiations were conducted with a linear electron accelerator, which delivers e-beams with a well-defined energy in the range of 5–10 MeV, at EB-Tech Co. (Daejeon, Korea) at an ambient temperature of approximately 20 °C. E-beams with an energy of 10 MeV were employed for e-beam irradiation of the samples, whereas X-rays were produced with 7.5 MeV e-beams. The applied dose rates were 8 MGy/h under e-beam irradiation, 24 kGy/h under X-ray irradiation, and 1 kGy/h under gamma irradiation. Geometrically, all the samples were irradiated in a direction perpendicular to the incident radiation.

2.3. PSL measurements

PSL of the non-irradiated control and irradiated red pepper powder samples were measured 24 h after irradiation, as described by EN 13751 (2009) using a SURRC Pulsed PSL system (Scottish Universities Research and Reactor Centre, Glasgow, UK), which comprised of a control unit, sample chamber, and detector. Briefly, 5 g of sample (less than 50 mesh) was placed in a disposable petri dish (Bibby Sterilin type 122, Glasgow, UK) with a diameter of 50 mm. All of the preparation, handling, measurement, and storage operations of the samples were carried out under subdued ambient light. The PSL signals (expressed as photon counts, PCs) were recorded in the measuring mode and were expressed as PCs/60 s. The accumulated PCs were interpreted with respect to two thresholds, namely the lower threshold (T_1 = 700 counts/60 s) and upper threshold (T_2 = 5000 counts/60 s). Samples exhibiting a signal less than T_1 were classified as non-irradiated samples, whereas those with a signal greater than T_2 were deemed to be irradiated samples. Samples with signal levels between the two thresholds were classified as intermediate samples requiring further investigation.

2.4. TL analysis

The TL signal intensities of the quartz, K-feldspar, and mineral samples separated from red pepper powder irradiated with three different types of radiation were analyzed by a TLD system instrument (Harshaw TLD-4500, Dreieich, Germany) under nitrogen flush (99.99%). For separating the minerals from red pepper powder, about 100 g of each sample was used to collect an aliquot of silicates following the procedure prescribed in the CEN Standard EN 1788 (2001). The isolated minerals were carefully deposited onto clean aluminum discs (diameter: 6 mm) and kept overnight at 50 °C in a laboratory oven. The discs were then heated from 50 to 350 °C at a scan rate of 5 °C/s. After the first glow (TL_1) measurement, the samples were annealed at 350 °C for 5 s to completely remove the TL characteristics. To normalize the TL response, the second glow (TL_2) was obtained from the TL_1 tested minerals that were re-irradiated with e-beams, gamma rays, and low or high-energy X-rays at a dose of 1 kGy. Re-irradiation of the discs with e-beams, gamma rays, and high-energy X-rays was performed under conditions that were identical to the first irradiation, whereas low-energy X-ray irradiation was carried out using a cabinet type X-ray machine (CP-160, Faxitron X-ray LLC,