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Thermoluminescence parameters and kinetics of irradiated inorganic dust collected from black peppers

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Abstract

Irradiated foods can be detected by thermoluminescence (TL) of contaminating inorganic dust particles. In this study, black peppers were irradiated with gamma rays at doses of 1, 3, 5 and 10 kGy. The inorganic dust particles collected from irradiated black pepper are investigated. This study reports the mineral composition of this dust being, mainly quartz, feldspar and little amount clay minerals. The TL detection method is clearly able to distinguish irradiated and non-irradiated black peppers at the doses ranging from 1 to 10 kGy. The paper provides a detailed calculation of the activation energy (E), frequency factor (s) and the order of kinetics of the 240 °C TL identification peak in polymineral dust material. The experimental and computerized deconvolution results are consistent with the presence of a closely overlapping second-order TL peaks in the identification peak at 240 °C. The TL kinetic parameters of overlapping peaks were estimated by computerized deconvolution method. The computer deconvolution shows the existence of four well-defined TL peaks at 118, 210, 270 and 305 °C. These four peaks probably due to the combined TL of quartz and feldspar minerals.

Keywords: Detection method; Food irradiation; Thermoluminescence; Black pepper

1. Introduction

To use of radiation to preserve foods and reduce the incidences of food-borne diseases can provide great benefits in the food industry. Through the irradiation process it is possible to sterilize, thus extending the shelf-life by reducing spoilage caused by organisms, inhibit sprouting, destroy insects and parasites, and delay the ripening process (IAEA, 2002; Rahman, Haque, & Sumar, 1995). Many countries started research to develop convenient methods and standard procedures for determining whether or not a particular food product has been irradiated with ionizing radiation. Significant progress has been made in the development of detection methods of irradiated foods. Detection methods for irradiated foods may be classified in three basic categories as chemical, physical and biological (Schreiber, Helle, & Bögl, 1993).

Thermoluminescence (TL) is one of the most important physical techniques for spices, herbs and dried fruits (Beneitez, Correcher, Millán, & Calderón, 1994; EN 1788, 2001; Göksu, Regulla, Hietel, and Popp, 1990; Khan, Bhatti, & Delincée, 2002). Thermoluminescence is the emission of light upon release of captured charges from their traps by heating. Quantitative measurement of the emitted TL light following irradiation is an established method in radiation dosimetry (McKeever, Moscovitch, & Townsend, 1995), and has been introduced by Bögl and coworkers for the identification of irradiated food (Heide & Bögl, 1987).

Extensive investigations have shown TL measurements to be suitable method for the rapid identification of irradiated dried food (Bögl & Heide, 1984; Heide & Bögl, 1986, 1988). It has been found that the origin of this TL signal in food samples mainly lies in the insoluble mineral and dust particles concomitant to food items and not in the organic matrix (Göksu et al., 1990; Sanderson, 1990). By separating these minerals from the food samples, such as herbs and spices, and performing TL analysis on these minerals alone

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produce much better results (Engin, 2004; Göksu-Ögelman & Regulla, 1989; Göksu et al., 1990). It appears not possible to relate the TL emission of the presence to a particular mineral. Such dust samples consist commonly of quartz, feldspars, different kinds of clay minerals and carbonates (Beneitez et al., 1994; Calderón et al., 1995; Calderón et al., 1994; Sanderson, Slater, & Cairns, 1989). The character (i.e., shape, emission wavelength, intensity) of a TL glow curve is critically dependent upon both the nature of the phosphor and the defects within it. The differences in the TL signal shapes for the different minerals do not change the validity of the detection tests for irradiated spices (Chabane, Pouliquen-Sonaglia, & Raffi, 2001). The intensity of TL glow peaks at the same gamma doses for inorganic dust particles collected from black peppers vary considerably from black pepper to black pepper and package to package (Engin, 2004; Mamoon, Abdul-Fattah, & Abulfaraj, 1994). In spite of the polymineral structure of inorganic dust particles, spices, herbs and dried fruits can be analysed successfully for previous radiation treatment by measuring first TL1 response of the adhering inorganic dust particles (Engin, 2004; Khan & Bhatti, 1999; Khan et al., 2002). This is true if we know the TL signal area of the unirradiated sample. Hence, in practice, where we do not know this unirradiated area, a clear identification needs a normalization step (Khan & Bhatti, 1999). For detection, it is a necessary prerequisite that adequate amounts of minerals can be isolated from the foodstuff. Signal intensities of polymineral inorganic dusts depend on irradiation dose, type and mass of minerals. Although the investigated inorganic dust particles are not typical for the dust on black peppers or another spices and herbs, it could be useful to determination of the TL glow peak structure and kinetic parameters of the inorganic dust particles collected from the irradiated black peppers. Because, so far, these type inorganic polymineral dust samples were used for identification of irradiated spices, herbs and all dried fruits with TL technique.

The purpose of the present study was to determine the glow curve structure, kinetics and trapping parameters of TL in inorganic dust particles collected from irradiated black pepper.

2. Experimental

Black peppers (*Piper nigrum*) were bought at random from the local market in Ankara (Turkey) and without selecting a specific producer, and used without any pretreatment. Different packages of black peppers were used in the experiments. The origin of the samples is unknown.

For TL analysis adequate amounts of minerals need to be isolated from the foods. For all the black peppers investigated, enough mineral grains could be isolated by the method described in the work of Göksu-Ögelman and Regulla (1989), Sanderson et al. (1989), Schreiber, Hoffmann, Helle, and Bögl (1994) and EN 1788 (2001): For separation of adhering inorganic dust particles from black pepper samples, about 400 g sample was mixed with water and after ultrasonic bath for 30 min and centrifugal treatments, a density gradient of polytungstate solution (density 1.7 g/cm^3) [$Na_6W_{12}O_{39}\cdot H_2O$] was used to separate remaining organic matrix from the minerals. Then, samples were washed with distilled water, dried with acetone and stored overnight at room temperature (25 °C) until further processed. The isolated minerals were deposited carefully on a very clean aluminium discs (5 mm radius, 0.5 mm thickness). Dust particles between 2 and 10 μm diameter subjected to the above treatment were used for the TL measurements. The irradiation of the samples was performed using a ^{60}Co gamma source with a dose rate of 4 kGy/h (irradiation unit "Isledovatel", former USSR at the Sarayköy Establishment of Turkish Atomic Energy Agency in Ankara). The preparation and irradiation of samples took place at ambient temperature. A standard gamma doses of 1, 3, 5 and 10 kGy (commercial irradiation dose range) were used for irradiations. The absorbed dose was checked Fricke dosimetry. After irradiation black pepper samples were stored at room temperature. Extra care was taken to keep all dust samples in the dark until the TL measurements were performed.

Measurements were performed with a TL reading system of a Pitman Model 654 TL Reader. The amount of sample for each experimental run was optimized at 10 mg. The collected sample material was put onto an aluminium disc fitting to the planchet of the TL reader. A linear heating rate of about $\beta = 1$ °C/s was used for the recording of glow curves. The heating rate of 10 °C/s was used for the isothermal decay experiment. This heating rate value is closer to the EN 1788 (2001) protocol conditions. The heating chamber of the TL reader was flushed with nitrogen and the sensitivity of photomultiplier tube was checked with a Carbon-14 light source. The TL curves were recorded immediately after the irradiation so that there was no significant fading of the peaks at room temperature. All measurements were carried out under red light illumination to avoid any drainage of TL due to white light. All data points are the average of at least five different aliquots of the sample. An electric furnace was used to maintain constant temperature (120 °C and 170 °C), for isothermal decay studies. The temperature was monitored with a Chromel–Alumel thermocouple; temperature control was within 5 °C from the desired setting.

3. Results and discussion

One type of spice was checked in the present study. The inorganic dust particles collected from irradiated black pepper are investigated. Chemical analyse by X-ray diffraction was done on inorganic dust particles separated from black pepper. This analyse confirms that dust particles have the polymineral structure. The results of the chemical analyse are shown in Table 1. Chemical analysis of the material indicated the presence of quartz, feldspars and clay minerals in all of the dust samples studied. These results confirm earlier findings (Beneitez et al., 1994; Calderón et al., 1995,