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Thermoluminescence characteristics of the irradiated minerals extracted from red pepper (*Capsicum annum L.*) spice

J. Marcazzó^{a,b}, E. Cruz-Zaragoza^{a,*}, L. Montiel^a, V. Chernov^c, T. Calderón^d

^a*Instituto de Ciencias Nucleares, Universidad Nacional Autónoma de México, A.P. 70-543, 04510 México D.F., México.*

^b*Instituto de Física Arroyo Seco, Universidad Nacional del Centro de la Provincia de Buenos Aires, Pinto 399, 7000 Tandil, Argentina.*

^c*Centro de Investigación en Física, Universidad de Sonora, A.P. 5-088, Hermosillo, Sonora, 83190 México.*

^d*Departamento de Química Agrícola, Facultad de Ciencias, Universidad Autónoma de Madrid, 28049-Cantoblanco, Madrid, España.*

*Corresponding author: Tel.: +52 55 56224683; Fax: +52 55 56162233. E-mail: ecruz@nucleares.unam.mx

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The inorganic polymineral content in the foodstuffs allows to analyze the main thermoluminescence (TL) characteristics that may be useful in the identification of irradiated food. The mineral fraction was separated from commercial Mexican red pepper (*Capsicum annum L.*). X-ray diffraction shows that the mineral composition of the samples was mainly quartz. From the mineral fraction of different grain sizes, samples of 149 μm were selected for this study because of the high TL signals. The samples were irradiated from 1 to 500 Gy by using a ^{60}Co irradiator. The TL characteristics like glow curve shape, dose-response, UV and sunlight bleaching and fading were analyzed. The glow curves show an intense TL peak at 82 °C followed by others with less intensity at 130, 170 and 340 °C. The T_M - T_{STOP} method shows six TL glow peaks that was taken into account for calculation the activation energies values. Because the complex structure of the glow curves, the kinetics parameters were determined by using a computerized deconvolution program assuming the general order kinetics model.

Keywords: Minerals; Red pepper; irradiated foodstuffs; thermoluminescence; glow curve structure.

La fracción inorgánica polimineral que está presente en los alimentos, permite analizar características termoluminiscentes (TL) que pueden ser útiles para la identificación de alimentos irradiados. La fracción mineral fue separada de muestras de pimentón rojo (*Capsicum annum L.*) de tipo comercial y de origen mexicano. La difracción de rayos-X mostró que la composición mineral es esencialmente cuarzo. Se seleccionaron distintos tamaños de grano y se eligieron aquellas muestras de 149 μm debido a su intensa señal TL. Las muestras fueron irradiadas desde 1 hasta 500 Gy utilizando un irradiador de ^{60}Co . Se analizaron las propiedades TL tales como la estructura de la curva de brillo, la relación dosis-respuesta, la disminución de las señales TL debido al blanqueo de las muestras a la luz solar y ultravioleta (UV) y durante el almacenamiento de las muestras a temperatura ambiente o fading. Las curvas de brillo mostraron un pico TL intenso en 82 °C, seguido de otros picos con menor intensidad en 130, 170 y 340 °C. El método T_M - T_{STOP} mostró que las curvas de brillo estaban compuestas de seis picos TL, los cuales se consideraron para el cálculo de los valores de las energías de activación. Debido a la estructura compleja de las curvas de brillo, los parámetros cinéticos se calcularon utilizando una deconvolución mediante un programa de cómputo considerando el modelo de cinética de orden general.

Descriptores: Minerales; Pimentón rojo; alimentos irradiados; termoluminiscencia; estructura de curvas de brillo.

PACS: 78.60.Kn; 61.80.Ba; 78.55.Hx

1. Introduction

It is well known that the exposure of food to ionizing radiation is an effective technological process used in many countries in order to eliminate pathogen microorganisms, to eradicate pests, and to extend the shelf-life of the foodstuffs. Food processed by ionizing irradiation has been recognized by the Codex Alimentarius Commission as a safe technology [1-3]. It is an effective method to prevent food spoilage and food-borne diseases by inhibiting the growth of microorganisms in food. Also the irradiation processing is an alternative to the chemical gases fumigation with ethylene oxide and methyl bromide which may result dangerous for humans and environment [4]. Food irradiation treatments normally include spices, dry vegetables, grains and fruits for preservation and sterilization too [5-7] which are exposed to different doses

radiation by the ^{60}Co commercial irradiators in the World [3, 8, 9]. However, the irradiation processing makes relevant the developing and assessment of physical methods for identification of irradiated foodstuffs. The physical, chemical and biological methods are frequently used to discriminate between irradiated and non-irradiated food [6, 10-12]. Among of those detection methods, the analysis of the thermoluminescence (TL) emission seems to be a good physical method due to the high precision [11-13]. TL is associated with contaminating polyminerals which are present on all herbs and spices which have been exposed to wind, water and soil. The minerals adhere only to the surface of the foodstuff. In many kind of foodstuffs the inorganic polymineral fraction normally is composed by quartz and feldspars minerals [14-18]. The TL phenomenon is based on the fact that the

inorganic mineral fraction emits light when is heated after exposure to ionizing radiation. During irradiation of the quartz or feldspars, the electron-holes pairs defects are generated in the lattice of the mineral and they can be trapped in localized states in the band gap. These point defects can remain trapped according to the depth energy concerned to the trapping levels distribution and environment temperature. When the temperature increases, the charge defects are free and a radiative recombine may be occurred emitting visible light. Then the thermally stimulated emission of light is obtained on the function of temperature that is named glow curve. In general, the luminescence emitted as TL response by the mineral fraction is usually proportional to the dose absorbed by the sample. However, the glow curve's structure and TL response of the samples depends on the mineral composition in each different foodstuff [16, 19-24]. Therefore, it is important to analyze the TL characteristics and the glow curve's structure of the mineral fraction in the commercial foodstuffs that can be useful for identification purposes of irradiated food. This work focuses on the TL properties of red pepper spice irradiated at different gamma doses and exposed to different light environment. The aim of this paper is to analyze the main TL characteristics of polyminerals separated from commercial Mexican red pepper. In particular, the TL dose-response, the UV and sunlight bleaching and fading results as well as the glow curve structure were analyzed. The T_M-T_{STOP} method [25] was used to identify separated peaks and their temperatures of maxima on TL glow curves. The activation energies and the frequency factors ascribed to the trap distribution of the mineral from red pepper were evaluated by using the general order kinetics (GOK) model [26, 27]

2. Experimental

The samples of red pepper (*Capsicum annum L.*) were obtained from the commercial batch in Mexico. The polymineral fraction was separated from organic part of the samples following by a centrifugation procedure. Initially, 25 g of the whole red pepper sample were mixed with an ethanol-water (70:30) solution and kept in constant agitation and centrifugation during 24 hours and 2 hours respectively, in order to separate the inorganic fraction. This procedure was repeated for ten batches with the same mass of red pepper samples. The collected polyminerals were washed with bidistilled water and followed by hydrogen peroxide to remove all residual organic matter. The polymineral powder was dried with acetone at room temperature (RT). All samples were kept in darkness at RT before and after irradiation. Four polymineral grain sizes 10, 53, 74 and 149 μm were selected. Approximately 4 mg of powder of each grain size was then deposited onto aluminum disks followed by an acetone drop for obtain a homogeneous grain deposition. Several sets with 20 disks samples were prepared with this procedure for irradiation and TL measurements. Because of the high TL signal the samples with 149 μm grain size were selected in this study. The mineral composition of the samples was determined by X-Ray

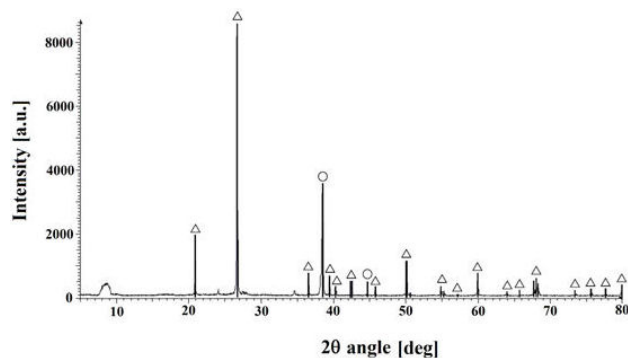


FIGURE 1. XRD spectrum of Mexican red pepper. The shifted peaks (triangles) are related to the quartz.

Diffraction (XRD) by using a Siemens D5000 diffractometer, equipped with a Cu-Ni filter. The XRD patterns of the samples were compared to the ASTM (American Standard of Test Materials) X-Ray Powder Diffraction data files. Figure 1 shows that the inorganic fraction is mainly composed by quartz (SiO_2). The SiO_2 is normally present in herbs, spices and condiments [17, 18, 20-24].

All samples were irradiated using a ^{60}Co gamma-source (Gammacell 200) giving a dose rate of 0.5 Gy/min determined by Frick solution dosimeter [28]. A double series of 30 disks samples, with 4 mg each one, were also exposed to ultraviolet (UV) and sunlight bleaching. An Hg lamp source model OS-9286 from Pasco Scientific Company, which provides a light beam irradiance of 0.1 $\mu\text{W}/\text{cm}^2$ at the sample position, was used to make the UV bleaching measurements. For the sunlight bleaching the samples were exposed directly to the sunlight beam and the room temperature was about 21-23 $^\circ\text{C}$. All the TL glow curves were recorded from RT (22 $^\circ\text{C}$) up to 400 $^\circ\text{C}$ with a linear heating rate of 2 $^\circ\text{C}/\text{s}$ by using a Harshaw 3500 TL reader. The heating planchet and sample in the TL reader were maintains in a continuous nitrogen flux to reduce spurious TL signals.

3. Results and discussion

Four different grain sizes were selected, i.e. 10, 53, 74 and 149 μm , from the inorganic fraction extracted of Mexican red pepper. An average of 67 mg of polyminerals was obtained from each 25 g of the whole red pepper samples. It was observed that the TL intensity increases significantly with the grain size. The most efficient grain size was 149 μm , and was chosen for the TL analysis. Figure 2 shows the TL glow curves of the polyminerals extracted from red pepper that was irradiated at different gamma doses. The glow curves show an intense TL glow peak at 82 $^\circ\text{C}$ followed by others peaks at about 130, 170 and 340 $^\circ\text{C}$. These peaks are related to the quartz present in the mineral fraction. The shape of the glow curves was similar to those reported previously by different authors for the quartz extracted from tiles or rocks [29, 30]. The TL signals as a function of the dose is shown in the inset of the Fig. 2 where a linearity behavior was observed in the

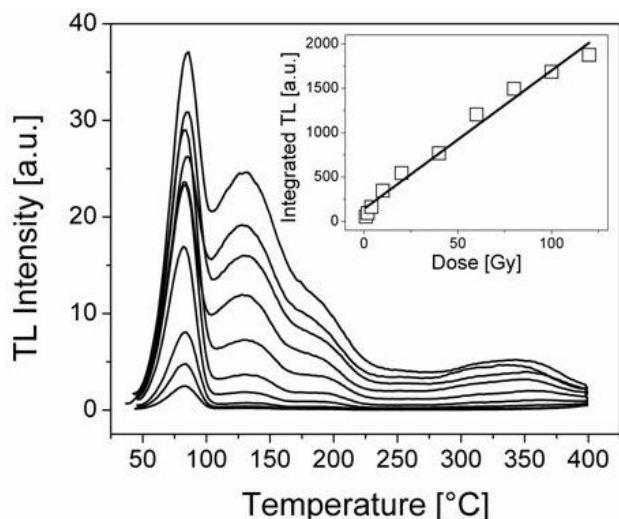


FIGURE 2. TL glow curves of the polyminerals expose to different doses, namely 120, 100, 80, 60, 40, 20, 10, 4, 2 and 1 Gy, from top to bottom. In the inset: integrated TL response as a function of the gamma doses (1-120 Gy).

range from 1 up to 120 Gy. It was observed that the TL response increases as the dose increases up to 500 Gy. This is an important result because no saturation effect was present in the dose range analyzed. Furthermore, the reproducibility of the TL response of the inorganic fraction after successive irradiation-readout cycles at two different doses (1 and 30 Gy), was about 3% of the standard deviation.

The influence of ultraviolet (UV) light and sunlight on TL glow curves of polymineral samples was analyzed. Samples were irradiated with a dose of 30 Gy and then exposed to UV light for different periods of time. Figure 3 shows the TL response, integrated under the glow curve, as a function of UV exposure time. The integrated TL signal decreases as a double exponential function during UV irradiation. This is because during the first 10 minutes the most important decrease is observed for peak 1 (82 °C) which is the most intense peak in the glow curve. After that the other peaks located at higher temperature side of the glow curves continue to decrease uniformly. The influence of the sunlight on the TL glow curves is similar to the results obtained with UV bleaching. The TL response decreases as a double exponential function (Figure 3). A comparison the effect of light bleaching on TL response relative to the response of the samples in darkness and without light, i.e. fading measurements were carried out too. The TL fading behavior is shown in the inset of Fig. 3. Samples were irradiated with 3 Gy and stored in darkness at RT for different periods of time. It is seems that the TL signal decreases as an exponential function and the TL response, was approximately 60% of the original value during the first month.

Because the complex of the glow curve's structure the T_M - T_{STOP} method [25] was used in order to determine the number of peaks and their temperature presents in the glow curve. Figure 4 shows the T_M - T_{STOP} relationship by using the steps of 7 °C, and 2 °C for refining intervals of the glow

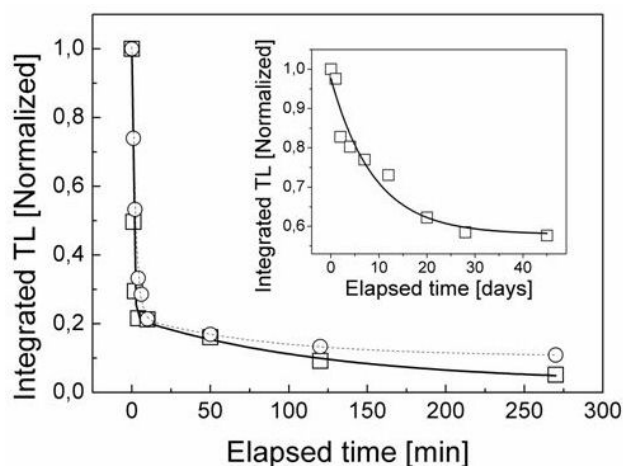


FIGURE 3. TL response as a function of exposure time to UV light (open circles) and sunlight (open squares). In the inset: TL fading of samples stored in darkness at RT.

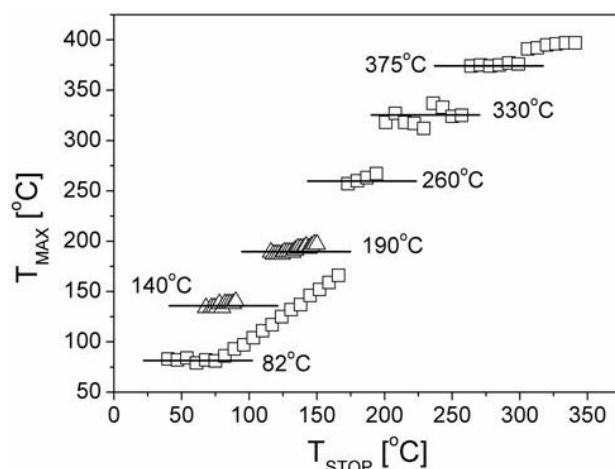


FIGURE 4. T_M - T_{STOP} measurements by using steps of 7°C (open squares) and 2°C (open triangles) for refining intervals of the data.

curve between 25-350 °C. Using the T_M - T_{STOP} information it was followed by a deconvolution procedure in order to obtain the kinetics parameters of the glow peaks. The glow curve deconvolution was carried out by assuming a General Order Kinetic (GOK) model [26] modified by Rasheedy [27]. The equation of the intensity includes the ratio n_0/N which takes into account the fraction of occupied traps. Then, the equation describing TL intensity is given by [27]:

$$I = n_0^b s \exp\left(\frac{-E}{kT}\right) / N^{b-1} \left[1 + \frac{s(b-1)(n_0/N)^{(b-1)}}{R} \times \int_{T_0}^T \exp\left(\frac{-E}{kT'}\right) dT' \right]^{1/(b-1)}$$

where n_0 stand for the initial concentration of electrons in traps, N stand for the corresponding concentrations of traps and R is the heating rate. Besides, E is the activation energy

TABLE I. Activation energies values obtained by using Initial Rise (IR) method and kinetic parameters values obtained by general order kinetics model by using GCD deconvolution procedure for glow curves at 120 Gy and 20 Gy doses.

	$T_M [^{\circ}\text{C}]$	E [eV] (IR)	E [eV] (GCD)	s [s^{-1}]	b	n_0/N	FOM
Dose 120 Gy							1.2%
Peak 1	82	1.00 ± 0.02	1.01	4.5×10^{13}	1.11	7.8×10^{-2}	
Peak 2	114	0.80 ± 0.02	0.85	4.0×10^{13}	1.83	7.3×10^{-5}	
Peak 3	140	0.84 ± 0.02	0.83	6.4×10^9	1.16	6.4×10^{-5}	
Peak 4	182	0.90 ± 0.05	0.88	8.9×10^9	1.45	1.6×10^{-3}	
Peak 5	243	0.93 ± 0.08	0.90	1.3×10^{11}	1.96	2.6×10^{-4}	
Peak 6	342	0.90 ± 0.12	0.93	2.0×10^9	1.90	6.2×10^{-4}	
Dose 20 Gy							1.6%
Peak 1	82	1.01 ± 0.02	1.01	4.3×10^{13}	1.08	5.7×10^{-2}	
Peak 2	118	0.80 ± 0.02	0.85	7.8×10^{13}	1.81	1.5×10^{-5}	
Peak 3	142	0.84 ± 0.02	0.88	6.2×10^9	1.02	5.3×10^{-6}	
Peak 4	186	0.90 ± 0.05	0.88	1.4×10^{10}	1.40	2.4×10^{-4}	
Peak 5	268	0.93 ± 0.08	0.85	1.4×10^{11}	2.00	4.2×10^{-5}	
Peak 6	368	0.90 ± 0.12	0.91	2.5×10^9	1.92	1.3×10^{-4}	

TABLE II. Kinetics parameters obtained by the general order kinetics model by using GCD deconvolution for the glow curve at 30 Gy and fading at 35 min after irradiation of the samples.

	$T_{max} [^{\circ}\text{C}]$	E [eV] (GCD)	s [s^{-1}]	b	n_0/N	FOM
Dose 30 Gy						1.2%
Peak 1	82	0.99	2.3×10^{13}	1.07	7.3×10^{-2}	
Peak 2	114	0.83	6.7×10^{13}	1.80	1.6×10^{-5}	
Peak 3	136	0.85	5.5×10^9	1.03	9.1×10^{-6}	
Peak 4	180	0.83	1.0×10^{10}	1.52	3.9×10^{-4}	
Peak 5	264	0.75	5.9×10^{10}	2.18	6.1×10^{-5}	
Peak 6	354	0.84	1.7×10^9	2.02	1.8×10^{-4}	
Dose 30 Gy Fading 35 min						1.3%
Peak 1	82	0.99	2.3×10^{13}	1.06	4.9×10^{-2}	
Peak 2	114	0.83	7.1×10^{13}	1.80	1.3×10^{-5}	
Peak 3	138	0.85	5.0×10^9	1.03	6.2×10^{-6}	
Peak 4	182	0.85	1.0×10^{10}	1.46	3.2×10^{-4}	
Peak 5	274	0.76	5.9×10^{10}	2.18	4.9×10^{-5}	
Peak 6	356	0.84	1.7×10^9	2.02	1.8×10^{-4}	

of the trap, s stand for the frequency factor, b the order of kinetic, k is the Boltzmann constant and T is the temperature in degrees Kelvin. The goodness of fit was evaluated by using the figure of merit (FOM) [31] which is given as:

$$FOM = 100 * \sum_{i=1}^m \frac{|I_{exp}(T_i) - I_{fit}(T_i)|}{A}$$

where $I_{exp}(T)$ and $I_{fit}(T)$ are the experimental and fitted glow curves, respectively; A is the area under curve $I_{exp}(T)$ and m is the number of experimental points. Figure 5 shows

that the structure of the glow curves by deconvolution is composed by six peaks along of the experimental glow curves at 120 Gy and 20 Gy doses. Table 1 shows the activation energy values obtained by Initial Rise (IR) method and the kinetics parameters using the glow curve deconvolution (GCD) procedure based on the GOK model [27]. The activation energy (E), calculated by IR method, has practically the same values (0.80-1.01 eV) for the glow curves obtained at two different doses (120 and 20 Gy). While using GCD computer calculation method the E values were similar for the glow curves at 120 Gy and 20 Gy, *i.e.*, 0.83-1.01 eV and 0.85-1.01 eV,

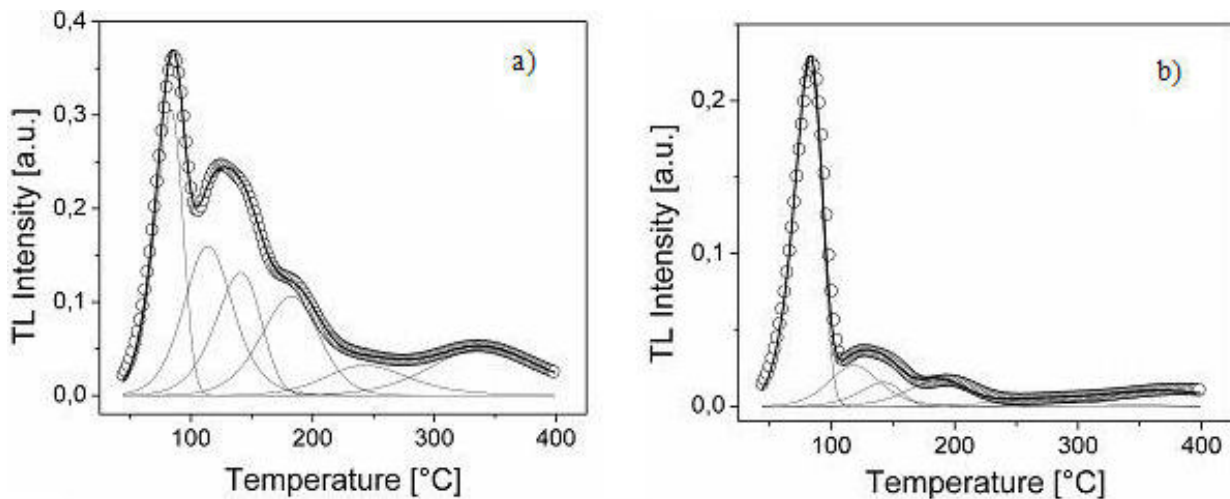


FIGURE 5. Glow curve's structure deconvolution by six TL peaks (thin solid lines) of the experimental TL glow curve (open circles) using GCD method. a) 120 Gy, b) 20 Gy. The thick solid line is the sum of all deconvoluted peaks.

respectively (Table I). The Figure Of Merit (FOM) values indicated that a very good fit was obtained by GCD deconvolution for the glow curves at 120 and 20 Gy doses, 1.2% and 1.6% of FOM, respectively. Furthermore, the development of the glow curve's structure during fading at 35 min after irradiation was investigated. The kinetics parameters values were similar between the glow curves at 30 Gy and after 35 min irradiation (Table II). The structure of the glow curves, *i.e.*, the peak numbers, their maximum temperatures, and the activation energy values was very similar and, no change during fading was observed.

4. Conclusion

The experimental glow curves from polymineral of Mexican red pepper, show a prominent peak at 82 °C followed by other peaks at 130, 17 and 34 °C respectively, which are related to the TL glow peaks characteristics of the quartz. Ultraviolet and sunlight bleaching measurements were carried out, and similar TL signal decreasing was observed. The TL response was found to be linear in the 1-120 Gy dose range and reproducible in spite of a strong TL fading observed. It is impor-

tant to mention that the glow peak located at 140 °C seems very stable, indicating to be a suitable TL peak for dose assessment in irradiated red pepper samples. The polimineral fraction shows a complex structure in its glow curves. The T_M-T_{STOP} method revealed the existence of a possible discrete traps distribution. A deconvolution program assuming the GOK model was used, and six glow peaks were determined for the glow curves. The activation energies values (0.80 - 1.01 eV) were similar to those obtained by Initial Rise Method. The similar kinetic parameters values obtained by different doses describe accurately the TL process.

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