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Observation of Several Detection Factors Derived from Thermoluminescence of Mineral Separated from Irradiated Korean Sesame and Perilla Seeds Stored under Different Storage Conditions

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Abstract

This study was carried out to observe changes in several detection factors derived from thermoluminescence (TL) of minerals separated from irradiated Korean perilla and sesame seeds during storage under normal room and darkroom conditions. The TL intensities of the first glow curves increased from 0 to 5 kGy but only slightly increase from 5 to 10 kGy. Maximum TL temperatures of the first glow curves in all irradiated samples were around 200°C, ranging from 150 to 250°C. Since the control (0 day of storage) glow curve ratios of G3 and G4, calculated from re-irradiated (1 kGy) sample were over 0.5, detection of irradiation was possible. However, because Gl ratios were below 0.1, they were classified as non-irradiated. There was a unique first glow curve shape that could be clearly seen in all irradiated samples, regardless of storage conditions, that was never seen in non-irradiated samples. In all samples, the maximum TL temperatures and shape of the second glow curve was in a lower temperature range than that of the first glow curve. Therefore, detection of irradiated Korean perilla and sesame seeds was possible for up to 3 months after irradiation, regardless of storage conditions, by examining several TL detection factors; including TL intensity, glow curve ratios, maximum TL temperatures, and the shapes of glow curves.

Key words: Korean perilla and sesame seeds, gamma irradiation, thermoluminescence (TL) characteristics

INTRODUCTION

Food irradiation is increasingly used as a hygienic process for reducing spoilage and extending the shelf-life of foods. As the use of irradiation increases, and new applications for its use are developed, there is also increased interest in detection methods for monitoring the presence of irradiated items in the food supply (1-3). Thermoluminescence (TL) detection exploits the tendency of minerals to trap free electrons when exposed to ionizing radiation. When heated, the minerals release the free electrons with an associated emission of light which can be detected and the intensity measured (4-6), because the method is very sensitivity and relatively simple, it is applicable to a wide ranges of foodstuffs containing salt (7-11) and minerals (12-16). Perilla and sesame seeds are used as sources of edible oils in Korea (17,18), with imported seeds increasingly used as domestic production decreases (19,20). If perilla or sesame seeds are contaminated with pathogenic microorganisms or infested with pests, great losses of the products will certainly be incurred. Although they are not currently approved by the Korean government for irradiation, if irradiation is adapted to make them safe, TL would be a useful method to detect irradiated perilla and sesame seeds.

Previously, a number of research studies to detect irradiated perilla and sesame seeds have been carried out (15-20). However, the changes in the TL characteristics induced by storage conditions have not yet been reported. Hence, this work investigated the changes in TL characteristics of minerals separated from Korean perilla and sesame seeds during storage under normal room and darkroom conditions, and evaluated the usefulness of TL for detecting irradiated perilla and sesame seeds after long-term storage.

MATERIALS AND METHODS

Materials, irradiation and storage

Korean perilla and sesame seeds were purchased from a local market (Daejon, Korea). Samples were packed in polyethylene bags and divided into two portions (normal room and darkroom conditions). Irradiation was carried out using an irradiator (AECL, Canada) equipped with

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a Co-60 source at the Korea Atomic Energy Research Institute. Samples were irradiated to final absorption doses of 1, 5, and 10 kGy with a dose rate of 10 kGy/h. The absorbed doses were determined using a ceric-cerous dosimeter. After irradiation, the samples of for normal room conditions were stored for 3 months in a laboratory with daylight or fluorescent light. The samples for darkroom conditions were stored for the same period in a chamber oven (K.M.C-1203P3, Vision Scientific Co., LTD, Seoul, Korea) to block exposure to light at room temperature.

Preparation of mineral samples

The preparation of mineral samples was carried out according to method DIN EN 1788 (4) as previously described by Yi et al. (15).

Measurement of first and second glow curves

The separated minerals (1 mg) were deposited onto a clean stainless steel disc (10 mm diameter, 0.5 mm thickness) and fixed with a silicon solution, which was a 1:5 mixture of silicon rubber (LDC 210, Dow Corning Korea Ltd, Seoul, Korea) and hexane, respectively. The fixed samples were then dried and used for the TL measurements. The first and second glow curves were measured by a thermoluminescence (TL) reader (Harshaw 3500, Wermelskirchen, Germany) according to method of Yi and Yong (16).

Calculation of glow curve ratios

The glow curve ratio I (G1) was determined as the ratio of the first glow curve of unirradiated sample divided by the second glow curve of irradiated sample at 1, 5 and 10 kGy. Ratio II (G2, G3 and G4) was determined as the ratio of the first glow curve per irradiation dose (G2=1 kGy, G3=5 kGy and G4=10 kGy) divided by the second glow curve of re-irradiated samples at 1, 5 or 10 kGy. Measurements of the first and second glow curves of all the samples were repeated three times (4,7,17).

Comparison of the shape and ratio of glow curve

Evaluation of the data was carried out according to DIN EN 1788 (4) and the method of Yi and Yang (7,16,17).

RESULTS AND DISCUSSION

TL intensity of first glow curve

As shown in Table 1, TL intensities for minerals separated from irradiated sesame seed linearly increased from 0 kGy to 5 kGy and slightly increased from 5 to 10 kGy at day 0 of storage. This trend was also similarly observed in minerals separated from irradiated perilla seed. The TL intensities measured immediately after irradiation were approximately two times higher in minerals separated from irradiated perilla seed than those from irradiated sesame seed. This tendency was also observed after 3 months of storage, regardless of storage conditions. Several other reports have indicated that the TL intensity of irradiated samples is higher than that of unirradiated samples (5-7), and that the intensity linearly increases up to 5 kGy and is slightly increased or maintained over 5 kGy (7,15-17).

Decay rate

The effect of storage conditions on the decay rate of TL intensity in irradiated perilla seeds is shown in Fig. 1. The decay rate in sesame seeds was higher in darkroom conditions than in normal room. On the other hand, the decay rate in perilla seeds was higher in normal room conditions than in darkroom. The minerals separated from perilla seed had a higher decay rate compared to the minerals separated from sesame seeds. Although the TL intensity decreased with time, detection of irradiation by remaining TL intensity was possible until 3 months of storage.

Similar results on the decay of TL intensity have been reported. Yi and Yang (16) reported that when minerals separated from irradiated shellfish were stored in a darkroom blocked from light or in a room lighted naturally, TL intensity of the sample dramatically decreased after 3 months of storage regardless of storage conditions. Their results were in agreement with the results reported in this paper.

Table 1. TL intensities of first glow curves of minerals separated from irradiated Korean sesame and perilla seeds measured immediately after irradiation and after storage of three months in room and darkroom conditions

(unit: nano coulombs (nC/mg))

Samples	Storage periods (months)	Storage conditions	Irradiation doses (kGy)			
			0	1	5	10
Sesame seed	0		159±18	852±150	2,041±447	2,215±171
	3	Room	158±20	679±51	1,596±189	1,786± 55
		Darkroom	258±11	587±25	1,284±101	1,570±133
Perilla seed	0		182±12	2,169±274	5,182±875	5,706±372
	3	Room	404±36	1,001±194	2,064±414	2,473±297
		Darkroom	250±46	1,116±291	2,454±266	2,671±329

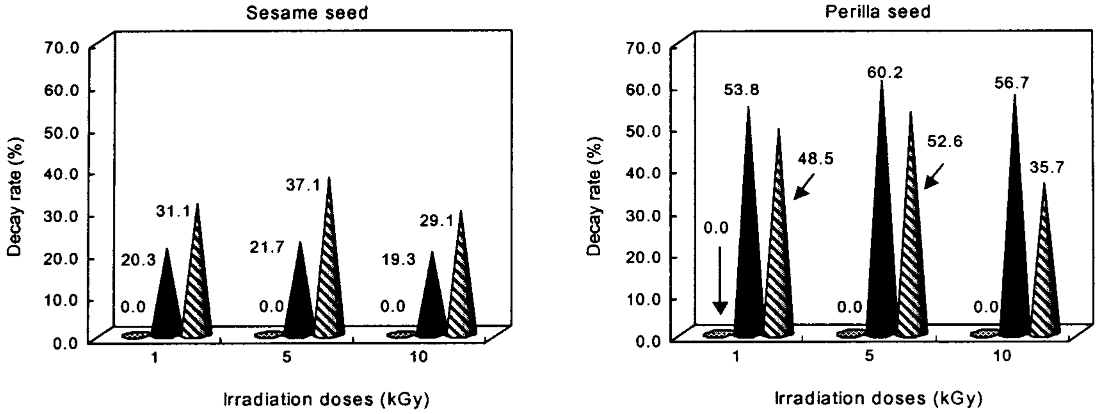

Fig. 1. Decay rates according to storage conditions in TL intensity of mineral separated from Korean perilla and sesame seeds irradiated at various doses.
□ Decay rate of sample measured immediately after irradiation.
■ Decay rate of sample stored during 3 months under room condition.
▨ Decay rate of sample stored during 3 months under darkroom condition.

TL intensity of second glow curve and glow curve ratio

Table 2 shows TL intensities of the second glow curves that are needed to calculate the glow curve ratios. TL intensities of the second glow curves increased with increasing irradiation doses, and, as in previous studies, were typically two fold higher trend approximately of the first glow curve.

The TL intensities of the second glow curves were higher in minerals separated from perilla seeds than from sesame seeds and the difference between storage conditions was not clearly observed. Glow curve ratios calculated from TL intensities of the first and second glow curves are shown in Table 3. According to DIN EN 1788 (4), TL glow curve ratios from re-irradiated samples at doses of 1 or 10 kGy are typically greater than 0.5, whereas those from the non-irradiated samples, which naturally have very low intensities in the first glow curves, are below 0.1 (4,15-17). Therefore, to confirm that the published range is ideal for glow curve ratio determination in oil seeds, re-irradiation was performed at 1, 5 and 10 kGy and the corresponding glow curve ratios were calculated. Gl, which was calculated from non-irradiated samples, were below 0.1 regardless of storage conditions and mineral samples. Therefore, they could be classified as non-irradiated samples based on definitions from previously published papers (4,7,15-17).

Because glow curve ratios, G2, calculated from mineral samples irradiated at 1 kGy were always below 0.5 under all conditions, they were also classified as non-irradiated samples. Since samples tested on day 0 of storage had glow curve ratios of G3 and G4 calculated from TL intensity of re-irradiation at a dose of 1 kGy were higher than 0.5, they were classified as irradiated samples. However, since samples calculated from TL intensity of re-irradiation doses of 5 or 10 kGy were below 0.5, except for glow curve ratios of G4 calculated from TL intensity of re-irradiation dose of 5 kGy, they were classified as non-irradiated samples. Korean sesame seeds had glow curve ratios above 0.5 after three months only in samples (G3 and G4) re-irradiated at 1 kGy and stored in normal room conditions. Korean perilla seeds had glow curve ratios above 0.5 after three months only in sample (G4) re-irradiated at 1 kGy and stored under darkroom conditions.

Table 2. TL intensities of second glow curves of minerals separated from irradiated Korean sesame and perilla seeds measured immediately after irradiation and after storage of three months in room and darkroom conditions

(unit: nano coulombs (nC/mg))

Samples	Storage periods (months)	Storage conditions	Re-irradiation doses (kGy)		
			1	5	10
Sesame seed	0		2,422±661	4,376±845	5,232±948
	3	Room	2,537±142	6,384±693	6,612±302
		Darkroom	2,810±166	6,985±24	9,147±817
Perilla seed	0		6,602±970	11,085±2215	16,334±1,073
	3	Room	7,295±1,994	16,153±3300	18,988±2,853
		Darkroom	7,009±2,328	14,165±5875	2,1698±3,848

Table 3. The changes according to storage conditions of glow curve ratios calculated from TL intensity of the first and second glow curves of irradiated Korean sesame and perilla seeds

Samples	Storage periods (months)	Storage conditions	Irradiation dose (kGy)	Glow curve ratios			
				GI	GII		
				G1[1)]	G2[2)]	G3[3)]	G4[4)]
Sesame seed	0		1	0.0659	0.3519	**0.8429**	**0.9147**
			5	0.0364	0.1947	0.4665	**0.5062**
			10	0.0305	0.1628	0.3901	0.4233
	3	Room	1	0.0623	0.2676	**0.6292**	**0.7041**
			5	0.0248	0.1063	0.2500	0.2798
			10	0.0239	0.1027	0.2414	0.2702
		Darkroom	1	0.0919	0.2091	0.4572	**0.5590**
			5	0.0370	0.0841	0.1839	0.2249
			10	0.0283	0.0642	0.1405	0.1717
Perilla seed	0		1	0.0276	0.3286	**0.7849**	**0.8643**
			5	0.0164	0.1957	0.4675	**0.5148**
			10	0.0111	0.1327	0.3173	0.3493
	3	Room	1	0.0555	0.1373	0.2829	0.3391
			5	0.0251	0.0620	0.1277	0.1531
			10	0.0213	0.0527	0.1087	0.1303
		Darkroom	1	0.0357	0.1593	0.3502	**0.5237**
			5	0.0176	0.0788	0.1733	0.2592
			10	0.0115	0.0515	0.1131	0.1692

[1)]G1=TL intensity of the first glow curve of unirradiated samples/TL intensity of the second glow curve of re-irradiated samples at 1, 5 or 10 kGy.
[2)]G2=TL intensity of the first glow curve of irradiated samples at 1 kGy/TL intensity of the second glow curve of re-irradiated samples at 1, 5 or 10 kGy.
[3)]G3=TL intensity of first glow curve of irradiated samples at 5 kGy/TL intensity of the second glow curve of re-irradiated samples at 1, 5 or 10 kGy.
[4)]G4=TL intensity of first glow curve of irradiated samples at 10 kGy/TL intensity of the second glow curve of re-irradiated samples at 1, 5 or 10 kGy.

Shape of glow curve

The shapes of the second glow curves of minerals separated from irradiated perilla and sesame seeds were measured immediately after irradiation and after 3 months of storage under normal room and darkroom conditions (Fig. 2 and 3). Since irradiated samples had the unique first glow curves when heated to around 200°C, but not non-irradiated samples did not, detection of irradiation was possible by comparing the unique first glow curves.

Shapes of the first and second glow curves of minerals separated from irradiated perilla and sesame seeds were not significantly different regardless of TL intensity. When TL intensity of the first glow curves measured immediately after irradiation was compared with the first glow curves measured after 3 months of storage under either room conditions, identification of the unique first glow curves was possible, even though the heights of the first glow curves were lower. Sesame seeds stored for 3 months in a darkroom had the first glow curves that were clearer and higher than those stored under normal conditions, but the storage effect was opposite in perilla seeds. If glow curve ratios between 0.1 and 0.5 are obtained, interpretation of the shape of the glow curves is needed to decide whether the sample has been irradiated or not (4,16). Accordingly, interpretation of the shape of the glow curves were used for glow curve ratios of irradiated samples with TL intensities below 0.5 in Table 3. The unique first glow curve shapes, which are only seen in irradiated samples, were observed in irradiated samples with glow curve ratios below 0.5, allowing them to be classified as irradiated samples. The G1, which had glow curve ratios below 0.1, were classified as non-irradiated samples because the unique first glow curve was not present. Since the second glow curves were lower domain than the first glow curves, as previously reported by other investigators (15-17), detection of irradiation by differences of distribution between the first glow curve and the second glow curve was possible.

Maximum TL temperature

Maximum TL temperature, defined as the temperature corresponding to the highest peak point on the glow curve, is an additional factor related to the first and second glow curves that can be used for detecting whether or not a sample is irradiated. Several papers have suggested that

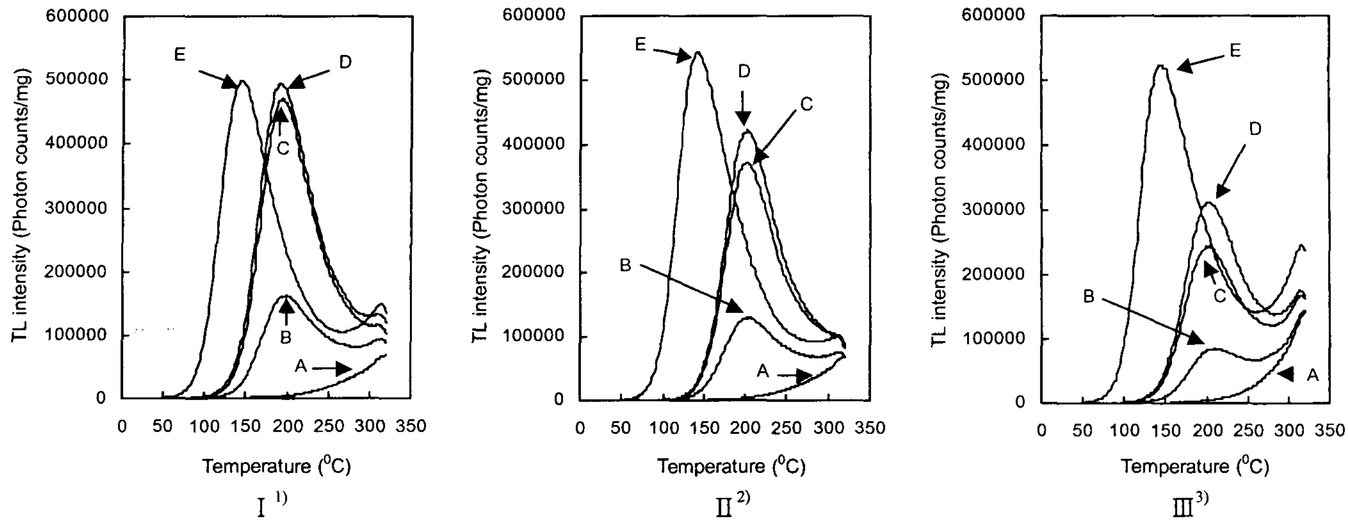

Fig. 2. Properties of the first and second glow curves according to storage conditions of minerals separated from Korean sesame seeds irradiated at various doses.
1)The first and second glow curves measured immediately after irradiation.
2)The first and second glow curves measured after storage during 3 months under room condition.
3)The first and second glow curves measured after storage during 3 months under darkroom condition.
A: The first glow curve of minerals separated from unirradiated samples.
B: The first glow curve of minerals separated from samples irradiated at 1 kGy.
C: The first glow curve of minerals separated from samples irradiated at 5 kGy.
D: The first glow curve of minerals separated from samples irradiated at 10 kGy.
E: The second glow curve of samples re-irradiated at 1 kGy.

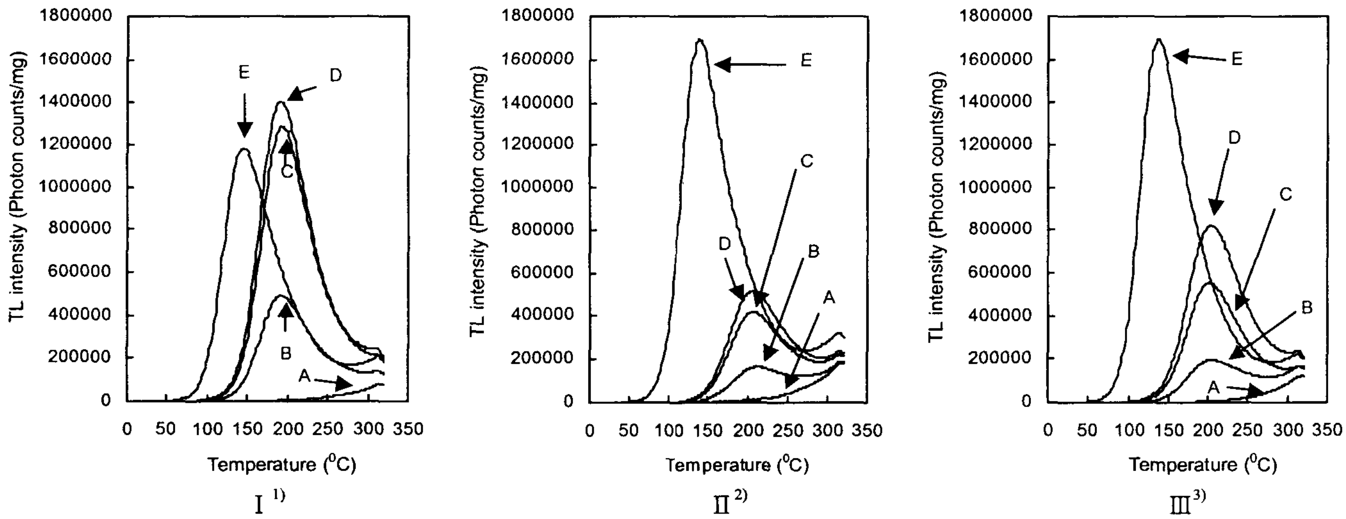

Fig. 3. Properties of the first and second glow curves according to storage conditions of the mineral separated from Korean perilla seed irradiated at various doses.
1)The first and second glow curves measured immediately after irradiation.
2)The first and second glow curves measured after storage during 3 months under room condition.
3)The first and second glow curves measured after storage during 3 months under darkroom condition.
A~E: Refer to the legend of Fig. 2.

a comparison of the maximum TL temperature combined with observation of the shapes of glow curves is more accurate for identifying irradiated and non-irradiated samples (15-17), because detection of irradiation by the shape of the first and second glow curves is an empirical method requiring careful and skillful observation. However, detection of irradiation by identifying the maximum TL temperature is a precise measurement that is not subject to interpretation. Therefore, the maximum TL temperature may often provide more conclusive detection of irradiated foods than comparison of the shape of the glow curves. Maximum TL temperatures of the first and second glow curves in minerals separated from irradiated sesame and perilla seeds were measured immediately after irradiation, and those of the irradiated seeds stored under normal room and darkroom conditions during 3 months were measured. The data are shown in Table 4 and 5. Maximum TL temperatures of the first glow curves in minerals separated from irradiated sesame seed were between 191.1±2.1 and 213.2±1.5°C, and those of irradiated perilla seed were between 193.7±1.6 and 215.5±2.1°C (Table 4). Maximum TL temperatures of the second glow curves of minerals separated from irradiated sesame and perilla seeds were measured immediately after irradiation, and those stored under normal room and darkroom conditions for 3 months were between 142.5±1.4~150.2±5.7 and 137.9±2.0~142.9±3.4°C, respectively (Table 5). Therefore, identification of non-irradiated and irradiated samples was possible by differences in their maximum TL temperatures. In the previous studies (15-17), maximum TL temperatures and the shapes of the second glow curve have always been in a lower range than those of the first glow curve, which is consistent with our results.

Therefore, we have demonstrated that detection of irradiation in Korean sesame and perilla seeds is possible, for seeds stored for up to 3 months, using several ther moluminescence (TL) characteristics; including the TL intensity, glow curve ratio, maximum TL temperature, and the shape of the glow curves.

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Table 4. The changes according to storage conditions in maximum TL temperatures of the first glow curves of minerals separated from irradiated Korean sesame and perilla seeds (unit: °C)

Samples	Storage periods (months)	Storage conditions	Irradiation dose (kGy)			
			0	1	5	10
Sesame seed	0		ND[1)]	197.7±5.7	195.1±3.9	191.9±2.1
	3	Room	Darkroom	203.2±3.4	205.1±1.3	203.7±2.4
		Darkroom	ND	213.2±1.5	204.5±4.2	206.8±2.1
Perilla seed	0		Darkroom	195.5±1.3	195.5±3.6	193.7±1.6
	3	Room	ND	215.5±2.1	210.5±2.8	209.1±1.4
		Darkroom	ND	210.0±7.5	205.0±3.6	207.8±1.3

[1)]Not detected.

Table 5. The changes according to storage conditions in maximum TL temperatures of the second glow curves of minerals separated from irradiated Korean sesame and perilla seeds (unit: °C)

Samples	Storage periods (months)	Storage conditions	Irradiation dose (kGy)		
			1	5	10
Sesame seed	0		145.7±4.7	147.0±2.8	149.1±1.7
	3	Room	145.2±1.5	142.5±1.4	142.9±2.1
		Darkroom	150.2±5.7	142.9±4.4	142.5±2.4
Perilla seed	0		142.9±3.4	140.0±0.8	141.5±2.1
	3	Room	142.4±2.4	140.2±2.1	137.9±2.0
		Darkroom	141.6±2.8	142.0±3.4	139.2±2.9

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