J. Food Sci. Nutr.
Vol. 6, No. 1, p. 23～28 (2001)

Changes in Thermoluminescence of Mineral Separated from Irradiated Shellfish under Various Storage Conditions

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Abstract

A study was carried out to establish a detection method of irradiated shellfish through thermoluminescence (TL). The TL intensity of first glow curves for irradiated bloody, freshwater, and short-neck shellfish increased from control until 5 kGy and increased slightly from 5 kGy until 10 kGy. Maximum TL temperatures of all irradiated samples tested were below 230°C, within temperature interval of 150～250°C recommended for evaluation. Since just in control, glow curve ratios of G3 and G4 calculated from re-irradiated (1 kGy) bloody, freshwater and short-neck were over 0.5, detection in control was possible. However, as glow curve ratios after three months were below 0.5, detection by glow curve ratios after three months was impossible. Gl, which calculated from unirradiated samples, exhibited below 0.1, they were classified as unirradiated. In all samples, all the irradiated shellfish could be classified correctly as irradiated by the maximum TL temperatures and shape of the second glow curve because those were shown in a lower temperature region than those of the first glow curve.

Key words: shellfish, gamma irradiation, thermoluminescence (TL)

INTRODUCTION

Foods are treated with ionizing radiation to decrease microbial content and insect infestations, and to extend shelf life. Treatment of food by ionizing radiation is accepted for specific purposes in many countries, although in other countries the sale of irradiated foods for human consumption is prohibited (1). Shellfish are particularly susceptible to bad handling and poor quality control, the major problem being associated with microbial contamination, especially by pathogens such as species of *Salmonella* and *Vibrio*. If they are not inhibited or destroyed, these microorganisms may present a public health hazard and growth of spoilage flora will result in a decrease in the shelf-life of the product (2). Therefore, irradiation of shellfish should be considered as a useful tool to increase assurance of their safety. With the necessity of increasing irradiation of shellfish, a detection technique should be requested particularly by consumer organizations and developed for correct and comprehensive information about food irradiation (3).

Thermoluminescence (TL) is radiation-specific phenomena from energy stored by trapped charge carriers following irradiation (4). Releasing such stored energy by thermal stimulation can result in a detectable luminescence emission (5,6). Thermoluminescence (TL) has been applied to various foods such as spices, herbs (7-10), fruits (11), onions, potato (12), and seafoods such as prawn, squid, shrimp, lobster, crayfish, scampi and fish (13-16) as to be an irradiation-detection technique. It was specifically shown to be an applicable method for the detection of all irradiated spices (17-22). Previous TL detection studies for shellfish were carried out just for shell powder, not separated mineral of mollusc shells such as blue mussel, chowder clam, dog cockle, softshell clam, periwinkle, common whelk, edible cockle (23), oyster and king scallop (15).

Therefore, the aim of this study was to observe the changes in TL, under different storage conditions (with and without light) and storage periods, of minerals separated from irradiated shellfish (bloody, freshwater, and short-neck) not yet examined in previous studies and add new data in this field such as changes of glow curve ratios and maximum TL temperatures with various irradiation dose and experimental conditions.

MATERIALS AND METHODS

Materials and irradiation

Shellfish (bloody, freshwater, and short-neck) were purchased from a local market. Samples (500 g) were packed in polyethylene bags and were irradiated using a Co-60 irradiator (AECL, IR-79, Ontario, Canada) with 1, 5, and 10 kGy, with a dose rate of 10 kGy/h at the Korea Atomic Energy Research Institute. To measure the exact total absorbed dose of gamma irradiation, the dose rates for cobalt-60 sources were determined using a ceric-cerous dosimeter.

Preparation of mineral extract

The 500 g samples were agitated in 5 L distilled water for 5 min. The suspended samples were filtered through a 250 μm nylon cloth, and the constituents retained were discarded. The filtered solution was allowed to settle for about 5 min to separate the sediment minerals from the supernatant. The sediment minerals were suspended in 5 mL sodium poly-

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tungstate solution which was adjusted to a density of 2.0 g/mL by the addition of water for the separation of minerals and adhering organic materials. The minerals were pelleted through centrifugation for 2 min at 1000 rpm after a 5 min ultrasonic treatment. The low-density layer was decanted off. This procedure was repeated until all the organic materials were removed. After the polytungstate solution was removed, the minerals were washed twice in water and pelleted through centrifugation at 1000 rpm, followed by a 10 min treatment with 1 M HCl to remove carbonates. After neutralizing with 1 M NH_4OH for 10 min, the solution was discarded. The minerals were washed twice in deionized water and centrifuged at 1000 rpm for 2 min to separate a mineral fraction. After the supernatant was decanted, the remaining water was then rinsed off with 3 mL of acetone twice and dried in a laboratory oven at 50°C for 3 hr. The dried minerals (1 mg) were deposited onto a clean stainless steel disc (10 mm diameter, 0.5 mm thickness), fixed with silicon solution, then dried and measured with a thermoluminescence (TL) reader (24). After mineral separation, samples were divided into two groups of storage conditions, with light and without light at room temperature.

Thermoluminescence (TL) measurement

TL measurement was carried out using a TL reader (Harshaw 3500, Wermelskirchen, Germany) with heat ranging from 50 to 320°C at a rate of 6°C/s and held at 320°C for 10 s. The light emission was recorded in a temperature-dependent mode as a glow curve and was measured in units of nano coulombs (nC). After the first glow curve was measured, the discs with the minerals were subsequently reirradiated using a Co-60 gamma rays with a normalizing dose of 1, 5 and 10 kGy. The TL intensity was measured again after the reirradiation step (second glow curve). The glow curve ratios I (G1) (first glow curve of unirradiated sample/second glow curve of irradiated sample at 1 and 5 kGy) and II (G2, G3, G4) (first glow curve per irradiation dose (G2=1 kGy, G3=5 kGy, G4=10 kGy)/second glow curve of irradiated spices at 1, 5 and 10 kGy) were then determined. TL measurements of all samples were repeated three times (24).

Statistical analysis

Significant differences were determined by using Duncan's multiple range test in a one way ANOVA with SPSS (Statistical Package for Social Science) version 7.5. All experiments were repeated three times.

RESULTS AND DISCUSSION

TL intensity of mineral separated from irradiated shellfish

Minerals were extracted with organic compounds from several shellfish such as bloody, freshwater, and short-neck. They were separated with sodium polytungstate solution (2.0 g/mL) following treatment by acid-base and dried at 50°C overnight. TL intensities of separated minerals with different irradiation doses and storage conditions (with and without light) were measured. As shown in Table 1 and Fig. 1, TL intensities of minerals from unirradiated and irradiated bloody, freshwater, and short- neck increased from 114.9±11.6, 175.9±15.9, and 164.3±19.5 nC, respectively, in the unirradiated control to 5,138.1±860.1, 10,673.0±815.1, and 10,225.9±1,661.3 nC at 5 kGy, respectively, and slightly increased to 5,662.2±553.1, 10,659.5±112.6, and 11,616.0±1,482.2 nC, respectively, at 10 kGy. These results were similar to our previously published data from TL intensities with increasing irradiation dose in minerals separated from Korean perilla and sesame seeds (25) and to those reported for shell powder by Ziegelmann

Table 1. TL intensities of first glow curves of minerals separated from irradiated shellfish measured immediately after irradiation (control) and after three months in conditions with and without light (Unit : nano coulombs (nC/mg))

Samples	Storage conditions		Irradiation dose (kGy)			
			Control[2)]	1	5	10
Bloody	Control[1)]		^{A}214.9±11.6$^{3)a}$	^{A}1,504.9±94.8^{b}	^{A}5,138.1±860.1^{c}	^{A}5,662.2±553.1^{c}
	After three months	With light	^{A}293.7±51.5^{a}	^{B}733.4±79.2^{b}	^{B}1,736.9±167.6^{c}	^{B}2,397.5±324.1^{d}
		Without light	^{A}235.3±24.3^{a}	^{B}705.7±114.9^{b}	^{B}2,166.6±154.7^{c}	^{B}2,938.0±438.2^{d}
Freshwater	Control		^{I}215.9±15.7^{a}	^{I}2,825.9±511.0^{b}	^{I}10,673.0±815.1^{c}	^{I}10,659.5±112.6^{c}
	After three months	With light	^{I}220.9±43.8^{a}	^{II}1,488.9±155.4^{b}	^{II}3,534.0±456.9^{c}	^{II}4,195.3±571.9^{c}
		Without light	^{II}210.7±15.8^{a}	^{II}1,452.0±425.3^{b}	^{II}3,150.8±772.3^{c}	^{II}3,646.2±1,359.8^{c}
Short-neck	Control		$^{\alpha}$264.3±19.5^{a}	$^{\alpha}$4,890.9±644.7^{b}	$^{\alpha}$10,225.9±1,661.3^{c}	$^{\alpha}$11,616.0±1,482.2^{c}
	After three months	With light	$^{\alpha}$248.3±50.5^{a}	$^{\beta}$1,729.2±224.1^{b}	$^{\beta}$3,505.9±375.7^{c}	$^{\beta}$3,264.6±636.9^{c}
		Without light	$^{\alpha}$230.8±32.9^{a}	$^{\beta}$1,584.7±363.7^{b}	$^{\beta}$2,822.7±259.8^{c}	$^{\beta}$3,760.5±428.3^{c}

[1)]Control : sample measured immediately after irradiation
[2)]Control : unirradiated sample
[3)]Means±standard deviation for 3 measurements
$^{a-c}$Means with the same superscripts in each row are not significantly different among group by Duncan's multiple range test in one way ANOVA ($p<0.05$).
$^{A-B, I-II, \alpha-\beta}$Means with the same superscripts in each column are not significantly different among group by Duncan's multiple range test in one way ANOVA ($p<0.05$).

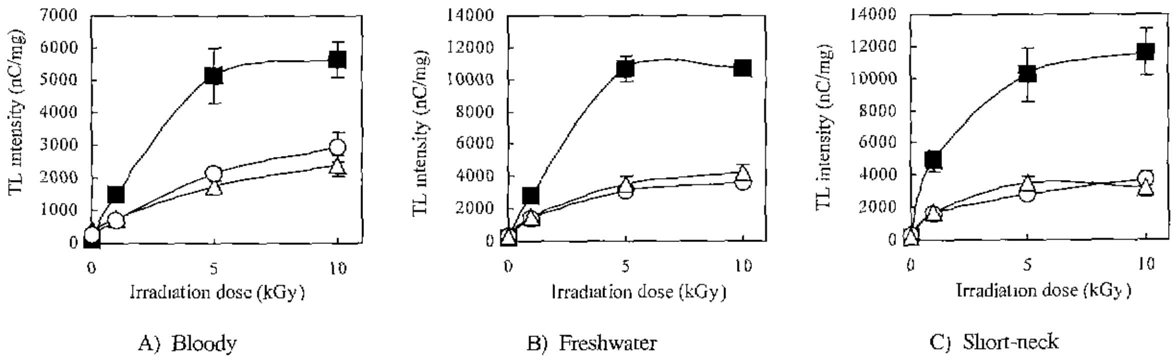

Fig. 1. The changes of TL intensities of minerals separated from irradiated shellfish measured immediately after irradiation (control) and after three months in conditions with and without light. ■—■: Control, △—△: After three months in condition with light, ○—○: After three months in condition without light

et al. (23). In all samples tested, TL intensities measured after three months showed a decrease with an increase of storage periods but there were no significant differences between storage conditions (with and without light). However, as irradiated samples showed higher TL intensities than those of the unirradiated samples, in all conditions, detection of irradiation for irradiated shellfish was still possible after three months. Therefore, TL method by separated mineral can be proposed as the method to detect the irradiation treatment of shellfish. Generally, the TL intensities of minerals separated from irradiated samples were higher than those of unirradiated samples (9,11,16). Schreiber et al. (26) reported that TL emission occurs when the excited electrons return to the original level at a certain temperature. Therefore, the principle of TL emission in mineral separated from irradiated shellfish used in this study seemed like that reported by Schreiber et al. (26).

TL intensities of second glow curve and glow curve ratios

Glow curve ratios of irradiated samples are typically greater than 0.5, whereas those of unirradiated samples are generally below 0.1. 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, since the shapes of first glow curve showed in higher temperature region than those of the second glow curve (24). A comparison of the glow curve ratio as well as an analysis of glow curve shapes were preferred for identifying irradiated or unirradiated samples in several studies (7,11,16). In addition, Hammerton and Banos (17) reported that all spices tested were clearly identified by re-irradiation as either unirradiated or irradiated and the TL ratios (glow curve ratio) varied between 0.0039 and 0.19 for unirradiated samples, and between 0.79 and 2.4 for samples irradiated with 5 kGy. TL intensities of second glow curves needed for calculations of glow curve ratios are shown in Table 2. TL intensities of second glow curves increased with increasing re-irradiation dose and that of 10 kGy increased slightly compared with that of 5 kGy. However, there was no difference of TL intensities under differing storage conditions and time periods. TL intensities of the reirradiated shellfish were in the order of freshwater, short-neck, bloody at 5 and 10 kGy. Irradiated freshwater and short-neck exhibited high TL intensities compared with the irra-

Table 2. TL intensities of second glow curves of minerals separated from irradiated shellfish measured immediately after irradiation (control) and after three months in conditions with and without light (Unit : nano coulombs (nC/mg))

Samples	Storage conditions		Irradiation dose (kGy)		
			1	5	10
Bloody	Control[1)]		^{A}5,025.9±316.4^{a}	^{A}12,911.4±2,339.1^{b}	^{A}14,825.2±3,012.7^{b}
	After three months	With light	^{B}5,903.2±836.3^{a}	^{B}15,520.6±1,269.3^{b}	^{B}21,148.4±1,130.1^{c}
		Without light	^{A}4,640.6±838.3^{a}	15,470.1±1,489.9^{b}	^{AB}17,678.8±3,309.1^{b}
Freshwater	Control		^{I}9,725.5±1,866.2^{a}	^{I}28,384.6±1,637.6^{b}	^{I}29,675.7±5,321.1^{b}
	After three months	With light	^{II}24,823.1±4,882.6^{a}	^{II}61,737.1±10,200.6^{b}	^{II}60,244.1±4,259.2^{b}
		Without light	^{III}16,667.8±4,737.9^{a}	^{III}37,015.3±6,074.9^{b}	^{III}37,977.4±9,622.8^{b}
Short-neck	Control		$^{\alpha\beta}$20,009.2±2,347.1^{a}	$^{\alpha}$31,161.2±3,513.3^{b}	$^{\alpha}$40,107.6±4,675.3^{c}
	After three months	With light	$^{\beta}$25,719.9±4,048.1^{a}	$^{\beta}$44,404.9±6,333.9^{b}	$^{\alpha}$46,758.7±20,969.5^{b}
		Without light	$^{\alpha}$16,080.6±3,942.5^{a}	$^{\alpha}$32,522.5±2,077.5^{b}	$^{\alpha}$42,526.5±5,197.3^{c}

[1)]Control : sample measured immediately after irradiation

[a-c]Means with the same superscripts in each row are not significantly different among group by Duncan's multiple range test in one way ANOVA ($p<0.05$).

[A-B, Ⅰ-Ⅲ, α-γ]Means with the same superscripts in each column are not significantly different among group by Duncan's multiple range test in one way ANOVA ($p<0.05$).

diated bloody. This difference observed among the shellfish seemed to be caused by the differences in mineral composition and size used for TL measurement. Glow curve ratios calculated from first glow curves per irradiation doses (control, 1, 5 and 10 kGy) divided by second glow curves per re-irradiation doses (1, 5 and 10 kGy) are shown in Table 3. Generally, TL glow curve ratio from irradiated samples are greater than 0.5, whereas those from unirradiated samples are below 0.1 (24). Since glow curve ratios of G3 and G4 calculated from re-irradiated (1 kGy) bloody, freshwater and short-neck in control measured immediately after irradiation were 1.0223 and 1.1266, 1.0974 and 1.0960, and 0.5110 and 0.5805, respectively, detection was possible by glow curve ratio G3 and G4. However, as glow curve ratios after three months were below 0.5, detection by glow curve ratios after three months was impossible. While Gl, which calculated from unirradiated samples, exhibited below 0.1, they were classified as unirradiated.

Maximum TL temperature and shape of glow curve

Maximum TL temperatures of first and second glow curves in the mineral separated from irradiated bloody, freshwater, and short-neck are shown in Table 4 and 5. Maximum TL temperatures first glow curves in the mineral separated from irradiated bloody, freshwater, and short-neck were between $193.7\pm2.8\sim222.3\pm6.4$, $194.6\pm5.5\sim218.6\pm2.1$ and $190.5\pm0.8\sim216.4\pm4.8$°C, respectively, and those of second glow curves were between $132.8\pm1.5\sim152.0\pm5.9$, $122.9\pm2.1\sim156.5\pm4.8$ and $132.5\pm2.8\sim151.1\pm2.8$°C, respectively. Therefore, detection of irradiation was possible by comparison of maximum TL temperatures because that of the second glow curve showed at the lower temperature than that of first glow curve. Maximum TL temperatures of all the irradiated samples were below 230°C, within the 150~250°C temperature interval recommended by DIN EN 1788 (24). If glow curve ratios between 0.1 and 0.5 or below 0.1 were obtained, interpretation of the shape of the glow curves was needed to decide whether the sample was irradiated or not. Usually, first glow curves of irradiated foodstuffs exhibit a maximum between 150°C up to 250°C, whereas low level natural radioactivity causes TL signals in the deep traps above 300°C (24). The general pattern of first and second glow curves in irradiated bloody are shown

Table 3. The changes of glow curve ratios of minerals separated from irradiated shellfish measured immediately after irradiation (control) and after three months in conditions with and without light

Samples	Storage conditions		Irradiation dose (kGy)	Glow curve ratios			
				G I	G II		
				G 1[2]	G 2[3]	G 3[4]	G 4[5]
Bloody	Control[1]		1	0.0220	0.2994	**1.0223**	**1.1266**
			5	0.0088	0.1165	0.3979	0.4385
			10	0.0077	0.1015	0.3465	0.3819
	After three months	With light	1	0.0497	0.1242	0.2942	0.4061
			5	0.0189	0.0472	0.1119	0.1544
			10	0.0138	0.0346	0.0821	0.1133
		Without light	1	0.0507	0.1520	0.4668	0.6331
			5	0.0152	0.0456	0.1400	0.1899
			10	0.0133	0.0399	0.1225	0.1661
Freshwater	Control		1	0.0180	0.2905	**1.0974**	**1.0960**
			5	0.0061	0.0995	0.3760	0.3755
			10	0.0059	0.0952	0.3596	0.3591
	After three months	With light	1	0.0088	0.0599	0.1423	0.1690
			5	0.0035	0.0241	0.0572	0.0679
			10	0.0036	0.0247	0.0586	0.0696
		Without light	1	0.0126	0.0854	0.1890	0.2187
			5	0.0056	0.0392	0.0851	0.0985
			10	0.0055	0.0382	0.0829	0.0960
Short-neck	Control		1	0.0082	0.2444	**0.5110**	**0.5805**
			5	0.0052	0.1569	0.3281	0.3727
			10	0.0096	0.0672	0.1363	0.1269
	After three months	With light	1	0.0055	0.0389	0.0789	0.0735
			5	0.0055	0.0389	0.0789	0.0735
			10	0.0053	0.0369	0.0749	0.0698
		Without light	1	0.0143	0.0985	0.1755	0.2338
			5	0.0070	0.0487	0.0867	0.1156
			10	0.0054	0.0372	0.0663	0.0884

[1]Control : sample measured immediately after irradiation
[2]G1 : first glow curve of unirradiated sample/second glow curve of re-irradiated sample at 1, 5 or 10 kGy
[3]G2 : first glow curve of irradiated sample at 1 kGy/second glow curve of re-irradiated sample at 1, 5 or 10 kGy
[4]G3 : first glow curve of irradiated sample at 5 kGy/second glow curve of re-irradiated sample at 1, 5 or 10 kGy
[5]G4 : first glow curve of irradiated sample at 10 kGy/second glow curve of re-irradiated sample at 1, 5 or 10 kGy

Table 4. Maximum TL Temperatures of first glow curves of minerals separated from irradiated shellfish measured immediately after irradiation (control) and after three months in conditions with and without light (Unit : °C)

Samples	Storage conditions		Irradiation dose (kGy)			
			Control[2)]	1	5	10
Bloody	Control[1)]		320.0±0.0	195.2±1.0	193.7±2.8	193.7±2.8
	After three months	With light	320.0±0.0	219.5±5.5	216.8±3.4	222.3±6.4
		Without light	320.0±0.0	219.1±7.0	214.1±10.9	216.4±0.8
Freshwater	Control		320.0±0.0	196.4±3.4	198.2±7.1	194.6±5.5
	After three months	With light	320.0±0.0	213.7±3.4	218.2±4.2	215.5±1.6
		Without light	320.0±0.0	211.8±3.7	218.6±2.1	215.9±2.7
Short-neck	Control		320.0±0.0	196.4±2.1	190.5±0.8	191.4±2.7
	After three months	With light	320.0±0.0	212.3±2.8	214.1±3.4	213.2±2.3
		Without light	320.0±0.0	209.6±8.2	205.9±5.7	216.4±4.8

[1)]Control : sample measured immediately after irradiation
[2)]Control : unirradiated sample means±standard deviation for 3 measurements

Table 5. Maximum TL Temperatures of second glow curves (GⅡ) of minerals separated from irradiated shellfish measured immediately after irradiation (control) and after three months in conditions with and without light (Unit · °C)

Samples	Storage conditions		Irradiation dose (kGy)		
			1	5	10
Bloody	Control[1)]		148.4±1.6	149.3± 1.3	149.4±2.1
	After three months	With light	138.4±1.3	136.1± 3.4	132.8±1.5
		Without light	150.6±2.7	152.0± 5.9	146.6±3.6
Freshwater	Control		156.5±4.8	151.5± 7.5	155.2±5.6
	After three months	With light	125.3±1.6	121.2±10.3	122.9±2.1
		Without light	142.9±2.1	143.3± 2.1	144.7±5.1
Short-neck	Control		151.1±2.8	145.7± 0.8	146.5±3.0
	After three months	With light	132.9±2.7	133.9± 3.8	132.5±2.8
		Without light	146.1±5.1	140.2± 1.6	141.1±1.3

[1)]Control : sample measured immediately after irradiation

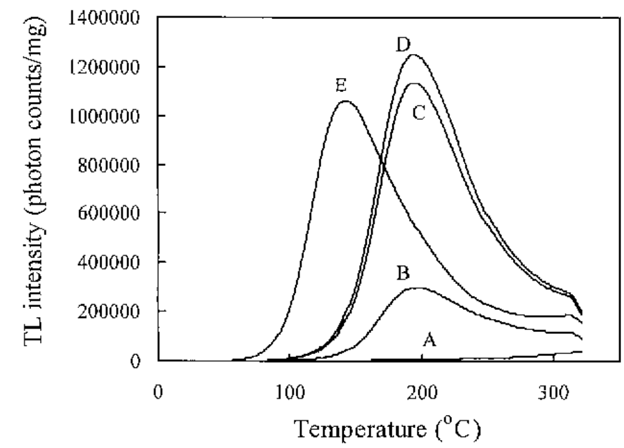

Fig. 2. TL intensity of first and second glow curves of mineral seperated from irradiated bloody at various doses.
A) Unirradiated sample, B) 1 kGy, C) 5 kGy, D) 10 kGy
E) Second glow curve re-irradiated at 1 kGy

in Fig. 2. A unique first glow curve which showed between 150°C up to 250°C was not found in unirradiated bloody but in the samples irradiated at 1, 5 and 10 kGy, unique first glow curve observed. In addition, since the shape of second glow curve was observed at a lower domain than that of first glow curve, detection of irradiated shellfish was possible by shape of glow curves. These results suggest that determination of irradiated shellfish was possible by intensity, glow curve ratio, maximum TL temperature and shape of glow curve of TL measured from mineral separated from shellfishes.

ACKNOWLEDGEMENTS

The authors would like to thank the Ministry of Science and Technology of Korea (Research of the Long-and-Mid-term Nuclear R & D Program) for financial support during this study.

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(Received January 15, 2001)