

## **Influence of Sample Form, Storage Conditions and Periods on Accumulated Pulsed Photostimulated Luminescence Signals of Irradiated Korean Sesame and Perilla Seeds**

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### **Abstract**

A study was carried out to examine the effect of sample form and storage conditions on the accumulated PPSL signals. Korean perilla and sesame seeds were tested as whole samples and separated minerals. Radiation-induced PPSL signals of perilla and sesame seeds themselves significantly increased with irradiation dose up to 5 kGy. On the other hand, a slight decrease in the accumulated PPSL signals was shown at 10 kGy. Similar results were also found in separated minerals. The accumulated PPSL signals of irradiated samples decreased with increasing storage periods. The decay rate was higher in 5 or 10 kGy-irradiated samples than in 1 kGy, in room conditions than in darkroom conditions, and in sesame and perilla seeds themselves than in separated minerals. The accumulated PPSL signals of the irradiated samples measured for 120 s were higher than those measured for 60 s. These results indicated that although the PPSL signal of all samples decreased with increasing the storage time, detection of irradiated samples was still possible after 12 months of storage regardless of sample form and measurement times (60 and 120 s) in both room and darkroom conditions.

**Key words:** Korean perilla and sesame seeds, irradiation, pulsed photostimulated luminescence (PPSL), storage condition

### **INTRODUCTION**

Irradiation is a well-known technique as a new approach in the preservation of foods (1) and its utilization has been allowed in many countries in recent years (2). Increase in the international trade of irradiated foods has created demands for detection methods that will provide regulatory agencies with the tools to ensure compliance with labeling regulations (3). As a part of the regulations for food irradiation, government organizations should be able to detect whether a particular food has been irradiated or not (4). A number of detection methods have been proposed for the identification of irradiated foods (5). Previously, the utilization possibility of pulsed photostimulated luminescence (PPSL) was investigated for irradiated foodstuffs positively suggesting that it is a new screening technique (6-17). However, the difficulty in the application of the PPSL to irradiated foodstuffs is changes in the PPSL signals by storage conditions and period (11). Perilla and sesame seeds and their oils have been used as spices in various foods in the Korean diet, and a large amount of these are imported from China and Sudan, because domestic production has decreased due to a change in agricultural circumstances (7,9,10). Pathogenic microorganisms and pests can be found in perilla and sesame seeds mainly due to improper handling after harvest or in importation stage (7,12). Although they are not commercially irradiated at present, if irradiation is adapted to make them safe from the pathogenic microorganisms and pests, PPSL technique would be

used to monitor the irradiation process. Additionally, as the technique of PPSL is simple, quick and applicable to a wide range of foodstuffs without further preparation (11,13), the utilization of PPSL in detection of irradiated food is considered positively. On identification of the irradiated perilla and sesame seeds, several results were reported (6,7,9,12), but there are no reports on the change in accumulated PPSL signals under various storage conditions for the long-term. The objective of this study was to examine differences in the PPSL signals according to the sample form measured (perilla and sesame seeds themselves; whole sample or minerals separated from the whole sample), and storage conditions and periods on the accumulated PPSL signals and thereby evaluating the possibility of PPSL on detection of irradiated perilla and sesame seeds.

### **MATERIALS AND METHODS**

#### **Materials and irradiation**

Korean perilla and sesame seeds were purchased from a local market (Daejeon, Korea). Samples were packed in polyethylene bags and divided into two portions (room and darkroom conditions). Irradiation was carried out using an irradiator (AECL, Canada) equipped with a Co-60 source at 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 dose rates for cobalt-60 sources were determined using a ceric-cerous dosimeter. After irradiation, the

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samples of room condition were stored for 12 months in laboratory condition, which existed sunlight or a fluorescent light. The samples at 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 by light at room temperature.

#### Preparation of test samples

The Korean sesame and perilla seeds (5 g, each) were directly measured without any other preparation. The separation of minerals was carried out with some modification of the European standard method (18) as follows. The 200 g samples were agitated with 2 L water for 5 min. The suspended samples were filtered through a 250  $\mu$ m-nylon cloth. The filtered solution was allowed to settle to separate the sediment minerals from the supernatant. The sediment minerals were suspended in 5 mL sodium polytungstate (Fluka 71913) solution (2.0 g/mL) for the separation of minerals and adhering organic materials. The minerals were pelleted through centrifugation for 2 min at 1,000 rpm after a 5 min ultrasonic treatment. The supernatant was decanted off. This procedure was repeated three times. 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. After neutralizing with 1 M NH<sub>4</sub>OH for 10 min, the solution was discarded. The minerals were washed twice with deionized water and centrifuged at 1,000 rpm for 2 min to separate a mineral fraction. After the supernatant was decanted to remove remaining water, the minerals were then rinsed off with 3 mL of acetone twice and dried at 50°C for 3 h (18). The dried minerals were used as a sample to measure accumulated PPSL signals.

#### Measurement of pulsed photostimulated luminescence (PPSL)

Measurement of accumulated PPSL signals was carried out using PPSL system purchased from Scottish Universities Research and Reactor Centre (SURRC, Glasgow, UK). The PPSL system is composed of a control unit, sample chamber, and detector head assembly. The control unit contains a stimulation source, which is comprised of an array of infrared light (880 ~ 940 nm) emitting diodes, which are pulsed symmetrically on and off for equal periods. PPSL signal is detected by a bi-alkali cathode photomultiplier tube operating in the photon counting mode and recorded automatically in personal computer connected with the PPSL system. Optical filtering is used to define both the stimulation and detection wavebands. The Korean sesame and perilla seeds themselves (5 g) and minerals (1, 5 and 10 mg) separated from them were introduced in 50 mm diameter of disposable petri dishes (Bibby Sterilin types 122, Glasgow, UK). The samples were measured in the sample chamber for 120 s. The radiation-induced photon counts (PPSL signals) emitting per second from the samples were automatically accumulated in a personal computer and presented the photon counts accumulated up to 60 s and 120 s (13).

## RESULTS AND DISCUSSION

#### Accumulated PPSL signals of Korean sesame and perilla seeds

Results in Table 1 show changes of accumulated PPSL signal according to storage conditions and periods of unirradiated and irradiated sesame and perilla seeds at 1, 5, and

**Table 1.** The changes of accumulated PPSL signals of irradiated sesame and perilla seeds measured by PPSL in the 60 and 120 s measurement times during storage of 12 months under room and darkroom conditions (unit: photon counts)

Samples	Storage periods	Storage conditions	Measurement times (sec)	Irradiation dose (kGy)				
				0	1	5	10	
Sesame seed	0 day		60	933 ± 131	350,055 ± 171,867	614,551 ± 141,950	464,374 ± 152,188	
			120	1,261 ± 157	620,422 ± 315,179	1,088,115 ± 239,982	886,135 ± 156,303	
	After 3 months	Dark room	60	503 ± 115	236,671 ± 111,464	431,232 ± 155,985	355,029 ± 31,131	
		120	674 ± 115	399,237 ± 216,314	737,840 ± 300,013	587,738 ± 45,235		
	Room	60	323 ± 68	11,283 ± 10,967	4,348 ± 2,525	14,566 ± 9,431		
		120	341 ± 48	13,983 ± 9,470	7,772 ± 4,694	26,253 ± 16,893		
	After 12 months	Dark room	60	596 ± 104	244,522 ± 21,514	260,239 ± 21,492	256,487 ± 44,908	
		120	751 ± 155	418,767 ± 18,625	432,800 ± 36,135	418,192 ± 74,685		
	Room	60	453 ± 211	1,699 ± 216	4,971 ± 2,486	4,083 ± 2,704		
		120	346 ± 46	2,979 ± 471	8,970 ± 4,418	7,328 ± 4,916		
	Perilla seed	0 day		60	752 ± 131	238,553 ± 16,857	605,498 ± 118,519	449,853 ± 61,547
				120	1,154 ± 144	392,666 ± 22,930	967,260 ± 178,860	723,872 ± 93,621
After 3 months		Dark room	60	583 ± 218	247,568 ± 22,783	264,983 ± 51,959	255,777 ± 29,045	
		120	742 ± 273	366,098 ± 52,363	425,376 ± 52,161	407,470 ± 59,141		
Room		60	564 ± 292	19,652 ± 9,411	46,433 ± 28,998	22,995 ± 1,653		
		120	743 ± 518	34,450 ± 16,606	79,345 ± 48,452	42,472 ± 2,913		
After 12 months		Dark room	60	636 ± 210	233,729 ± 34,567	232,248 ± 27,016	252,689 ± 22,544	
		120	857 ± 104	387,382 ± 53,883	384,586 ± 50,358	404,069 ± 54,976		
Room		60	502 ± 231	23,676 ± 6,615	47,031 ± 31,042	33,397 ± 11,646		
		120	566 ± 339	43,540 ± 8,346	81,955 ± 49,999	55,843 ± 19,804		

10 kGy. The results of the effect of gamma irradiation on the sesame and perilla seeds themselves showed that the number of radiation-induced PPSL signals significantly increased with a irradiation dose up to 5 kGy. On the other hand, a slight decrease in the accumulated PPSL signals at a dose of 10 kGy as compared to 5 kGy was observed. Detection of irradiation by the PPSL system is based on higher radiation-induced PPSL signal than that of unirradiated sample. Therefore, although the signal of 5 to 10 kGy was non-linear, detection whether the sample has been irradiated or not was possible because of the higher signals of irradiated sample than that of unirradiated sample. These results are consistent with the literature (9), which has shown that the accumulated PPSL signals of irradiated Chinese sesame and perilla seeds, and Sudanese sesame seed themselves measured immediately after irradiation were higher than that of unirradiated ones and linearly increased up to 5 kGy, afterward, slightly decreased up to 10 kGy. However, the accumulated PPSL signals of Chinese and Sudanese sesame seeds were lower than that of Korean sesame seed. In the case of perilla seed, showed a contrary tendency, showing higher the accumulated PPSL signals than those of Chinese and Sudanese perilla seeds. As

shown in several papers on sesame and perilla seeds, generally, minerals, mixed in harvest stage, exist with the sesame and perilla seeds (6,9,12). Hence, this difference between samples was guessed due to differences of composed minerals mass, which showed higher radiation-induced PPSL signal owing to properties trapped greater radiation energy than in organic compounds. Slightly lower PPSL signal in the sample treated at 10 kGy than 5 kGy was guessed due to a radiation damage explained in study of Oduko and Spyrou (19). They reported that TL response of 20 kGy-irradiated chicken bone powder was lower than that of 15 kGy, possibly due to radiation damage, which was known to cause a decrease in TL response at high-absorbed dose for some TL materials, to the material. There were no significant differences in accumulated PPSL signals in unirradiated sesame and perilla seed samples. Threshold level, which can distinguish unirradiated sample from irradiated sample, was below  $1,261 \pm 157$  photon counts regardless of sample species tested, storage conditions and periods during 12 months. Based on the above results, since irradiated samples showed higher accumulated PPSL signal than the unirradiated samples under all conditions, detection of irradiation was possible by PPSL mea-

**Table 2.** The changes of accumulated PPSL signals of separated from irradiated sesame seed measured by PPSL with different sample amounts in the 60 and 120 s measurement times during storage of 12 months under room and darkroom conditions

(unit: photon counts)

Samples	Storage periods	Storage conditions	Measurement times (sec)	Irradiation dose (kGy)				
				0	1	5	10	
1	0 day		60	8,517 ± 990	1,145,408 ± 155,222	1,563,196 ± 584,968	985,793 ± 171,004	
			120	13,981 ± 2,615	1,562,917 ± 292,933	2,840,431 ± 340,867	1,539,354 ± 330,573	
	After 3 months	Dark room	60	7,927 ± 1,055	783,633 ± 47,750	1,107,565 ± 229,663	1,017,519 ± 54,474	
			120	14,032 ± 1,765	1,293,495 ± 101,887	1,870,915 ± 330,460	1,674,116 ± 66,735	
		Room	60	4,463 ± 2,463	468,875 ± 92,223	1,177,220 ± 153,546	747,592 ± 296,916	
			120	7,889 ± 4,368	780,219 ± 155,635	1,742,603 ± 511,966	1,224,102 ± 505,022	
	After 12 months	Dark room	60	7,301 ± 647	567,518 ± 16,263	1,181,800 ± 118,467	904,882 ± 88,373	
			120	12,848 ± 1,281	955,308 ± 278,975	1,932,415 ± 177,360	1,496,076 ± 129,714	
		Room	60	2,136 ± 498	190,389 ± 81,844	563,364 ± 4,199	292,070 ± 30,717	
			120	3,589 ± 1,071	311,694 ± 130,618	923,616 ± 13,064	487,974 ± 49,610	
	5	0 day		60	27,276 ± 4,380	1,960,782 ± 434,701	4,582,919 ± 188,672	4,673,016 ± 553,548
				120	48,382 ± 7,530	2,981,029 ± 128,539	7,712,995 ± 275,931	8,291,921 ± 2,048,235
After 3 months		Dark room	60	26,253 ± 12,357	1,619,125 ± 264,197	4,175,147 ± 999,680	3,820,305 ± 658,388	
			120	38,841 ± 8,383	2,761,521 ± 423,944	7,290,068 ± 2,252,371	6,385,943 ± 1,097,431	
		Room	60	16,885 ± 5,784	1,194,081 ± 219,442	2,701,222 ± 518,203	2,520,786 ± 759,668	
			120	30,038 ± 10,222	2,018,997 ± 351,201	4,578,086 ± 877,968	4,352,799 ± 1,122,330	
After 12 months		Dark room	60	17,841 ± 2,164	1,752,949 ± 119,131	4,517,200 ± 271,790	3,763,358 ± 199,553	
			120	31,752 ± 3,787	2,967,513 ± 181,249	7,708,083 ± 373,793	6,330,511 ± 329,079	
		Room	60	13,648 ± 6,343	855,345 ± 37,354	1,906,035 ± 560,986	1,166,123 ± 97,598	
			120	24,036 ± 11,179	1,438,966 ± 53,920	3,124,499 ± 949,045	1,949,051 ± 159,996	
10		0 day		60	44,723 ± 1,349	2,628,149 ± 695,775	8,257,229 ± 444,891	7,801,859 ± 743,488
				120	80,113 ± 3,198	4,208,246 ± 649,142	14,067,998 ± 909,629	13,775,553 ± 2,262,194
	After 3 months	Dark room	60	31,331 ± 11,261	2,241,449 ± 96,226	7,971,974 ± 2,011,044	6,017,703 ± 1,256,516	
			120	56,042 ± 20,477	3,812,144 ± 182,809	13,318,634 ± 3,397,355	10,116,310 ± 2,050,917	
		Room	60	30,923 ± 8,760	2,119,594 ± 335,413	5,746,684 ± 929,041	4,120,132 ± 538,454	
			120	55,901 ± 15,762	3,598,684 ± 558,497	9,739,919 ± 1,551,629	7,042,516 ± 921,679	
	After 12 months	Dark room	60	39,884 ± 10,838	2,132,585 ± 225,539	6,789,460 ± 206,453	5,276,331 ± 402,026	
			120	71,490 ± 19,719	3,794,892 ± 190,954	11,610,507 ± 395,106	9,102,290 ± 659,645	
		Room	60	18,317 ± 5,119	1,650,449 ± 217,343	2,778,119 ± 123,583	1,909,011 ± 299,563	
			120	31,982 ± 8,900	2,783,459 ± 376,085	4,604,733 ± 143,616	3,181,917 ± 507,682	

surement of sesame and perilla seeds themselves. Difference in accumulated PPSL signals between the sesame and perilla seeds was not significantly observed.

The accumulated PPSL signals of minerals separated from the irradiated and unirradiated Korean perilla and sesame seeds are shown in Table 2 and 3. The accumulated PPSL signals of the mineral measured exhibited an increase, showing decrease from 5 kGy to 10 kGy, with increasing irradiation dose, as shown in sesame and perilla seeds themselves, and mineral content of 1, 5 and 10 mg. This trend was similar in all storage conditions and periods. The accumulated PPSL signals of minerals separated were exceedingly greater than those of sesame and perilla seeds. This result indicates that the radiation-induced PPSL signals is much more trapped in the minerals separated than in organic compound.

Threshold levels, meaning accumulated PPSL signals of minerals separated from unirradiated sesame and perilla seeds, were  $80,113 \pm 3,198$  and  $58,827 \pm 21,417$  photon counts at 10 mg, respectively. Difference in the signals between minerals separated from Korean and Chinese sesame seeds, and Sudanese perilla seed was also observed as a discussion on

the accumulated PPSL signals of sesame and perilla seeds (9).

#### Decay rate of accumulated PPSL signals

If the PPSL response of irradiated materials is significantly greater than that of unirradiated materials, and if the fading of the PPSL response is low overlong-term storage, PPSL measurement may be suitable to determine whether foodstuffs have been irradiated (8). Therefore, decay rates of accumulated PPSL signals of unirradiated and irradiated sesame and perilla seeds themselves, and minerals separated from them were monitored during 12 months under different storage conditions. Table 4 shows that accumulated PPSL signals of irradiated sesame and perilla seeds were strongly influenced by the storage conditions. The accumulated PPSL signals of sesame seeds irradiated at 1, 5 and 10 kGy after 12 months resulted in the signal intensity falling to approximately 32.5, 60.2, and 52.8%, respectively, in darkroom condition and 99.9, 99.2 and 99.2%, respectively, in room condition compared to the initial values (0 day). This tendency according to storage conditions also was similarly observed in perilla seeds expect for decay rate (about 1.3%) at 1 kGy in darkroom condition.

In our previous work on PPSL (11), when marjoram irra-

**Table 3.** The changes of accumulated PPSL signals of separated from irradiated perilla seed measured by PPSL with different sample amounts in the 60 and 120 s measurement times during storage of 12 months under room and darkroom conditions (unit: photon counts)

Samples	Storage periods	Storage conditions	Measurement times (sec)	Irradiation dose (kGy)				
				0	1	5	10	
1	Control		60	$3,022 \pm 1,732$	$589,704 \pm 44,483$	$1,692,484 \pm 181,997$	$1,117,834 \pm 108,075$	
			120	$5,056 \pm 2,849$	$997,768 \pm 81,578$	$2,842,091 \pm 285,502$	$1,929,297 \pm 272,661$	
	After 3 months	Dark room	60	$6,750 \pm 1,470$	$562,489 \pm 53,975$	$1,371,977 \pm 459,487$	$846,958 \pm 86,303$	
			120	$12,161 \pm 2,582$	$1,008,534 \pm 71,059$	$2,319,337 \pm 765,969$	$1,425,443 \pm 218,247$	
		Room	60	$6,088 \pm 627$	$594,007 \pm 70,260$	$774,935 \pm 217,488$	$674,583 \pm 99,798$	
			120	$11,328 \pm 1,788$	$981,532 \pm 47,414$	$1,331,768 \pm 382,390$	$1,221,899 \pm 266,070$	
	After 12 months	Dark room	60	$2,723 \pm 1,552$	$574,555 \pm 44,818$	$1,338,544 \pm 292,669$	$714,001 \pm 91,285$	
			120	$4,714 \pm 2,842$	$1,024,251 \pm 101,826$	$2,272,031 \pm 520,697$	$1,223,873 \pm 147,106$	
		Room	60	$3,169 \pm 1,144$	$105,681 \pm 37,840$	$356,288 \pm 45,938$	$258,301 \pm 44,900$	
			120	$5,519 \pm 1,894$	$187,420 \pm 71,015$	$628,487 \pm 80,273$	$409,159 \pm 89,909$	
	5	Control		60	$18,053 \pm 10,195$	$2,357,409 \pm 232,837$	$5,672,295 \pm 349,104$	$3,660,351 \pm 311,625$
				120	$43,935 \pm 38,082$	$3,700,036 \pm 1,126,836$	$9,784,131 \pm 581,619$	$6,380,419 \pm 540,396$
After 3 months		Dark room	60	$18,999 \pm 6,403$	$2,197,906 \pm 163,928$	$3,158,364 \pm 320,094$	$2,191,033 \pm 880,079$	
			120	$34,173 \pm 11,070$	$3,853,697 \pm 266,968$	$5,451,004 \pm 597,091$	$3,841,156 \pm 1,524,454$	
		Room	60	$10,595 \pm 2,639$	$1,215,241 \pm 147,319$	$2,674,268 \pm 527,745$	$2,098,249 \pm 685,173$	
			120	$18,973 \pm 4,952$	$2,120,521 \pm 261,987$	$4,703,855 \pm 929,688$	$3,645,125 \pm 1,117,690$	
After 12 months		Dark room	60	$18,438 \pm 4,651$	$2,164,777 \pm 164,651$	$3,061,898 \pm 244,392$	$2,085,364 \pm 732,871$	
			120	$32,852 \pm 9,021$	$3,793,359 \pm 282,575$	$5,342,352 \pm 524,581$	$2,203,703 \pm 1,607,066$	
		Room	60	$4,548 \pm 1,017$	$586,050 \pm 179,504$	$977,922 \pm 85,614$	$738,880 \pm 92,093$	
			120	$7,255 \pm 483$	$1,215,589 \pm 14,322$	$1,762,861 \pm 133,255$	$1,420,107 \pm 84,205$	
10		Control		60	$33,383 \pm 12,545$	$3,532,351 \pm 266,902$	$8,641,921 \pm 449,464$	$6,087,570 \pm 291,582$
				120	$58,827 \pm 21,457$	$6,167,958 \pm 441,745$	$14,538,001 \pm 1,241,836$	$10,331,114 \pm 950,029$
	After 3 months	Dark room	60	$22,791 \pm 5,682$	$3,300,698 \pm 951,989$	$6,764,899 \pm 1,798,467$	$3,301,157 \pm 880,812$	
			120	$40,766 \pm 9,967$	$5,792,218 \pm 1,591,622$	$11,911,567 \pm 3,190,993$	$5,819,446 \pm 1,588,906$	
		Room	60	$13,767 \pm 3,300$	$2,437,195 \pm 395,664$	$4,862,256 \pm 304,202$	$1,750,338 \pm 285,064$	
			120	$24,487 \pm 5,986$	$4,319,030 \pm 689,882$	$8,525,910 \pm 495,102$	$3,108,802 \pm 490,332$	
	After 12 months	Dark room	60	$21,584 \pm 3,393$	$3,143,254 \pm 945,403$	$6,592,220 \pm 1,562,945$	$3,374,476 \pm 734,788$	
			120	$38,862 \pm 7,048$	$5,203,459 \pm 1,789,142$	$11,357,653 \pm 2,776,113$	$5,771,253 \pm 1,229,342$	
		Room	60	$14,587 \pm 3,108$	$1,252,573 \pm 91,236$	$2,466,877 \pm 184,103$	$942,560 \pm 154,291$	
			120	$25,223 \pm 5,473$	$2,235,666 \pm 152,331$	$4,353,789 \pm 168,068$	$1,478,305 \pm 138,604$	

**Table 4.** The changes of decay rate calculated from accumulated PPSL signals of irradiated sesame and perilla seeds measured by PPSL in the 60 and 120 s measurement times during storage of 12 months under room and darkroom conditions (unit: %)

Samples	Storage periods	Storage conditions	Measurement times	Irradiation dose (kGy)				
				0	1	5	10	
Sesame seed	0 day		60	0	0	0	0	
			120	0	0	0	0	
	After 3 months	Dark room	60	NC <sup>1)</sup>	62.4	29.8	23.6	
		120	NC	35.7	32.2	33.7		
		Room	60	NC	96.7	99.3	96.9	
			120	NC	97.7	99.3	97.0	
	After 12 months	Dark room	60	NC	30.1	57.7	44.8	
		120	NC	32.5	60.2	52.8		
		Room	60	NC	99.9	99.2	99.1	
			120	NC	99.5	99.2	99.2	
	Perilla seed	0 day		60	0	0	0	0
				120	0	0	0	0
After 3 months		Dark room	60	NC	ND <sup>2)</sup>	56.2	43.2	
		120	NC	6.8	56.0	43.7		
		Room	60	NC	91.7	92.3	94.9	
			120	NC	91.2	91.8	94.1	
After 12 months		Dark room	60	NC	2.0	61.7	43.8	
		120	NC	1.3	60.2	44.2		
		Room	60	NC	90.1	92.2	92.6	
			120	NC	88.9	91.5	92.3	

<sup>1)</sup>NC: Not calculated. <sup>2)</sup>ND: Not decreased.

diated at 5 kGy was stored during 24 weeks under room and darkroom conditions, the accumulated PPSL signals at 5 kGy resulted in the signal intensity falling to approximately 74% in room condition and 38.2% in darkroom condition of its former control level. The decreasing rate was about twofold under room conditions. These results agreed with the results in this paper. Also, we suggested that the cause of the greater decrease in accumulated PPSL signals under storage at room conditions was due to the difference in exposure periods to light. Because the storage under room conditions, which can be stimulated by sunlight and other light instead of infrared, after irradiation compared to darkroom conditions can create considerable higher instability followed by the emission of the radiation-induced PPSL photon counts trapped within the sample. Consequently, we think that this condition led to greater decrease in accumulated PPSL signals in sesame and perilla seeds as explained in the spices paper (11).

The changes in the decay rate calculated from accumulated PPSL signals of separated from irradiated sesame and perilla seeds measured by PPSL with different sample amounts in the 60 and 120 s measurement times during storage periods of 12 months under room and darkroom conditions are shown in Table 5 and 6. As shown in Table 4, decay rate of the accumulated PPSL signals decreased with increasing storage periods, and the sample stored in room condition showed higher reduction. The decay rate was less than sesame and perilla seeds and exhibited higher decrease in 5 and 10 kGy than in 1 kGy. Although, the accumulated PPSL signals of all sesame and perilla seeds decreased with in-

**Table 5.** The changes of decay rate calculated from accumulated PPSL signals of separated from irradiated sesame seed measured by PPSL with different sample amounts in the 60 and 120 s measurement times during storage of 12 months under room and darkroom conditions (unit: %)

Mineral contents	Storage periods	Storage conditions	Measurement times	Irradiation dose (kGy)				
				0	1	5	10	
1	0 day		60	0	0	0	0	
			120	0	0	0	0	
	After 3 months	Dark room	60	NC <sup>1)</sup>	31.6	29.2	ND <sup>2)</sup>	
		120	NC	17.3	34.1	ND		
		Room	60	NC	59.1	24.7	24.2	
			120	NC	50.1	38.7	20.5	
	After 12 months	Dark room	60	NC	50.5	24.4	8.2	
		120	NC	38.9	31.9	2.8		
		Room	60	NC	83.3	63.9	70.3	
			120	NC	80.1	67.5	68.3	
	5	0 day		60	0	0	0	0
				120	0	0	0	0
After 3 months		Dark room	60	NC	17.4	8.9	18.3	
		120	NC	7.3	5.5	22.9		
		Room	60	NC	39.1	41.1	46.1	
			120	NC	32.3	40.7	47.5	
After 12 months		Dark room	60	NC	10.6	1.4	19.5	
		120	NC	0.5	0.1	23.7		
		Room	60	NC	56.3	58.4	75.1	
			120	NC	51.7	59.5	76.5	
10		0 day		60	0	0	0	0
				120	0	0	0	0
	After 3 months	Dark room	60	NC	14.7	3.5	22.9	
		120	NC	9.4	5.3	26.6		
		Room	60	NC	19.3	30.4	47.2	
			120	NC	15.5	30.8	48.9	
	After 12 months	Dark room	60	NC	18.9	17.8	32.3	
		120	NC	9.8	17.5	33.9		
		Room	60	NC	37.2	66.4	75.5	
			120	NC	33.9	67.3	76.9	

<sup>1)</sup>NC: Not calculated. <sup>2)</sup>ND: Not decreased.

creasing storage times, irradiated samples showed higher photon counts than those of unirradiated samples in room and darkroom conditions. In both conditions, however, detection of irradiation was still possible after 12 months. These results indicate that PPSL can be used as a method for detecting the irradiation treatment by both sample forms, such as sesame and perilla seeds themselves and minerals separated, stored in different conditions during long-term storage.

#### Difference in accumulated PPSL signal with different measurement time

Difference in different measurement times (60 and 120 s) of accumulated PPSL signal measured by PPSL in irradiated sesame and perilla seeds and minerals separated stored under various storage conditions and periods during 12 months are shown in Fig. 1, 2 and 3. The accumulated PPSL signal of all the irradiated sesame and perilla seed samples and separated minerals measured for 120 s were higher than those measured for 60 s. In the all samples, the photon counts of minerals of all irradiated sesame and perilla seed samples

**Table 6.** The changes of decay rate calculated from accumulated PPSL signals of separated from irradiated perilla seed measured by PPSL with different sample amounts in the 60 and 120 s measurement times during storage of 12 months under room and dark-room conditions (unit: %)

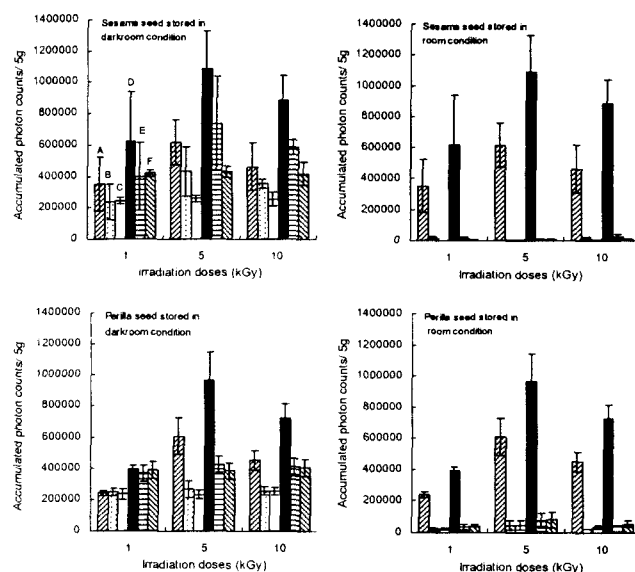
Mineral contents	Storage periods	Storage conditions	Measurement times	Irradiation dose (kGy)				
				0	1	5	10	
1	0 day		60	0	0	0	0	
			120	0	0	0	0	
	After 3 months	Dark room	60	NC <sup>1)</sup>	4.6	18.9	24.2	
		120	NC	ND <sup>2)</sup>	18.4	26.1		
	Room	60	NC	ND	54.2	39.7		
		120	NC	1.6	53.2	36.7		
	After 12 months	Dark room	60	NC	2.6	20.9	36.1	
		120	NC	ND	20.1	36.6		
	Room	60	NC	82.1	78.9	76.9		
		120	NC	81.2	77.9	78.8		
	5	0 day		60	0	0	0	0
				120	0	0	0	0
After 3 months		Dark room	60	NC	6.8	44.3	40.2	
		120	NC	ND	44.3	39.8		
Room		60	NC	48.5	52.9	42.7		
		120	NC	42.7	51.9	42.9		
After 12 months		Dark room	60	NC	8.2	46.0	43.0	
		120	NC	ND	45.4	65.5		
Room		60	NC	75.2	82.8	79.8		
		120	NC	67.2	81.9	77.7		
10		0 day		60	0	0	0	0
				120	0	0	0	0
	After 3 months	Dark room	60	NC	6.6	21.7	45.8	
		120	NC	6.1	18.1	43.7		
	Room	60	NC	31.0	43.7	71.3		
		120	NC	29.9	41.3	69.9		
	After 12 months	Dark room	60	NC	11.0	23.7	44.6	
		120	NC	15.6	21.9	44.1		
	Room	60	NC	64.5	71.5	84.5		
		120	NC	63.8	70.1	85.7		

<sup>1)</sup>NC: Not calculated. <sup>2)</sup>ND: Not decreased.

were higher than those of the unirradiated ones regardless of measurement times of 60 and 120 s. Hence, the authors believe that the detection of irradiation regardless of difference in sample forms, perilla and sesame seeds themselves and separated minerals, is possible in both 60 s and 120 s measurement time. These results are similar to our previous reports on imported perilla and sesame seeds (9), corn powder, shrimp-taste seasoning powder (10), spices (11) and irradiated foodstuffs such as cereals, beans and starches (8).

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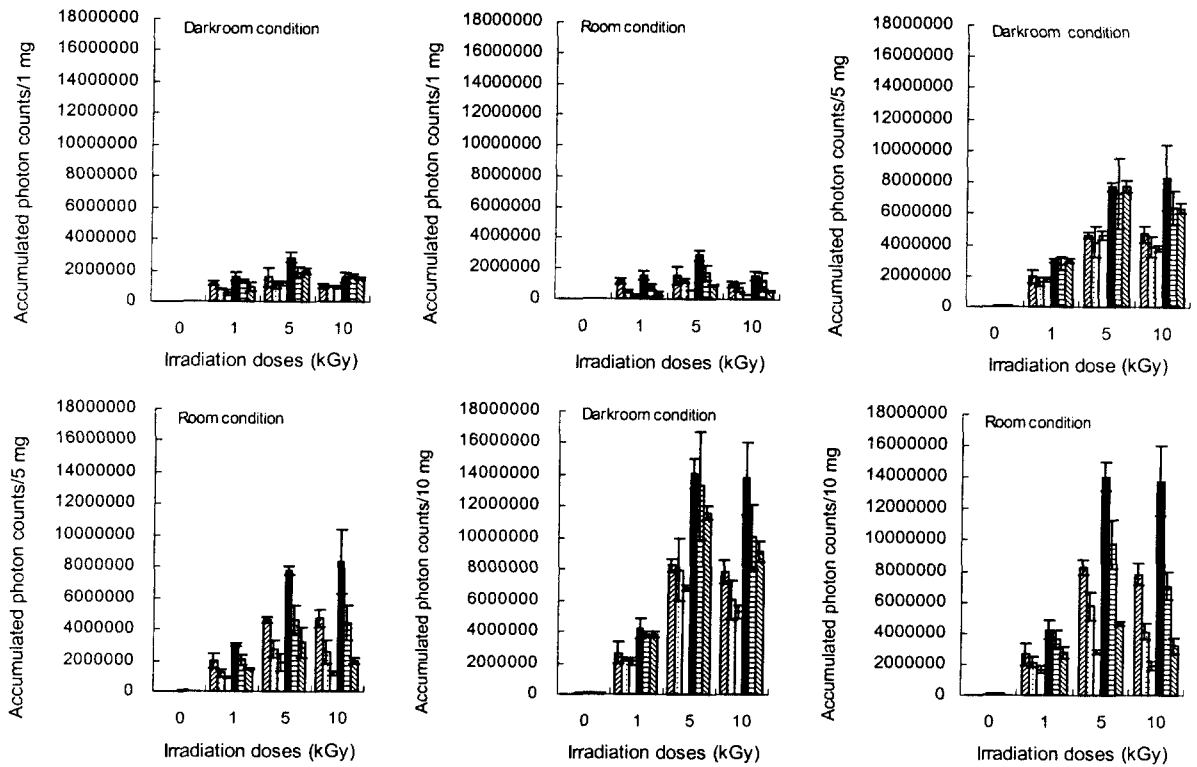


**Fig. 1** Difference in measurement times of accumulated PPSL signals measured by PPSL in irradiated sesame and perilla seeds stored under different storage conditions and periods.

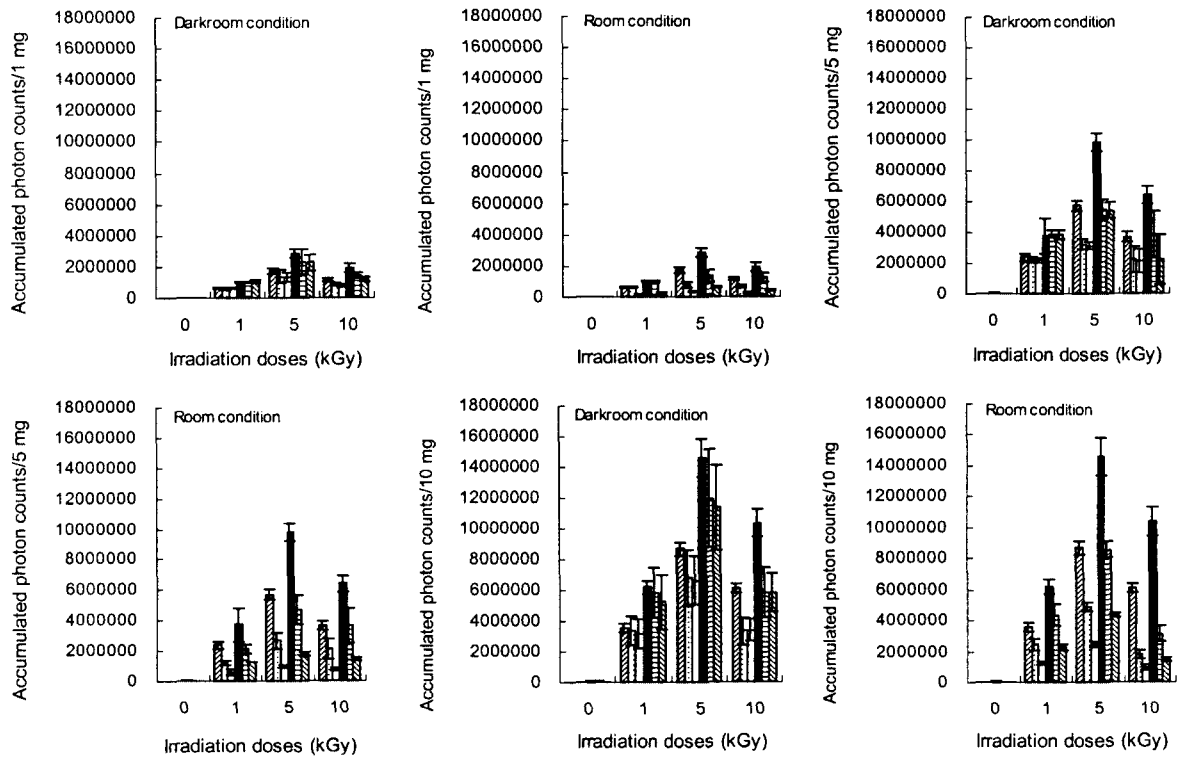
- Accumulated PPSL signals of sample measured immediately after irradiation for 60 s.
- ▨ Accumulated PPSL signals of sample measured at 3 months after irradiation for 60 s.
- ▩ Accumulated PPSL signals of sample measured at 12 months after irradiation for 60 s.
- Accumulated PPSL signals of sample measured immediately after irradiation for 120 s.
- ▨ Accumulated PPSL signals of sample measured at 3 months after irradiation for 120 s.
- ▩ Accumulated PPSL signals of sample measured at 12 months after irradiation for 120 s.

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**Fig. 2.** Difference in measurement times and sample amount tested of accumulated PPSL signals measured by PPSL in minerals separated from irradiated sesame seeds stored under different storage conditions and periods.   
 ▨, □, ◻, ■, ▩, ▪: Refer to the legend in Fig. 1.



**Fig. 3.** Difference with measurement times and sample amount tested of accumulated PPSL signals measured by PPSL in minerals separated from irradiated perilla seeds stored under different storage conditions and periods.   
 ▨, □, ◻, ■, ▩, ▪: Refer to the legend in Fig. 1.

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