J. Food Sci. Nutr.
Vol. 6, No. 3, p. 152~157 (2001)

Detection of Pulsed Photostimulated Luminescence Signals Emitted by Infrared Stimulation of Irradiated Spices during Storage under Two Conditions

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Abstract

Accumulated photon counts in immediate measurement after irradiation of marjoram, basil and thyme were shown to be below 625±162, 577±178 and 1261±640 Pc, respectively. The accumulated photon counts increased linearly with increasing irradiation doses up to 5 kGy and slightly increased from 5 kGy to 10 kGy. This trend was similar after storage periods. According to storage conditions, the difference of the accumulated photon counts was not clearly observed. The accumulated photon counts of irradiated spice samples decreased with increasing storage periods. The rate of decrease was higher in 5 and 10 kGy irradiated samples than that in 1 kGy, and in room conditions than that in darkroom conditions. The photon counts of the irradiated spice samples measured for 120 s were higher than those measured for 60 s. The irradiated spice samples showed higher photon counts than those of unirradiated samples in both room and darkroom conditions during all the storage periods. These results indicate that detection of irradiation was still possible after 24 weeks, although the PPSL signal of all spice samples decreased with increasing storage times.

Key words: spices, irradiation, pulsed photostimulated luminescence (PPSL)

INTRODUCTION

Irradiation can be used to achieve the safety of food through the reduction of pathogenic microorganisms and by reducing those organisms which cause food spoilage related to a decrease in the shelf life of foods (1). The irradiation also can be applied to inhibit the metabolic processes leading to ripening and spoilage of high value food products such as exotic fruits, vegetables and spices (2). Therefore, if only consumer acceptance of irradiated foods was reconsidered, it would be very useful technology in food hygiene and could be applied to various foods for the health of consumers and benefit of the food industry. As a means to improve such consumer acceptance, establishment of detection methods for irradiated foods are considered to play a very important role in contributing to a belief in the safety of irradiated foods.

Among many detection methods, pulsed photostimulated luminescence (PPSL) is one method which has the highest application capability. The simplicity of the procedure of this detection method is such that even a beginner can use it very easily (3-6). In addition, the cost of the apparatus is relatively very cheap compared with other detection apparatus such as thermoluminescence (TL) (7-9) and electron paramagnetic resonance (ESR) (10-12). But, one of problems not yet examined for utilization of PPSL is stability under various storage conditions of the signal emitted by infrared stimulation. Although previous detection studies using PPSL were carried out on many irradiated foods such as bown shrimp (13), white ginseng powder (14), pepper powder, dried herbs, fresh shrimp, potato, soybean, dried fig, chestnut, dried squid, dried cod (15), cereals, starches, beans (4), corn powder (5) and sesame, and perilla seeds (3), these results are based on data measured immediately after irradiation and there have been no studies conducted on irradiated foods in long term storage under various conditions.

Thus, to investigate the potential of using PPSL in the identification of irradiated spices such as basil, marjoram and thyme after long-term storage, this paper described changes in photon counts emitted by infrared stimulation of irradiated samples under various storage conditions.

MATERIALS AND METHODS

Materials and irradiation

Basil, marjoram and thyme harvested in U.S.A. were purchased from a local supplier. Samples were packed in polyethylene bags (50 g), split into two portions (room and darkroom conditions) and irradiated using a Co-60 irradiator (AECL, Canada) with 1, 5, and 10 kGy at the Korea Atomic Energy Research Institute. The dose rate was 1 kGy/h. After irradiation, the samples under darkroom conditions were stored in a chamber oven (K.M.C-1203P3, Vision Scientific Co., LTD, Seoul, Korea) for 24 weeks (from April to September) to block exposure by a light at room temperature. The samples at room conditions were stored under usual laboratory conditions. To measure the exact total absorbed dose of gamma irradiation, the dose rates for Co-60 sources were determined using a ceric-cerous dosimeter.

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Measurement of pulsed photostimulated luminescence
The PPSL system (serial ; 0021, SURRC ; Scottish Universities Research and Reactor Center, UK) that is composed of a control unit, sample chamber, and detector head assembly was used in this experiment. The control unit contains a stimulation source that is comprised of an array of infrared light emitting diodes. The diodes are pulsed symmetrically on and off for equal periods. PPSL is detected by a bialkali cathode photomultiplier tube operating in photon counting mode. Optical filtering was used to define both the stimulation and detection wavebands. The samples (5 g) were introduced in 50 mm diameter disposable petri dishes (Bibby Sterilin type 122) with no other preparation and measured in the sample chamber for 60 and 120 s. The photon counts of the samples were recorded in the measuring mode (16). The PPSL measurement was performed in triplicate under the same laboratory and instrumental conditions.

RESULTS AND DISCUSSION

Threshold levels and difference of accumulated photon counts among the spice samples
To observe the difference in accumulated photon counts according to storage conditions, this experiment was carried out under room conditions (laboratory conditions) with light and darkroom conditions without light. The accumulated photon counts were obtained by directly measuring 5 g of spice-sample with no other preparation using a PPSL system. In case of zero time measured immediately after irradiation (during 120 s), accumulated photon counts for marjoram, basil and thyme were showed below 625±162, 577±178 and 1261±640 Pc, respectively. According to storage conditions, the difference of the accumulated photon counts was not clearly observed among the unirradiated control samples regardless of the storage times and conditions. Hence, we think that the accumulated photon count values in the unirradiated control samples indicated threshold levels between unirradiated and irradiated spices such as marjoram, basil and thyme. The accumulated photon counts of the irradiated marjoram, basil and thyme measured (during 120 s) immediately after irradiation at 1 kGy in darkroom conditions were 302,038±41,737, 67,206±10,186 and 118,940±2,457 Pc, respectively, and showed clear differences in the accumulated photon counts among the samples. This phenomena was hypothesized due to the difference in mineral content of the spice samples as shown from our previous data showing higher accumulated photon counts in minerals separated from Chinese perilla, sesame seeds and Sudanese sesame seeds than in the perilla and sesame seeds themselves (17).

Changes of accumulated photon counts and decay rate according to storage conditions and irradiation doses
Changes of accumulated photon counts and decay rate of irradiated marjoram according to storage conditions are shown in Table 1 and Fig. 1. The photon counts increased linearly with increasing irradiation doses up to 5 kGy and from 5 kGy to 10 kGy, slightly increased. This trend was similar to other storage times. Changes of accumulated photon counts according to storage conditions were also observed. The accumulated photon counts decreased with in

Table 1. The changes of accumulated photon counts and decay rate of unirradiated and irradiated marjoram according to storage conditions and periods (unit: P. C. = photon counts, D. R. = %)

Storage periods & conditions		Measurement time (s)	Irradiation dose (kGy)							
			Control[1)]		1		5		10	
			P. C.	D. R.[2)]	P. C.	D. R.	P. C.	D. R.	P. C.	D. R.
Zero time	Room	60	291±73[3)]	NC[4)]	172,722±5,528	NC	453,731±42,563	NC	483,899±19,075	NC
		120	373±41	NC	277,244±10,321	NC	700,496±71,026	NC	748,752±34,632	NC
	Darkroom	60	538±40	NC	187,267±25,721	NC	494,035±107,115	NC	591,341±14,048	NC
		120	446±92	NC	302,038±41,737	NC	763,752±158,124	NC	899,752±18,786	NC
After 1 week	Room	60	393±171	NC	141,942±10,354	17.8	332,901±28,973	26.6	414,713±99,015	14.3
		120	407±111	NC	227,408±15,715	17.9	524,378±43,339	25.1	639,274±139,628	14.6
	Darkroom	60	437±104	NC	189,961±19,851	0.0	392,632±34,671	20.5	466,608±31,789	21.1
		120	625±162	NC	298,285±28,474	1.3	684,588±130,548	10.3	734,559±46,367	18.3
After 4 weeks	Room	60	408±25	NC	157,550±17,618	8.7	328,089±24,563	27.7	380,762±60,548	21.3
		120	457±87	NC	250,793±25,533	9.5	508,841±40,054	27.3	586,023±87,327	21.7
	Darkroom	60	443±97	NC	142,824±14,260	23.7	421,379±24,289	14.7	473,698±24,178	19.9
		120	431±57	NC	230,407±20,641	23.7	652,325±39,143	14.6	713,014±73,252	20.7
After 12 weeks	Room	60	547±73	NC	105,402±22,110	38.9	174,852±22,610	61.5	196,928±16,257	59.3
		120	468±86	NC	172,021±37,105	37.9	267,000±44,768	61.9	306,448±41,004	59.1
	Darkroom	60	522±187	NC	182,246±16,311	2.7	339,736±16,950	31.2	465,672±58,584	21.3
		120	513±185	NC	289,325±26,856	4.2	531,045±29,950	30.5	677,007±69,272	24.8
After 24 weeks	Room	60	273±55	NC	76,195±14,179	55.9	168,702±10,109	62.8	175,364±13,591	63.7
		120	391±91	NC	119,717±25,123	56.8	265,932±21,971	62.0	273,900±23,644	63.4
	Darkroom	60	413±281	NC	147,996±7,346	20.9	262,488±5,096	53.1	309,246±30,481	52.3
		120	336±274	NC	239,068±13,176	20.8	431,579±96,480	43.5	483,566±45,714	46.3

[1)]Unirradiated sample. [2)]Decay rate. [3)]Means±standard deviation for 3 measurements. [4)]Sample not calculated.

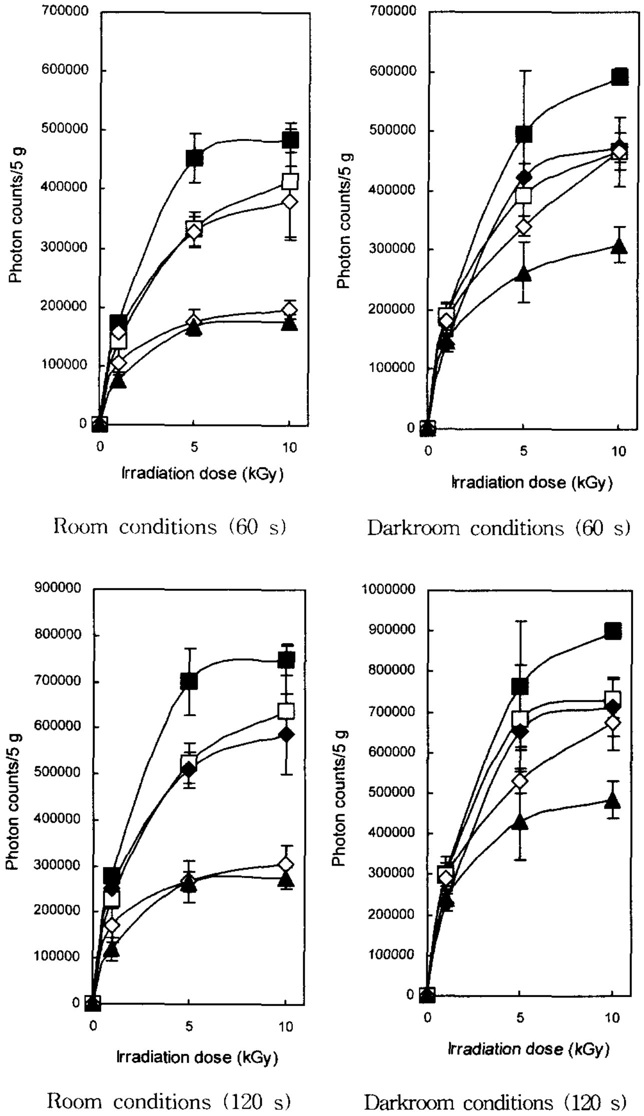

Fig. 1. The differences of accumulated photon counts of irradiated marjoram measured by PPSL during the 60 and 120 s.
■-■ : Control (sample measured immediately after irradiation), □-□ : sample measured after 1 week, ◆-◆ : sample measured after 4 weeks, ◇-◇ : sample measured after 12 weeks, ▲-▲ : sample measured after 24 weeks.

creasing storage times and exhibited a greater decrease in 5 and 10 kGy than in 1 kGy, and in room conditions than in darkroom conditions. The accumulated photon counts with storage times were strongly influenced by the storage conditions. The accumulated photon counts measured for 120 s of marjoram stored during 24 weeks in room conditions after irradiated at 5 kGy resulted in the signal intensity falling to approximately 74% (from its former control level) showing a larger decrease of about twofold compared with 38.2% in darkroom conditions. This tendency to change according to storage conditions was also observed in both samples (basil and thyme) and irradiation doses. Since storage under room conditions after irradiation compared to darkroom conditions showed considerable higher instability due to a greater emittance of the radiation-induced PPSL photon counts, which were trapped within the sample by irradiation, due to stimulation by sunlight and other light existing under room conditions instead of infrared, the reason for the greater decrease in photon counts under storage at room conditions was possibly due to the difference in the exposure periods to light. Consequently, it is proposed that this condition led to a greater decrease in accumulated photon counts. As can be seen from Table 2 and 3, and Fig. 2 and 3, the PPSL signal strength of irradiated basil and thyme stored under darkroom conditions was significantly higher than for those kept under room conditions and increased with increasing irradiation doses as indicated in marjoram. Although, the PPSL signal of all spice samples decreased with increasing storage times, irradiated samples showed higher photon counts than those of unirradiated samples in room and darkroom con-

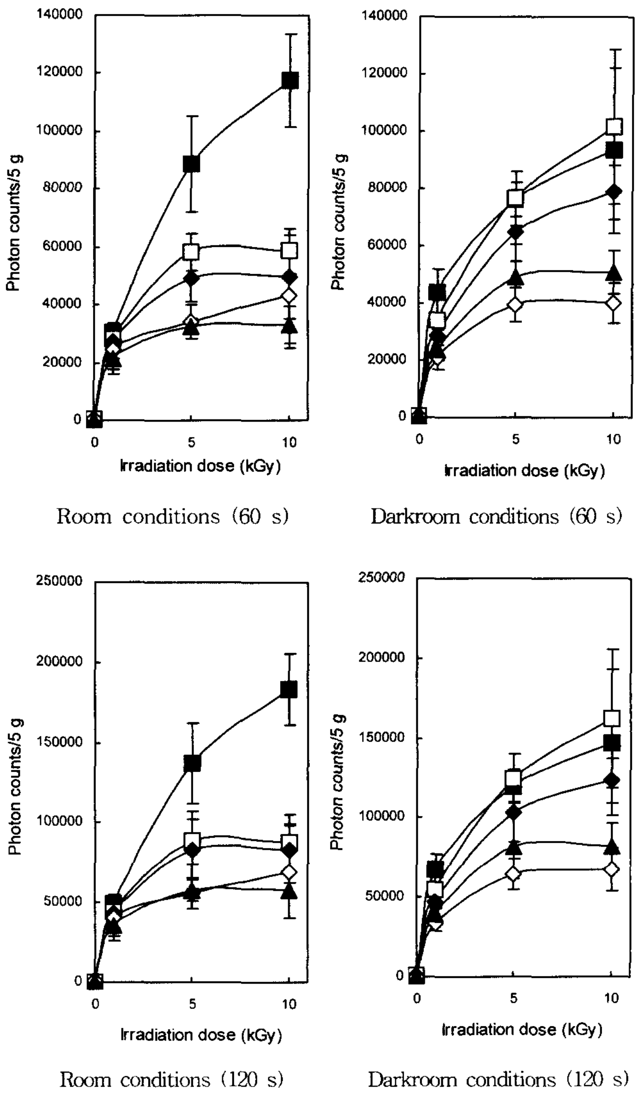

Fig. 2. The differences of accumulated photon counts of irradiated basil measured by PPSL during the 60 and 120 s.
■-■, □-□, ◆-◆, ◇-◇, ▲-▲ : Refer to the legends in Fig. 1.

Table 2. The changes of accumulated photon counts and decay rate of unirradiated and irradiated basil according to storage conditions and periods (unit: P. C. = photon counts, D. R.=%)

Storage periods & conditions		Measurement time (s)	Irradiation dose (kGy) Control[1)] P. C.	D. R.[2)]	1 P. C.	D. R.	5 P. C.	D. R.	10 P. C.	D. R.
Zero time	Room	60	392±148[3)]	NC[4)]	30,644±3,230	NC	88,662±16,378	NC	117,449±15,893	NC
		120	439±87	NC	49,624±4,609	NC	137,331±25,257	NC	183,153±22,140	NC
	Darkroom	60	369±192	NC	43,796±8,160	NC	76,163±6,034	NC	93,362±28,998	NC
		120	356±134	NC	67,206±10,186	NC	119,937±9,913	NC	147,089±45,608	NC
After 1 week	Room	60	416±7	NC	28,199±3,349	8.0	58,176±6,287	34.3	58,656±7,815	50.1
		120	450±55	NC	44,641±4,888	10.1	88,482±13,676	35.6	87,932±17,269	51.9
	Darkroom	60	417±128	NC	34,277±7,202	21.7	76,652±9,381	ND[5)]	101,503±27,177	ND
		120	564±201	NC	54,912±11,606	18.3	124,449±15,493	ND	162,184±43,276	ND
After 4 weeks	Room	60	312±80	NC	27,380±6,185	10.7	49,176±7,799	44.5	49,878±14,364	57.5
		120	406±101	NC	43,194±9,433	12.9	82,810±24,214	39.7	83,489±15,191	54.4
	Darkroom	60	385±57	NC	28,862±1,721	34.1	65,008±10,623	14.7	78,750±9,413	15.7
		120	501±189	NC	47,040±2,831	30.0	103,607±18,867	13.6	123,075±13,560	16.3
After 12 weeks	Room	60	476±99	NC	24,596±8,614	19.8	34,120±5,887	61.5	43,088±18,172	63.3
		120	577±178	NC	40,556±14,382	18.3	55,808±9,912	59.4	69,920±29,846	61.8
	Darkroom	60	448±134	NC	21,276±4,313	51.4	39,437±5,816	48.2	40,084±7,174	57.1
		120	326±233	NC	33,564±4,282	50.1	64,921±9,681	45.9	67,321±13,459	54.2
After 24 weeks	Room	60	242±56	NC	21,394±4,003	30.2	32,555±2,702	63.3	33,142±6,272	71.7
		120	248±75	NC	35,696±6,287	28.1	58,278±6,744	57.6	57,790±4,815	68.5
	Darkroom	60	295±257	NC	23,704±3,720	45.9	49,211±11,094	35.4	50,882±7,570	45.5
		120	350±667	NC	39,971±4,064	40.5	82,105±20,118	31.6	82,380±13,906	44.0

[1)~4)]Refer to the legends in Table 1. [5)]Sample not decreased.

Table 3. The changes of accumulated photon counts and decay rate of unirradiated and irradiated thyme according to storage conditions and periods (unit: P. C. = photon counts, D. R. = %)

Storage periods & conditions		Measuremen time (s)	Irradiation dose (kGy) Control[1)] P. C.	D. R.[2)]	1 P. C.	D. R.	5 P. C.	D. R.	10 P. C.	D. R.
Zero time	Room	60	555±102[3)]	NC[4)]	69,755±22,311	NC	228,181±5,348	NC	293,171±36,586	NC
		120	1,261±640	NC	116,365±21,909	NC	340,540±12,431	NC	433,758±57,537	NC
	Darkroom	60	430±65	NC	76,398±1,185	NC	231,286±26,934	NC	246,771±34,404	NC
		120	536±122	NC	118,940±2,457	NC	340,755±23,714	NC	360,693±36,824	NC
After 1 week	Room	60	560±62	NC	78,832±16,017	ND[5)]	159,586±13,251	30.1	176,509±29,036	39.8
		120	1,183±546	NC	119,934±12,492	ND	249,376±23,259	26.8	276,868±43,987	36.2
	Darkroom	60	299±134	NC	88,446±9,863	ND	193,005±22,977	16.6	192,475±23,226	22.0
		120	657±141	NC	14,0288±16,701	ND	263,266±33,393	22.7	264,704±2,852	26.6
After 4 weeks	Room	60	560±62	NC	67,379±15,098	3.4	149,264±17,599	34.6	175,828±2,614	40.0
		120	1,187±482	NC	107,528±22,884	7.6	228,381±26,538	32.9	268,210±6,238	38.2
	Darkroom	60	440±229	NC	89,585±10,536	ND	183,355±22,737	20.7	208,998±2,979	15.3
		120	415±169	NC	144,018±16,701	ND	283,298±33,115	16.9	318,849±3,431	11.6
After 12 weeks	Room	60	611±109	NC	51,638±6,117	25.9	87,244±17,070	61.8	90,132±13,049	69.3
		120	964±145	NC	83,600±9,225	28.2	139,367±9,823	59.1	142,417±20,048	67.2
	Darkroom	60	565±125	NC	59,900±6,868	21.6	116,040±7,882	49.8	117,233±14,029	52.5
		120	672±112	NC	97,691±9,878	17.9	195,998±31,111	42.5	188,524±26,806	47.7
After 24 weeks	Room	60	492±170	NC	33,344±4,905	52.2	55,162±2,416	75.8	62,536±18,536	78.7
		120	579±63	NC	55,185±8,086	52.6	88,563±4,894	74.0	106,956±36,051	75.3
	Darkroom	60	598±181	NC	57,687±12,308	24.5	133,435±8,899	42.3	135,319±15,965	45.2
		120	815±284	NC	98,488±41,182	17.2	210,565±13,370	38.2	209,180±19,812	42.0

[1)~4)]Refer to the legends in Table 1. [5)]Sample not decreased.

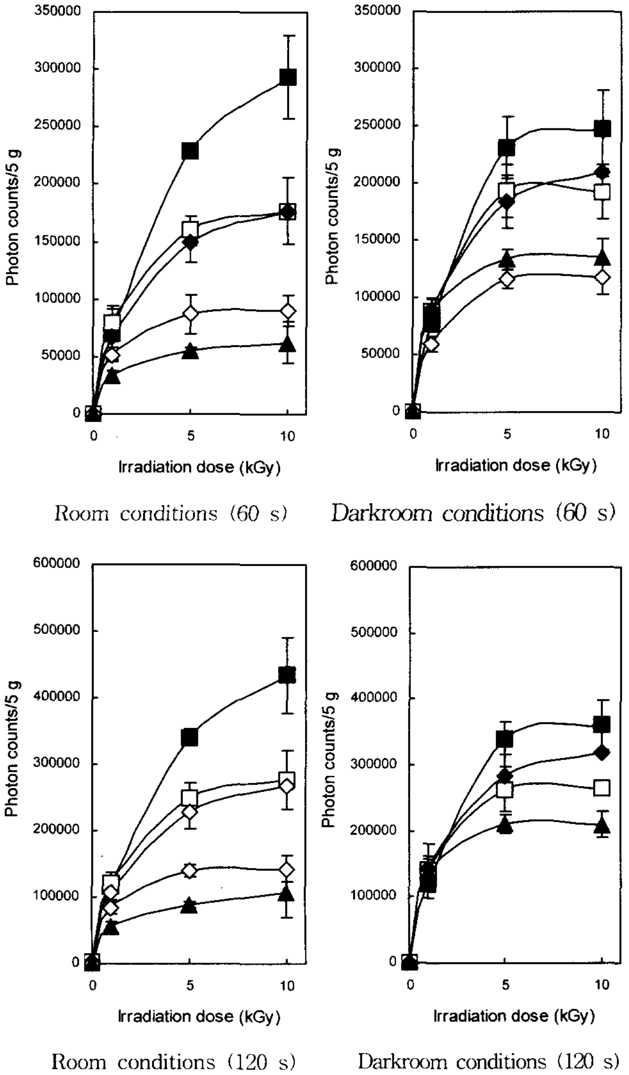

Fig. 3. The differences of accumulated photon counts of irradiated thyme measured by PPSL during the 60 and 120 s. ■-■, □-□, ◆-◆, ◇-◇, ▲-▲: Refer to the legends in Fig. 1.

ditions, in both conditions, detection of irradiation was still possible after 24 weeks. Therefore, PPSL can be proposed as a method for detecting the irradiation treatment of irradiated spices such as marjoram, basil and thyme.

Similar results for PPSL have been reported. Sanderson et al. (6) reported that the photon counts of irradiated samples were higher than unirradiated ones. Yi et al. (5) reported that the differences in photon counts according to storage conditions (room and darkroom) were clearly observed and in darkroom conditions, a significant decrease in the photon count of corn powder was not observed, but the photon count after one month in room conditions was barely observable. The above result under darkroom conditions agreed with our result, but those at room conditions disagreed. This difference in the room conditions was assumed to be due to difference of mineral contents (17) and general components in spices and corn powder.

Influence of measurement time

The photon counts of the marjoram, basil and thyme measured during 60 and 120 s exhibited an increase with increasing irradiation dose. Also, the photon count of the spice samples measured during 120 s were higher than those measured for 60 s. In all samples, the photon counts of the spice samples were higher than those of the unirradiated ones in samples measured immediately after irradiation (control). In both 60 s and 120 s measurement time, difference in decay rate of irradiated spice samples according to increasing storage times, irradiation doses and different storage conditions was not clearly observed. Hence, the authors believe that detection of the spices such as marjoram, basil and thyme is possible in both 60 s and 120 s measurement times.

Several papers (3-5,17) reported that the photon counts of irradiated samples measured for 60 and 120 s exhibited an increase with increasing irradiation dose and also, the photon count measured for 120 s were higher than those measured for 60 s. Our results also agree with the several papers.

ACKNOWLEDGEMENTS

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

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(Received May 9, 2001)