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The Application of a Pulsed Photostimulated Luminescence (PPSL) Method for the Detection of Irradiated Foodstuffs

Sang-Duk Yi[†] and Jae-Seung Yang

Detection Lab. of Irradiated Food, Korea Atomic Energy Research Institute, Taejon 305-353, Korea

Abstract

The properties of pulsed photostimulated luminescence (PPSL) were measured to use as basis data for the detection of irradiated foodstuffs (34 different foods). Samples were packed in polyethylene bags and irradiated at 1, 5, and 10 kGy with a dose rate of 10 kGy/h. The samples irradiated were introduced in the sample chamber without other preparation and measured PPSL photon counts for 60 and 120 s. The PPSL photon counts of the irradiated samples were higher than the unirradiated, increased with increasing irradiation dose, and showed a good relationship between irradiation doses and photon counts in a multinomial expression. These results suggest that the detection of irradiated foodstuffs was possible by PPSL. Therefore, PPSL can be proposed as the method for the detection of irradiated foodstuffs.

Key words: pulsed photostimulated luminescence (PPSL), detection method, gamma irradiation

INTRODUCTION

Optically stimulated luminescence (OSL) has gained importance after its application as a method of accumulated dose determination in the dating of quartz extracted from sediments (1) and demonstrated that transient luminescence could be stimulated in feldspars using infrared light in the wavelength of 530~1030 nm and measured a stimulation spectrum with a complex structure with several peaks (2,3). Following this study, a great number of studies have been carried out both on the technique and its applications relevant to dating (4-12). Sanderson and Clark were the first to investigate the potential of pulsed OSL from alkali feldspars relevant to dating and infrared stimulated pulsed PSL has been investigated in connection with the problem of detecting irradiated foods (4).

Pulsed photostimulated luminescence (PPSL), which uses light rather than heat to stimulate the release of trapped charge carriers, is radiation specific phenomena from energy storage by trapped charge carriers following irradiation (13,14). This detection method is designed to allow direct measurements for rapid screening purposes without the need for sample preparation or re-irradiation such as the TL of irradiated foods (15-17). When exposed to ionizing radiation, most materials store energy by trapping charge carriers at structural, interstitial or impurity sites. Subsequent stimulation by electromagnetic radiation of the appropriate wavelength releases the trapped charge carriers, resulting in the emission of photostimulated luminescence (PSL) (15,16). Therefore, the PPSL detection method has a possible application as a rapid screening purpose for many irradiated foods.

Based on this background, to use as a basis data for the detection of irradiated foodstuffs (34 different foods), this study was conducted on the properties of PPSL of those foodstuffs not yet examined in previous studies (16-19).

MATERIALS AND METHODS

Materials and irradiation

Unclean wheat rice, clean wheat rice, white wheat, dark northern spring, and durum were obtained from Mildawon Co. (Kongju, Chungnam). Corn, potato, and sweet potato starches (Dusan Co., Ich'on, Kyonggi, Korea), strong and weak flours (Cheiljedang Co., Seoul, Korea) and other samples used for this study were purchased from a local market. The samples were packed in polyethylene bags and were irradiated using a Co-60 irradiator (AECL, 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.

Measurement of pulsed photostimulated luminescence

The PPSL system (serial ; 0021, SURRC ; Scottish Universities Research and Reactor Center, UK) is composed of a control unit, sample chamber, and detector head assembly, and the control unit contains a stimulation source which is comprised of an array of infra-red light emitting diodes which 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 is used to define both the stimulation and detection wavebands. The samples (starches; 3 g, other 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 cham-

[†]Corresponding author. E-mail: ysduk@hanmail.net
Phone: 82-42-868-8064, Fax: 82-42-868-8064

ber for 60 and 120 s (20). The photon counts of the samples were recorded in the measuring mode.

Statistical analysis

Significant differences were determined by using Duncan's multiple range test ($p<0.05$) in a one-way ANOVA, and regression expressions and coefficients were determined through the regression analysis of the SPSS (Statistical Package for Social Science) version 7.5. All experiments were repeated three times.

RESULTS AND DISCUSSION

To investigate the capability of detection of irradiated foodstuffs by PPSL, the study was carried out on 34 different foods not yet examined in previous studies (16-19). Table 1 shows the changes of photon counts of irradiated starches with increasing dose levels. The photon counts of the unirradiated and irradiated potato and sweet potato starch measured (120 s) immediately after irradiation exhibited an increase from 992±209, and 1577±459, respectively, in the unirradiated control to 519136±97066, and 1252722±11633 in the irradiated sample at 10 kGy. The photon counts of corn starch measured for 120 s exhibited an increase from 494±118 in the unirradiated control to 129528±31231 in the irradiated sample at 5 kGy, but photon counts of corn starch in the 10 kGy slightly decreased rather than that in the 5 kGy.

The photon counts of irradiated perilla and sesame seeds measured by PPSL for 60 and 120 s are shown in Table 2. The photon counts exhibited an increase with increasing irradiation dose. Also, the photon counts of all the irradiated sesame and perilla seeds measured for 120 s were higher than those measured for 60 s. In all the samples, the photon counts of the samples were higher than the unirradiated ones.

Gamma irradiation is a useful technique for cereal preservation against insect infestation and microbial contamination as a dose to 5 kGy completely kills the spores (21). Therefore, gamma irradiation for cereals and detection technique for irradiated cereals will become increasingly important. As shown in Table 3, the photon counts of the irradiated cereals were higher than those of the unirradiated ones and the pho-

Table 1. The changes of accumulated photon counts of unirradiated and irradiated starches (Unit : Photon counts)

Sample	Measurement time (s)	Irradiation dose (kGy)				Regression expressions and coefficient
		Control[1)]	1	5	10	
Corn starch	60	494^{a} ±118	44229^{a} ±4222	167003^{b} ±40365	129528^{b} ±31231	$y=-4074x^2+53928x-2067.2$ $R^2=0.9988$
	120	591^{a} ±107	81837^{a} ±13902	251683^{b} ±54980	202250^{b} ±48742	$y=-6017.3x^2+80001x+3631.7$ $R^2=0.9992$
Potato starch	60	657^{a} ±99	153305^{b} ±5623	273586^{c} ±21577	335336^{d} ±59731	$y=144778x^{0.3898}$ $R^2=0.9995$
	120	992^{a} ±209	245798^{b} ±6402	424310^{c} ±31207	519136^{d} ±97066	$y=225986x^{0.3921}$ $R^2=0.9995$
Sweet potato starch	60	974^{a} ±237	299282^{b} ±12837	794200^{c} ±36393	826987^{c} ±8550	$y=335688x^{0.4235}$ $R^2=0.9984$
	120	1577^{a} ±459	484653^{b} ±23668	1222847^{c} ±46943	1252722^{c} ±11633	$y=523966x^{0.4205}$ $R^2=0.9986$

[1)]Control = unirradiated sample x: irradiation dose (kGy), y: photon counts
[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 ($p<0.05$).

Table 2. The changes of accumulated photon counts of unirradiated and irradiated sesame and perilla seeds (Unit : Photon counts)

Sample	Measurement time (s)	Irradiation dose (kGy)				Regression expressions and coefficient
		Control[1)]	1	5	10	
Sesame seed	60	933^{a} ±131	350056^{b} ±171867	614552^{b} ±141950	464374^{b} ±152188	$y=-15436x^2+192709x+72870$ $R^2=0.9193$
	120	1261^{a} ±157	620422^{b} ±315179	1088115^{b} ±239982	886135^{b} ±156303	$y=-26066x^2+334700x+131249$ $R^2=0.9194$
Perilla seed	60	752^{a} ±99	238553^{b} ±16857	605498^{c} ±118519	449853^{d} ±61547	$y=-15262x^2+194821x+25133$ $R^2=0.9908$
	120	1154^{a} ±144	392666^{b} ±22930	967260^{c} ±178860	723872^{d} ±93621	$y=-24288x^2+310243x+45340$ $R^2=0.9882$

[1)]Control = unirradiated sample x: irradiation dose (kGy), y. photon counts
[a-d]Means with the same superscripts in each row are not significantly different among group by Duncan's multiple range test in one way ANOVA ($p<0.05$).

Table 3. The changes of accumulated photon counts of unirradiated and irradiated cereals (Unit : Photon counts)

Sample	Measurement time (s)	Irradiation dose (kGy)				Regression expressions and coefficient
		Control[1]	1	5	10	
Glutinous sorghum	60	320^{a} ±108	60541^{b} ±23817	75948^{bc} ±29744	31586^{ab} ±3940	$y=-2434.1x^2+25751x+15769$ $R^2=0.7721$
	120	544^{a} ±194	100061^{b} ±26012	117763^{b} ±43288	51080^{c} ±6084	$y=-3737.4x^2+39460x+27248$ $R^2=0.7271$
Italian millet	60	704^{a} ±86	233127^{b} ±54898	275454^{b} ±115412	80073^{a} ±18335	$y=-9537.9x^2+96553x+61573$ $R^2=0.7626$
	120	1053^{a} ±94	359987^{b} ±81825	416341^{b} ±178251	129258^{a} ±28196	$y=-14260x^2+144796x+96662$ $R^2=0.7444$
Glutinous indian millet	60	185^{a} ±53	53373^{b} ±5393	47374^{b} ±7768	37694^{b} ±28140	$y=-1175.3x^2+13608x+17246$ $R^2=0.4567$
	120	253^{a} ±33	87574^{b} ±5923	74535^{b} ±9581	67429^{b} ±53677	$y=-1691.6x^2+20444x+28956$ $R^2=0.4247$
Job's tears	60	389^{a} ±27	60795^{b} ±10776	60744^{b} ±37228	51545^{b} ±21021	$y=-1175.3x^2+13608x+17246$ $R^2=0.4567$
	120	328^{a} ±111	95975^{b} ±13958	92232^{b} ±52782	82342^{b} ±32593	$y=-2102.7x^2+25886x+30411$ $R^2=0.5308$
Millet	60	502^{a} ±255	70033^{b} ±25470	75642^{b} ±10738	38111^{c} ±2310	$y=-2297.4x^2+24535x+20300$ $R^2=0.6515$
	120	666^{a} ±326	112149^{b} ±40451	119640^{b} ±14693	61956^{c} ±3248	$y=-3604.3x^2+38615x+32678$ $R^2=0.6391$
Polished barley	60	322^{a} ±43	1441^{a} ±408	5565^{b} ±2026	5017^{b} ±2593	$y=-115.43x^2+1642.9x+151.36$ $R^2=0.9954$
	120	385^{a} ±263	1910^{a} ±802	7971^{b} ±2752	7472^{b} ±3653	$y=-161.08x^2+2350.4x+107.99$ $R^2=0.9944$
Glutinous rice	60	603^{a} =246	34461^{b} ±5900	42441^{b} ±9711	47015^{b} ±7059	$y=28310x^{0.2767}$ $R^2=0.9957$
	120	714^{a} ±368	54962^{b} ±8158	66599^{b} ±15510	72312^{b} ±10729	$y=43253x^{0.2947}$ $R^2=0.9943$
Glutinous rice powder	60	326^{a} ±123	18491^{b} ±4201	44236^{bc} ±11832	32582^{b} ±9898	$y=-1116.1x^2+14148x+2475$ $R^2=0.9863$
	120	524^{a} ±197	29284^{b} ±7014	66666^{bc} ±15147	51287^{b} ±15429	$y=-1639.1x^2+21032x+4445\ 2$ $R^2=0.9802$
Polished rice	60	391^{a} ±109	43314^{b} ±10757	62626^{b} ±10247	100782^{c} ±23076	$y=41776x^{0.3381}$ $R^2=0.9984$
	120	506^{a} ±235	71179^{b} ±17101	101012^{b} ±15613	158835^{c} 35571	$y=65935x^{0.3521}$ $R^2=0.9986$
Brown rice	60	399^{a} ±101	89247^{b} ±22438	201315^{c} ±16982	193825^{c} ±25256	$y=90723x^{0.3924}$ $R^2=0.9982$
	120	437^{a} ±279	147961^{b} ±35917	321483^{c} ±26478	314239^{c} ±39343	$y=142299x^{0.4178}$ $R^2=0.9983$

[1]Control = unirradiated sample x: irradiation dose (kGy), y: photon counts

[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 ($p<0.05$).

ton counts of the unirradiated cereals were below 704±86 (60 s) and 1053±94 (120 s). Therefore, if photon counts of the unknown cereals measured by PPSL are below or above that, it is possible to distinguish whether they are unirradiated or not. Based on the above results, in all the samples, detection of irradiated cereals by PPSL seems possible.

The photon counts of irradiated legumes measured by PPSL are shown in Table 4. The photon counts increased with increasing irradiation doses and showed clear differences between unirradiated and irradiated samples. Legumes have been utilized in many kinds of traditional oriental foods, including soybean curd (*Tofu*), soybean sauce, soybean paste, soybean sprouts, and others, and irradiation of legumes has been used to reduce losses of legumes from insect infestation and micro-

Table 4. The changes of accumulated photon counts of unirradiated and irradiated legumes (Unit : Photon counts)

Sample	Measurement time (s)	Irradiation dose (kGy) Control[1)]	1	5	10	Regression expressions and coefficient
Black soybean	60	483[a] ±89	111910[b] ±3790	88686[c] ±14106	59895[d] ±8022	$y=-2421.5x^2+26074x+37226$ $R^2=0.3801$
	120	629[a] ±125	180.289[b] ±6844	144161[c] ±19179	99080[d] ±13991	$y=-3903.6x^2+42312x+59758$ $R^2=0.3844$
Kongnamul-kong	60	878[a] ±62	977258[b] ±49159	1005557[b] ±343116	336302[c] ±81561	$y=-34099x^2+343512x+280073$ $R^2=0.6609$
	120	1322[a] ±52	1531111[b] ±67210	1603201[b] ±552607	545012[c] ±126085	$y=-54165x^2+547829+435049$ $R^2=0.6710$
Mung bean	60	437[a] ±40	23862[b] ±2125	27041[b] ±1793	11806[c] ±5120	$y=-851.04x^2+8933.7+6859.7$ $R^2=0.7031$
	120	595[a] ±135	37283[b] ±3001	44242[b] ±5117	19705[c] ±9030	$y=-1385.4x^2+14677x+10389$ $R^2=0.7324$
Mung bean powder	60	282[a] ±108	5296[ab] ±1057	14314[b] ±7728	15207[b] ±8940	$y=7751.9x^{0.2431}$ $R^2=0.9983$
	120	266[a] ±16	8360[ab] ±2037	20731[b] ±10655	25477[b] ±14957	$y=11601x^{0.2762}$ $R^2=0.9888$
Red bean	60	436[a] ±45	186848[b] ±36108	183935[b] ±2771	83819[c] ±13904	$y=-5795.1x^2+60147x+55717$ $R^2=0.5937$
	120	585[a] ±40	294097[b] ±55060	288337[b] ±4353	137619[c] ±21748	$y=-8963.7x^2+93626x+88011$ $R^2=0.5847$
Yellow soybean	60	1219[a] ±664	323242[b] ±119011	334047[b] ±76182	245515[b] ±41169	$y=-8641.7x^2+100124x+97724$ $R^2=0.5874$
	120	1928[a] ±1365	510419[b] ±186354	547681[b] ±118267	399145[b] ±63759	$y=-14218x^2+165280+151550$ $R^2=0.6190$
Yellow soybean powder	60	437[a] ±174	30917[ab] ±12031	88203[bc] ±53588	140194[c] ±31030	$y=13466x+11076$ $R^2=0.9757$
	120	459[a] ±284	46192[ab] ±17190	127110[bc] ±71487	204102[c] ±44617	$y=19537x+16317$ $R^2=0.9758$
Small black soybean	60	1807[a] ±873	201173[b] ±49451	173632[b] ±58580	75412[c] ±26708	$y=-5539.6x^2+55822x+64214$ $R^2=0.5073$
	120	2762[a] ±1320	316168[b] ±71046	274960[b] ±92221	125090[c] ±44497	$y=-8659.1x^2+87934x+100770$ $R^2=0.5055$

[1)]Control = unirradiated sample x: irradiation dose (kGy), y: photon counts
[a-d]Means with the same superscripts in each row are not significantly different among group by Duncan's multiple range test in one way ANOVA ($p<0.05$)

bial contamination. Hence, the necessity of a detection technique for irradiated legumes will also increase (22).

The changes of photon counts with irradiation doses of wheat and wheat flour are given in Table 5. Photon counts of wheat and wheat flour exhibited an increase with increasing irradiation doses, and showed a very high relationship between irradiation doses and photon counts in a multinomial expression.

In all irradiated foodstuffs, photon counts emitted by infrared exhibited an increase from unirradiated control to 5 kGy, but photon counts of irradiated foodstuffs in the 10 kGy slightly decreased rather than those in the 5 kGy. However, in all irradiated foodstuffs, detection for irradiation is possible, because the photon counts of the irradiated foodstuffs were higher than the unirradiated ones.

In principle, as Bailiff and Barnett (9) explained, infrared stimulated luminescence in feldspar is a two-stage process, and occurs by the photoexcitation of charge from the ground state of an electron trap to an excited state, followed by elevation of the electron to the conduction band by phonon absorption. Hence, the theory of the process of photon emission from irradiated foodstuffs stimulated by infrared is considered to be similar to that of feldspar as explained by Bailiff and Barnett (9).

Several researchers have carried out studies for PPSL. Sanderson et al. (16) analyzed unirradiated and irradiated 45 herbs, spices and seasonings, and reported that more than 90% of irradiated samples can be readily identified on the basis of simple intensity measurements. Sanderson (17) also reported that the photon counts of intestinal grits and irradiated

Table 5. The changes of accumulated photon counts of unirradiated and irradiated wheat and wheat flour (Unit : Photon counts)

Sample	Measurement time (s)	Irradiation dose (kGy)				Regression expressions and coefficient
		Control[1)]	1	5	10	
Buckwheat	60	436^{a} ±232	15163^{b} ±4646	41445^{c} ±4105	46007^{c} ±3889	$y=-732.43x^2+11707x+2007.5$ $R^2=0.9944$
	120	540^{a} ±241	24138^{b} ±7465	65404^{c} ±6301	73533^{c} ±6625	$y=-1140.6x^2+18411x+3188.9$ $R^2=0.9937$
Buckwheat powder	60	348^{a} ±175	17323^{b} ±6096	33474^{c} ±1143	26954^{c} ±7413	$y=-799.88x^2+10312x+3472.2$ $R^2=0.9500$
	120	324^{a} ±107	27998^{b} ±9654	57874^{c} ±8476	43107^{c} ±11527	$y=-10353x+8032.7$ $R^2=0.9062$
Unclean wheat rice	60	351^{a} ±107	46479^{b} ±15924	113673^{c} ±22777	96317^{c} ±16995	$y=-2626.1x^2+35229x+6013.2$ $R^2=0.9870$
	120	369^{a} ±120	75766^{b} ±25216	181550^{c} ±34175	156148^{c} ±28050	$y=-4153.5x^2+56019x+10220$ $R^2=0.9847$
Clean wheat rice	60	257^{a} ±125	5170^{a} ±1010	20351^{b} ±11692	25607^{b} ±6064	$y=-296.48x^2+5514x+129.84$ $R^2=0.9999$
	120	335^{a} ±64	7925^{a} ±986	26283^{b} ±20113	40558^{b} ±9618	$y=-234.88x^2+6300.2x+973.77$ $R^2=0.9987$
Korean wheat	60	312^{a} ±148	21301^{b} ±9314	41649^{c} ±6859	22464^{b} ±978	$y=-1218.1x^2+140.15x+3741.5$ $R^2=0.9562$
	120	388^{a} ±206	35426^{b} ±15752	66017^{c} ±10818	37219^{b} ±1305	$y=-1902.4x^2+22013x+6636.8$ $R^2=0.9424$
Korean wheat powder	60	443^{a} ±188	4161^{ab} ±1952	8326^{bc} ±1015	11243^{c} ±4820	$y=992.38x+2074.1$ $R^2=0.9093$
	120	562^{a} ±142	6725^{ab} ±3150	14089^{bc} ±817	17531^{c} ±7271	$y=-1200.8x^2+8946.4x-31.057$ $R^2=0.9846$
White wheat	60	301^{a} ±64	33353^{b} ±5797	85795^{c} ±15602	75386^{c} ±9860	$y=-1926.1x^2+26369x+3905.1$ $R^2=0.9911$
	120	462^{a} ±210	54596^{b} ±8481	135509^{c} ±24507	120442^{c} ±16075	$y=-3016.9x^2+41436x+7040.9$ $R^2=0.9882$
Dark northern spring	60	402^{a} ±49	15546^{b} ±920	34142^{c} ±8902	32203^{c} ±10132	$y=-718.93x^2+10101x+2813.9$ $R^2=0.9753$
	120	509^{a} ±45	25317^{b} ±1398	58044^{c} ±9611	50507^{c} ±14448	$y=-1309.3x^2+11701x+4032.5$ $R^2=0.9807$
Durum	60	188^{a} ±38	10831^{ab} ±163	19713^{bc} ±708	31697^{c} ±17951	$y=-156.75x^2+4421.7x+2858$ $R^2=0.9576$
	120	253^{a} ±48	17553^{ab} ±392	32057^{bc} ±1461	49571^{c} ±27942	$y=-295.27x^2+7410.8x+4516.4$ $R^2=0.9561$
Strong flour	60	550^{a} ±255	6609^{ab} ±1823	12094^{bc} ±5629	22197^{c} ±9837	$y=1987.9x+2411.4$ $R^2=0.9668$
	120	822^{a} ±454	11665^{ab} ±3621	19428^{bc} ±8445	33697^{c} ±14164	$y=2956.9x+4575.6$ $R^2=0.9453$
Weak flour	60	259^{a} ±154	11412^{b} ±3503	30495^{c} ±2442	30248^{c} ±4474	$y=15049x^{0.2962}$ $R^2=0.9915$
	120	260^{a} ±134	18247^{b} ±5031	45979^{c} ±4379	44685^{c} ±6192	$y=22104x^{0.3228}$ $R^2=0.9954$

[1)]Control = unirradiated sample x: irradiation dose (kGy), y: photon counts
[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 ($p<0.05$).

bown shrimp samples were higher than unirradiated samples. Chung et al. (19) reported that PPSL values of irradiated white ginseng powder were negative for unirradiated samples but were positive for irradiated ones. Hwang et al. (18) also reported that the photon counts of irradiated pepper powder, dried herbs, fresh shrimp, potato, soybean, dried fig, chestnut, dried squid, and dried cod increased with dose. Our results above agree with those reported by other researchers (16-19).

Based on the results, the detection of irradiated foodstuffs is possible by PPSL and PPSL can be proposed as a method for the detection of the irradiated foodstuffs.

In order to detect irradiated foodstuffs by PPSL, future work is needed for the detailed demonstration of emitting phenomena in irradiated foodstuffs by PPSL, the settlement of threshold levels of foodstuffs not yet examined in this study, and the changes of photon counts of irradiated samples under a variety of storage conditions and time periods.

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