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Pulsed Photostimulated Luminescence of Irradiated Black and White Peppers and Effects of Long-Term Storage

Sang-Duk Yi, Jae-Seung Yang*, Chi-Kwang Song, Hyun Kyo Jeoung, Kyu-Seob Chang and Man-Jin Oh†**

Department of Food Science and Technology, Chungnam National University, Taejon 305-764, Korea
**Detection Lab. of Irradiated Food, Korea Atomic Energy Research Institute, Taejon 305-353, Korea*
***R&D center, Haechandle Foods Co. Ltd, Chungnam 320-833, Korea*

Abstract

Changes in accumulated pulsed photostimulated luminescene (PPSL) signals were observed after storage, which affected the ability to detect irradiation in black and white peppers. The PPSL curves were accumulated linearly during the 120 s measurement times, and PPSL signals increased according to irradiation doses. Threshold levels of black and white peppers were below 557±220 and 503±92 photon counts in 60 s, and below 679±351 and 812±648 photon counts in 120 s, respectively. The PPSL signals of black and white peppers linearly increased with irradiation dose up to 5 kGy, but very little from 5~10 kGy. The accumulated PPSL signals of irradiated black and white peppers had higher decay rates when stored in normal room conditions than in a darkroom. Detection of irradiation was possible for up to 12 months after irradiation, if the samples were stored in a darkroom.

Key words: black pepper, white pepper irradiation, pulsed photostimulated luminescence (PPSL)

INTRODUCTION

Black and white peppers are widely used food spices (1) that are frequently irradiated because of their natural propensity for contamination with molds, yeasts, bacteria, and insects (2,3). Black and white peppers may also be contaminated by mycotoxigenic species, such as *Aspergillus parasiticus* (*flavus*), that produce the food poisons, aflatoxin B1, B2, G1 and G2 (4-6); *A. ochraceus* that produces ochratoxin A (7) which is carcinogenic in humans; and *Penicillium citrinum*, and *P. islandicum* which produce citrinin and paltulin (8,9), respectively. Hence, irradiation can be used both for reducing health hazards associated with pathogenic and mycotoxigenic microorganisms, and for the reduction of microbial contamination and disinfecting (10).

Gamma irradiation is used to sanitize black and white peppers in 26 countries as of 1995 and this technique has become increasingly important with the increased awareness of the benefits of food irradiation, since irradiation does not adversely affect the quality and safety of the product (11). With increasing use of irradiation technology, detection methods for identifying irradiated food have been requested by consumer organizations and have developed by many researchers (10,12-23), because consumers should be provided with correct and comprehensive information about food irradiation and irradiated food, thus enabling them to make informed choices.

Pulsed photostimulated luminescence (PPSL), which uses light rather than heat to stimulate the release of trapped charge carriers following irradiation, has been applied to detection studies on irradiated foodstuffs. PPSL has been designed to allow direct measurements for rapid screening (15-17,22). Previous studies have used PPSL for detection of food irradiation in corn powder (11), perilla and sesame seeds (14,16), starches, beans and cereals (15), seasoning powder (17,19), spices (18), white ginseng powder (20) and foods like pepper powder, dried herbs, fresh shrimp, potato, soybean, dried fig, chestnut, dried squid, dried cod (21). As a result of the success of those studies, PPSL measurement is proposed as a screening method for detecting irradiation treatment. However, there are no reports on changes in PPSL signals of irradiated black and white peppers during the long-term storage under varied conditions. Therefore, the objective of this study was to establish PPSL methods for detecting irradiated black and white peppers by determining the effects of storage conditions and periods on the accumulated PPSL signals.

MATERIALS AND METHODS

Materials and packing

Black and white peppers produced in Malaysia were

†Corresponding author. E-mail: ohmanjin@cnu.ac.kr
Phone: 82-42-821-6728. Fax: 82-42-821-6728

purchased from a Korean supplier (Dongbang Trade Co. Ltd, Eumseong, Chungcheongbuk-do). Samples were packed in polyethylene bags, divided into two portions (room and darkroom conditions), sub-divided for irradiation dose, and irradiated using a Co-60 irradiator (AECL, IR-79, Ontario, Canada) at 1, 5, and 10 kGy, respectively, at a dose rate of 1 kGy/h at Korea Atomic Energy Research Institute. After irradiation, the samples for normal room conditions were stored in a laboratory condition, and the samples for darkroom conditions were stored in a chamber oven at room temperature (K.M.C-1203P3, Vision Scientific Co., LTD, Seoul, Korea) to block light exposure. Absorbed dose rates were documented using a ceric-cerous dosimeter.

Measurement of pulsed photostimulated luminescence

The PPSL system (serial; 0021, SURRC; Scottish Universities Research and Reactor Center, Glasgow, UK) 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 emitting diodes which are pulsed symmetrically on and off for equal periods. PPSL is detected by a bialkali cathode photomultiplier tube operating in the photon counting mode. Optical filtering is used to define both the stimulation and detection wavebands (Fig. 1). The samples (5 g) were placed in a 50 mm diameter disposable petri dish (Bibby Sterilin type 122, SURRC, Glasgow, UK) without other preparations, and measured in the sample chamber for 120 s. The radiation-induced photon counts (PPSL signals) emitting per sec from the irradiated samples were automatically accumulated in personal computer and presented the photon counts accumulated up to 60 s and 120 s (22). The samples were measured in triplicate under the same laboratory and instrumental conditions.

Statistical analysis

Mean values ± standard deviations were determined using Microsoft office 2000 (Excel, Microsoft Corporation, Redmond, WA, USA). All experiments were repeated three times.

RESULTS AND DISCUSSION

Properties of PPSL curves

PPSL curves, plotted from PPSL emissions per sec from irradiated and stored black and white peppers, are shown in Fig. 2. Immediately after irradiation, the PPSL signals were accumulated linearly up to the 120 sec. PPSL signals increased according to irradiation doses except for the PPSL signals from 5 to 10 kGy in white pepper. On the other hand, the PPSL curves measured after storage of 12 months were very low.

Threshold levels

Others have reported the effects of storage on PPSL emissions (12-18). Accordingly, this study evaluated the effects of storage in both light and dark conditions during the 12 months. The accumulated photon counts were obtained by directly measuring 5 g of black and white pepper samples, without other preparation, using PPSL. Threshold levels, defined as accumulated PPSL signals for non-irradiated samples, and border line levels, between irradiated and non-irradiated samples, of black and white peppers were below 557±220 and 503±92 photon counts in 60 sec, respectively, and below 679±351 and 812±648 photon counts in 120 sec, respectively, regardless of storage conditions and time (Table 1 and 2). Therefore, if the PPSL signals of unknown black or white pepper samples are higher or lower than the threshold levels, they

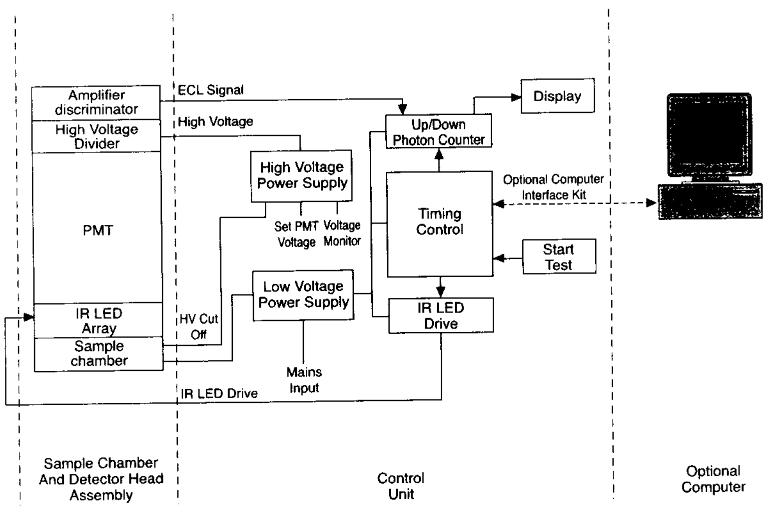

Fig. 1. Boundaries diagram of PPSL System.

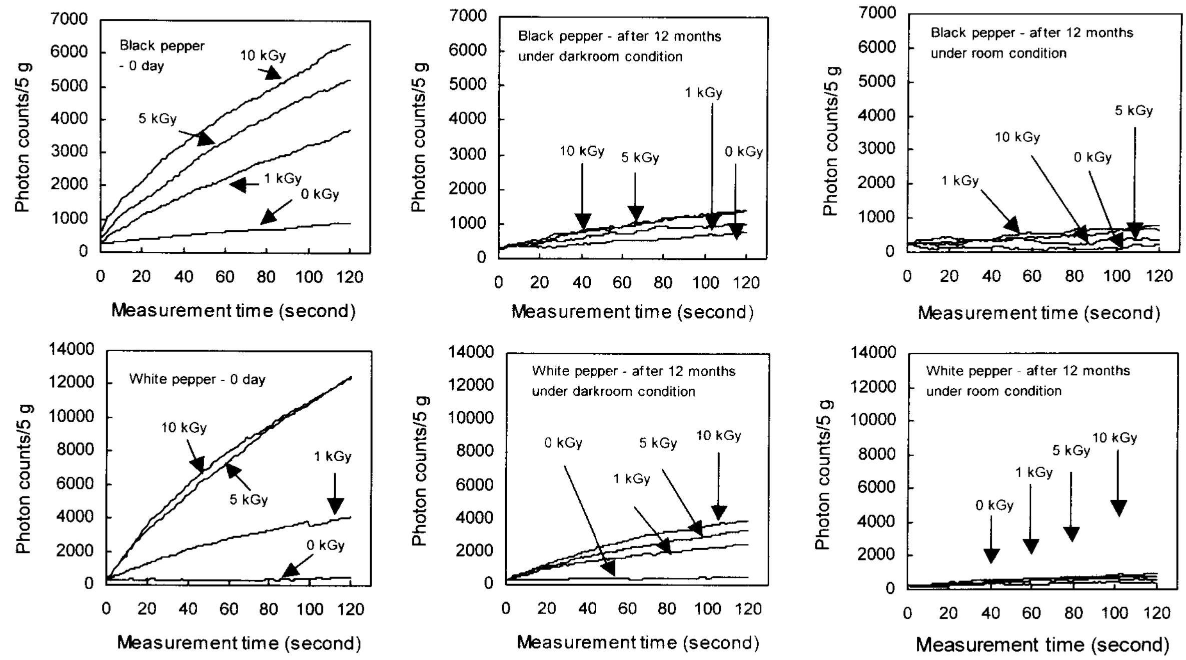

Fig. 2. Properties of PPSL curves of irradiated black and white peppers measured for 120 s depending on storage conditions.

Table 1. The changes of accumulated photon counts of unirradiated and irradiated black peppers according to storage conditions and periods (unit: photon counts)

Storage period & condition		Measurement time (s)	Irradiation dose (kGy) 0	1	5	10
0 day		60	347±68[1]	2,243±124	3,560±185	3,773±415
		120	493±46	3,538±238	5,737±457	6,213±912
1 months	Room	60	278±60	624±267	1,860±333	1,403±340
		120	372±78	930±391	2,486±430	1,937±521
	Darkroom	60	469±179	817±88	3,425±979	3,193±584
		120	679±351	1,261±387	5,065±1,704	4,770±1,533
3 months	Room	60	290±50	678±525	1,135±110	1,391±424
		120	442±50	809±534	1,615±623	2,074±1,026
	Darkroom	60	228±77	1,063±315	1,348±429	1,329±158
		120	306±131	1,646±455	2,142±869	2,399±778
6 months	Room	60	276±7	690±202	521±262	541±180
		120	250±82	925±136	712±317	518±126
	Darkroom	60	297±146	862±136	1,355±349	1,220±360
		120	332±49	1,034±163	2,237±382	2,502±846
12 months	Room	60	275±194	653±91	354±275	418±108
		120	270±138	910±321	506±330	730±101
	Darkroom	60	557±220	779±101	1,301±485	1,316±373
		120	516±220	1,057±69	1,922±762	1,807±743

[1]Mean value±standard deviation for 3 measurements.

can be considered to be irradiated.

Influence of irradiation dose

Accumulated PPSL signal is the sum of light emitting per second from irradiated samples by infrared stimulation at a second intervals. Therefore, even if samples are prepared for measurement under identical conditions, the accumulated PPSL signals will differ, depending on the measurement time. This problem can be controlled for by consistently using the same measurement times and uniform experimental conditions. Yi et al. (12-18) proposed that measurement times of 60 or 120 sec be used. If the accumulated PPSL signal of irradiated samples is too low to be counted, a longer measurement time is needed (120 s) to obtain a high enough PPSL signal for detection

Table 2. The changes of accumulated photon counts of unirradiated and irradiated white peppers according to storage conditions and periods (unit: photon counts)

Storage period & condition		Measurement time (s)	Irradiation dose (kGy) 0	1	5	10
0 day		60	362± 70[1)]	2,962±700	8,390±1,437	8,062±1,434
		120	510± 27	4,400±923	13,318±1,985	12,785±1,472
1 months	Room	60	503± 92	1,566±121	2,822±1,180	2,380± 560
		120	812±648	2,462± 57	4,256±1,790	3,671± 782
	Darkroom	60	405±149	1,763±585	5,385± 323	5,881±1,016
		120	662±175	2,866±969	8,693±1,762	9,376±1,749
3 months	Room	60	292± 56	1,339±296	2,468± 678	1,994± 918
		120	423± 56	2,089±485	3,544±1,067	2,897±1,802
	Darkroom	60	157±183	1,868±331	3,718±1,171	2,268± 838
		120	175±156	2,747±642	4,705±2,164	3,275±1,542
6 months	Room	60	324± 49	1,280±278	1,296± 256	1,839± 620
		120	365±107	1,562±336	1,656± 180	3,018±1,518
	Darkroom	60	135± 43	1,863±534	3,609± 821	2,960± 966
		120	240± 31	2,917±639	4,136±2,375	3,783± 231
12 months	Room	60	304±206	569±221	526± 245	912± 700
		120	394±201	863±345	677± 271	1,317±1,132
	Darkroom	60	291± 49	2,005±349	2,130± 347	2,045± 502
		120	403± 96	3,020±587	3,260± 668	3,254± 835

1)Mean value±standard deviation for 3 measurements.

of irradiation. On the other hand, if the accumulated PPSL signal of tested samples is high enough, the shorter measurement time (60 s) is adequate and more efficient. Hence, this paper reported photon counts accumulated for both 60 s and 120 s.

Changes in black pepper PPSL signals accumulated for 60 and 120 s, according to storage conditions, either not irradiated or irradiated at 1, 5, or 10 kGy are shown in Table 1. The number of radiation-induced PPSL signals increased linearly with irradiation dose up to 5 kGy but did not increase substantially from 5~10 kGy in samples stored in the darkroom for 12 months. However, the PPSL signals of irradiated samples were not observed in samples stored in normal room condition for 6 months. Discrimination of irradiated sample by PPSL signals is based on higher PPSL signals in irradiated than non- irradiated samples (23). Therefore, although the PPSL signals were not linearly changed according to irradiation dose and decreased with the lapse of time, detection of irradiated samples was possible for up to 12 months after irradiation, if they were stored in darkroom conditions, because the signals of irradiated samples were higher than those of non-irradiated samples.

These results are consistent with published data on other irradiated foods such as corn powder (12), perilla and sesame seeds (14,16), cereals, beans and wheat (15), seasoning powders (17), spices (18) and white ginseng powder (20), which have higher accumulated PPSL signals when measured immediately after irradiation than those of non-irradiated samples, with a linear increase up to 5 kGy, and little or no increase from 5~10 kGy.

Table 2 shows the changes in PPSL signals accumulated for 60 and 120 sec for non-irradiated and irradiated white peppers stored under different conditions. Properties of accumulated PPSL signals of irradiated white peppers were similar to those observed in black pepper, except that the accumulated PPSL signals were approximately 2-fold higher in white pepper than in black pepper. This trend was also observed after storage for 12 months in darkroom conditions. Yi et al. (16) reported that the accumulated PPSL signals of extracted minerals from sesame and perilla seeds were much greater than those from the seeds themselves, indicating that the radiation-induced PPSL signals are primarily trapped in minerals rather than the organic portion of contaminated seeds. Hence, one reason for higher accumulated PPSL signals in white rather than black pepper might be due to the different mineral contents between samples.

Decay of PPSL signal

Decay rates of PPSL signals from irradiated black pepper, stored under different conditions, are shown in Fig. 3 and 4. The accumulated PPSL signals of samples stored for 12 months following 10 kGy irradiation decreased by approximately 88 and 71% below initial levels under normal versus darkroom conditions, respectively. This trend according to storage conditions was also observed in

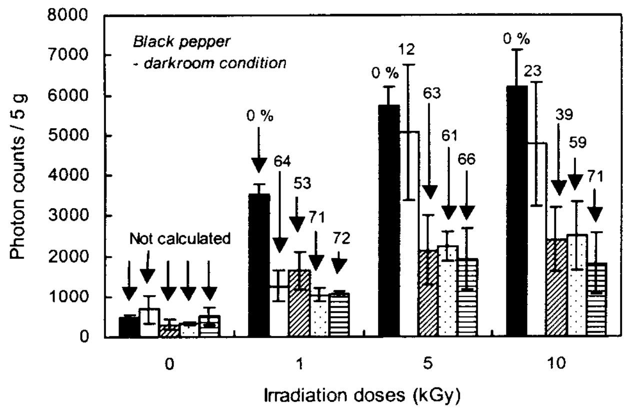

Fig. 3. The changes of decay rate calculated from accumulated PPSL signals of irradiated black pepper measured for 120 s during storage of 12 months under darkroom condition.
■: Accumulated PPSL signals of samples measured immediately after irradiation for 120 s.
□: Accumulated PPSL signals of samples measured at 1 month after irradiation for 120 s.
▨: Accumulated PPSL signals of samples measured at 3 months after irradiation for 120 s.
□: Accumulated PPSL signals of samples measured at 6 months after irradiation for 120 s.
▤: Accumulated PPSL signals of samples measured at 12 months after irradiation for 120 s.
Mean value±standard deviation for 3 measurements.

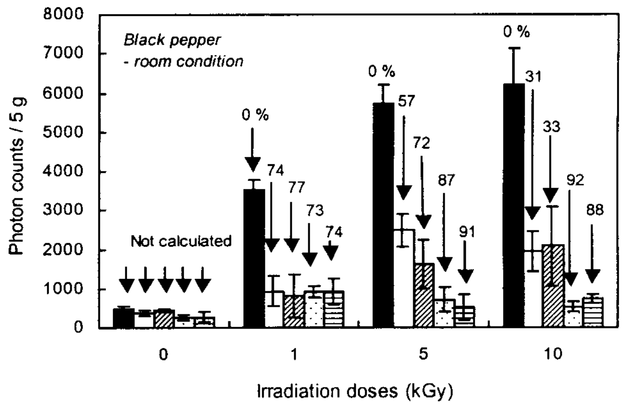

Fig. 4. The changes of decay rate calculated from accumulated PPSL signals of irradiated black pepper measured for 120 s during storage of 12 months under room condition.
■, □, ▨, □, ▤: Refer to the legend of Fig. 3.
Mean value±standard deviation for 3 measurements.

black pepper irradiated at 1 and 5 kGy.

Others have suggested that decreased accumulated PPSL signals after storage in normal room conditions are a result of stimulation by either sunlight or artifical light, rather than infrared light, that causes a release of radiation induced PPSL photon counts (12,18,23).

Decay rates of the accumulated PPSL signals measured for irradiated white pepper in normal room and darkroom conditions are shown in Fig. 5 and 6. The trends were similar to black pepper except for the decay rate (about 31%)

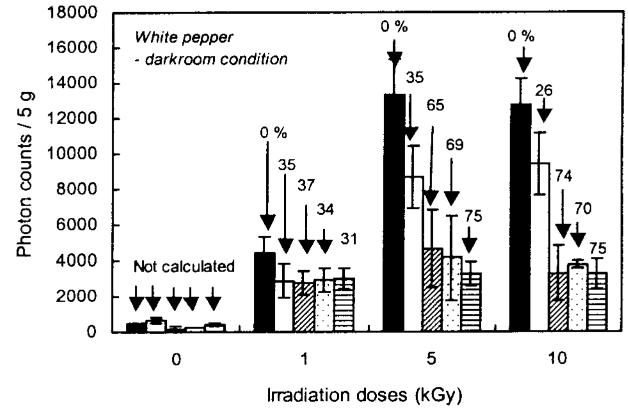

Fig. 5. The changes of decay rate calculated from accumulated PPSL signals of irradiated white pepper measured for 120 s during storage of 12 months under darkroom condition.
■, □, ▨, □, ▤: Refer to the legend of Fig. 3.
Mean value±standard deviation for 3 measurements.

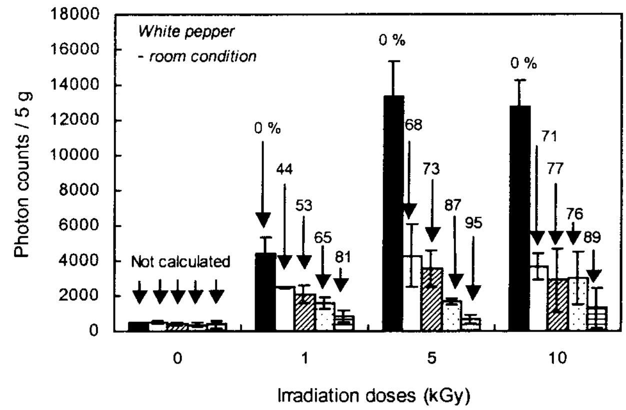

Fig. 6. The changes of decay rate calculated from accumulated PPSL signals of irradiated white pepper measured for 120 s during storage of 12 months under room condition.
■, □, ▨, □, ▤: Refer to the legend of Fig. 3.
Mean value±standard deviation for 3 measurements.

of the 1 kGy samples stored under darkroom conditions.

These results suggest that, although PPSL signals of irradiated black and white peppers decrease with increasing storage time, the higher photon counts in irradiated than in non-irradiated samples permit detection for 3 to 6 months in samples stored under normal room conditions and up to 12 months when stored in a darkroom.

Therefore, we propose that PPSL is a suitable technique for detecting irradiation in black and white peppers.

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