

[報文]

## Detection of Irradiated Prawns by Photostimulated Luminescence

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### PSL 法によるエビの照射履歴の検知

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#### Summary

The purpose of this study was to investigate how photostimulated luminescence (PSL) can be applied to verify whether prawns have been irradiated by analyzing their intestinal tracts. Prawns from five different locations which were irradiated at doses of 1 kGy of  $\gamma$ -radiations were analyzed using the Japanese model PSL system. The results showed that the integrated photon counts of all the irradiated samples exceeded the upper threshold value ( $T_2 = 4000$  counts/90 s), whereas those of the non-irradiated samples were below the lower threshold value ( $T_1 = 1000$  counts/90 s). Moreover, using the other parameters which were decrease of intensity after optically stimulation and increase of intensity by optically stimulation, a clear difference was observed between non-irradiated and 1 kGy irradiated samples. Therefore, the Japanese model PSL system can be used as a screening method for detecting irradiated prawns by analyzing their intestinal tracts.

**Key words:** food irradiation, prawn, photostimulated luminescence (PSL), screening method

#### Introduction

Over the last few years, there has been an increasing demand for new techniques of food preservation to replace the use of hazardous chemicals. Among these techniques, food irradiation has been demonstrated to be particularly effective in inactivating pathogens, decreasing microbial load, and extending shelf life without an appreciable alteration in food quality<sup>1)</sup>. Food irradiation is a cold process involving a negligible rise in temperature, and thus, it is suitable

for maintaining the freshness of heat-sensitive foods such as seafood. For this reason, seafood, including prawns, is irradiated in many countries, such as Belgium, Vietnam, France, and Indonesia<sup>2)</sup>.

However, despite repeated assurances that irradiation is one of the safest methods to preserve foodstuffs, consumers should be allowed to choose between irradiated and non-irradiated foods. Thus, any irradiated food or any irradiated food ingredient of a compound food must be labeled with “irradiated” or “treated with ionizing radiation.” To enforce the

correct labeling or to detect non-authorized products, several analytical methods have been standardized by the European Committee for Standardization (CEN)<sup>3)</sup>. Thus, there is a great interest in the methods suitable for identifying irradiated foods as well as in the applicability of these methods to foods with different composition.

Recently, the thermoluminescence (TL) and 2-alkylcyclobutanone (2-ACB) methods were successfully applied to identify irradiated prawns<sup>4)</sup>. However, the TL and 2-ACB methods are time consuming, complex, expensive, and thus they are not suitable for rapid screening of irradiated prawns. Therefore, a simple, dependable, and a routine analytical method is a priority.

When foods are exposed to ionizing radiations, mineral debris, which is present on many foodstuffs, stores energy in charge carriers trapped in the structural or interstitial sites. Optical stimulation of these minerals by LED will release this stored energy as weak light that can be measured using a photon detector. This phenomenon, called photostimulated luminescence (PSL), has been successfully applied to identify irradiated foodstuffs. Analytical protocols using a European model PSL screening system have been tested in interlaboratory trials for shellfish, herbs, spices, and seasonings<sup>5), 6)</sup>, and a screening method for detecting irradiated foods was formulated by the European Standard (EN 13751)<sup>7)</sup>. Recently, a Japanese model PSL system suitable for Japanese humid conditions was developed. This PSL system is applicable to a large variety of foods such as oregano, peppers, and garlic<sup>8), 9)</sup>. However, there are no reports to apply this system for detecting the irradiated prawns.

In the present study, the suitability of the Japanese model PSL system to distinguish the radiation history of prawns was examined.

## Materials and Methods

### 1. Prawn samples

Five lots of beheaded prawns from different areas (A: India, white tiger; B: Indonesia, white tiger; C: Thailand, white vannamei; D: Vietnam, black tiger;

E: Indonesia, black tiger) were purchased from a local supermarket in Tsukuba, Ibaraki, Japan and preserved at  $-20^{\circ}\text{C}$ . Before analysis, the samples were protected against light exposure, and all dispensing and handling of samples were performed under subdued lighting.

### 2. Irradiation

The frozen prawn samples in aluminum-sealed polyethylene bags were irradiated with  $\gamma$ -rays from a cobalt-60 source (Gammacell 220; MDS Nordion International Co. Ltd., Ottawa, Ontario, Canada) at the National Food Research Institute (NARO) of Japan. The dose rate was 6 kGy/h. The samples were irradiated at doses of 1 kGy and protected from light in aluminum-sealed polyethylene bags storage at  $-20^{\circ}\text{C}$  until analysis. An alanine pellet dosimeter (Bruker Biospin Ltd, Rheinstetten, Germany) was attached to the surface of each sample, and the absorbed dose was determined with an electron spin paramagnetic spectrophotometer (Bruker EMX, Bruker Biospin Ltd, Rheinstetten, Germany). The non-irradiated samples were used as the control and stored under the same condition before use. All the samples were used for the PSL analysis after three days of storage at  $-20^{\circ}\text{C}$ .

### 3. Preparation of sample

The intestinal tract was the measurement target in this study. After the intestinal tract was removed from the prawn with forceps, the intestinal material was squeezed into a filter paper moistened with 0.5 mL of distilled water and laid out as thin as possible. Then, the filter paper was set into the stainless Petri dish ( $d = 50 \text{ mm}\phi$ ;  $h = 15 \text{ mm}$ ), which is the dedicated holder for PSL measurement.

### 4. PSL measurement

PSL measurements of prawn samples were carried out by Japanese model PSL system (ES-7340A, Japan Radiation Engineering Co. Ltd., Ibaraki, Japan). The PSL signals of the control and irradiated samples were measured for 90 s after 10 s spontaneous luminescence.

scense (background) measurement without the optical stimulation. Three parameters were calculated to evaluate the measurement results.

One parameter was the “photon count decrease,” which is the difference of the average photon counts between the initial 10 s and the last 10 s after optical stimulation. Another parameter was the “photon count increase,” which is the difference of the average photon counts before and after optical stimulation. Afterward, the results were expressed as “integrated net counts” for the recorded 90 s. Before and after each measurement, an empty Petri dish test was operated to ensure that it was free from contamination. All the samples were measured at  $25 \pm 1^\circ\text{C}$  and fewer than 50% of RH. All the handling and measurement of the samples were carried out under subdued lighting following the EN 13751 recommendations, and were repeated at least 6 times.

### Results and Discussion

Fig. 1 shows the picture of the measurement target in this study. The intestinal tracts were removed from the prawn samples (a), and then the intestinal materials were squeezed into a filter paper (b).

Fig. 2 shows the time course of PSL signals for the control and irradiated samples. The PSL signal of irradiated samples (b) significantly increased immediately after optical stimulation and then diminished with time. In contrast, the PSL signal of the non-irradiated samples (a) was unchanged before and after optical stimulation.

Table 1 summarizes the results of the evaluation parameters of photon count increase and decrease. The PSL of irradiated samples (1 kGy) increased

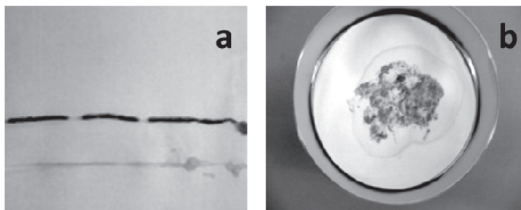


Fig. 1 Pictures of the samples. a) intestinal tracts; b) intestinal materials

immediately after optical stimulation and then diminished with time. On the contrary, PSL was not observed in non-irradiated samples (0 kGy), and the photon counts remained at the background level after optical stimulation. Thus, the values of these two parameters for non-irradiated samples were both approximately 0, which were significantly different from those of the irradiated samples. Therefore, it is possible to distinguish between irradiated and non-irradiated samples by using these parameters.

The results of the integrated net photon counts of the five lots are presented in Fig. 3. The photon counts of all of the non-irradiated samples (hollow symbols) were less than  $T_1 = 1000$  counts/90 s, which was defined as the lower threshold, whereas the counts of most of the irradiated samples (solid symbols) exceeded  $T_2 = 4000$  counts/90 s, which was defined as the upper threshold. The PSL photon count of the Vietnam black tiger (D lot) was closest to the upper

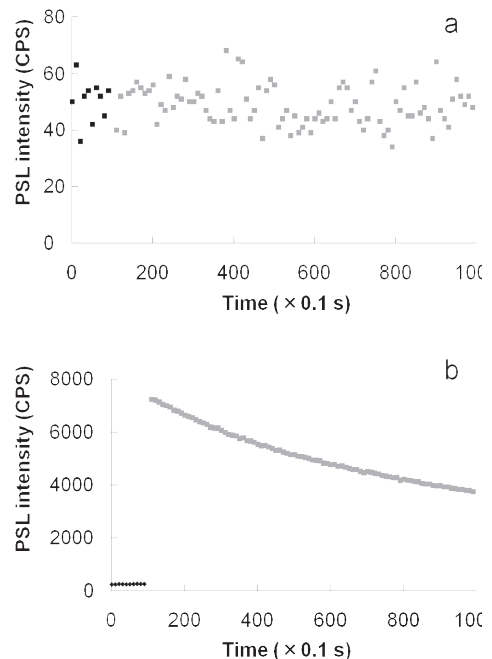


Fig. 2 PSL signals. a) control sample (0 kGy); b) sample irradiated at doses of 1 kGy

Table 1 Disparity of photon counts decrease parameter and increase parameter in five non-irradiated and irradiated (1 kGy) lots.

Sample	Decrease (counts)*		Increase (counts)**	
	0 kGy	1 kGy	0 kGy	1 kGy
A	-0.24±0.34	1654.17±1007.74	0.07±0.43	3349.85±2217.62
B	0.06±0.34	327.03±243.30	-0.04±0.27	534.85±408.33
C	-0.22±0.26	663.17±186.81	0.05±0.47	1272.62±355.23
D	0.07±0.26	86.13±83.15	0.13±0.51	138.17±131.12
E	-0.11±0.31	524.78±369.67	-0.04±0.47	831.60±604.55

<sup>1</sup> Mean values represent the mean ± SD of 6 determinations

\* Difference of the average photon counts between the initial 10 s and the last 10 s after optical stimulation.

\*\* Difference of the average photon counts before and after optical stimulation.

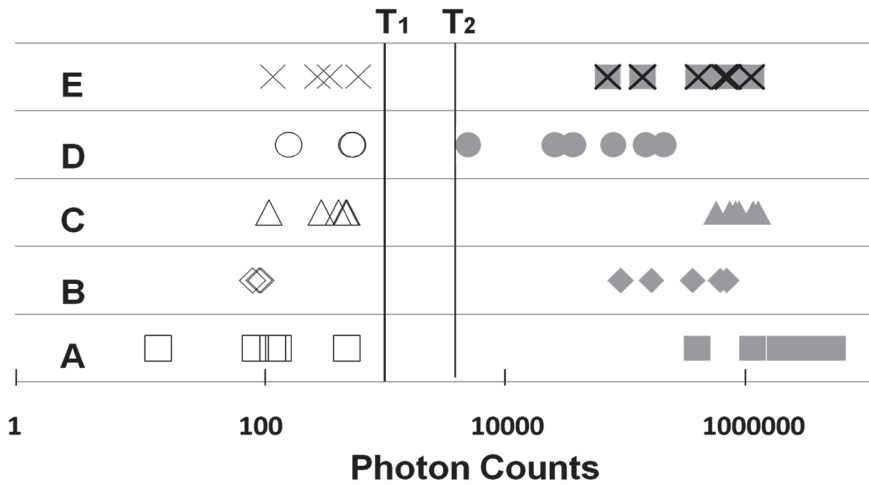


Fig. 3 Integrated PSL photon counts (counts/90 s) of irradiated and non-irradiated intestinal tracts of each lot (n=6) after 3 days of storage at -20°C.

$T_1 = 1000$  counts/90 s;  $T_2 = 4000$  counts/90 s.

Hollow symbols, non-irradiated samples; Solid symbols, samples irradiated at 1 kGy

A: India, white tiger, B: Indonesia, white tiger, C: Thailand, white vannamei, D: Vietnam, black tiger, E: Indonesia, black tiger.

threshold, and the values of all of the irradiated samples were higher than the upper threshold value, indicating a clear positive result. In contrast, the values of the non-irradiated samples were below the lower threshold value, indicating a negative result. Therefore, it is possible to use the threshold values ( $T_1$  and  $T_2$ ) to evaluate the irradiated prawns, which is similar to EN 13751 method <sup>7</sup>, although further analysis of

original samples is required.

In conclusion, PSL was not observed in any non-irradiated prawns tested in this study, and the PSL system can be used as a screening method for the detection of irradiated prawns.

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#### 和文要旨

本研究では、産地や種類の異なる5種類のエビ背腸の内容物について、国産PSLシステムで光ルミネッセンス測定を行った。その結果、非照射試料のPSL積算発光量は下限しきい値 ( $T_1 = 1000$  counts/90 s) を下まわり、一方、1 kGy照射した試料は全て上限しきい値 ( $T_2 = 4000$  counts/90 s) を超えることが確認された。また、LEDによる照射前後の発光量の増加とPSL発光の減少を評価パラメータとして用いても、非照射と1 kGy照射の試料との間に明確な差が確認された。したがって、本PSL法はスクリーニング方法としてエビの照射履歴を検知することができる。

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