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Calibrated photostimulated luminescence is an effective approach to identify irradiated orange during storage



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HIGHLIGHTS

• Photostimulatedluminescence (PSL) was studied to identify irradiated orange for quarantine application.

• PSL detection efficiency was compared amonggamma, electron, and X irradiation during shelf-life of oranges

- PSL properties of samples were characterized by standard samples
- Calibrated PSL gave a clear verdict on irradiation extending potential of PSL technique

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ABSTRACT

Photostimulated luminescence (PSL) has been employed as a fast screening method for various irradiated foods. In this study the potential use of PSL was evaluated to identify oranges irradiated with gamma ray, electron beam and X-ray (0–2 kGy) and stored under different conditions for 6 weeks. The effects of light conditions (natural light, artificial light, and dark) and storage temperatures (4 and 20 °C) on PSL photon counts (PCs) during post-irradiation periods were studied. Non-irradiated samples always showed negative values of PCs, while irradiated oranges exhibited intermediate results after first PSL measurements. However, the irradiated samples had much higher PCs. The PCs of all the samples declined as the storage time increased. Calibrated second PSL measurements showed PSL ratio < 10 for the irradiated samples after 3 weeks of irradiation confirming their irradiation status in all the storage conditions. Calibrated PSL and sample storage in dark at 4 °C were found out to be most suitable approaches to identify irradiated oranges during storage.

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1. Introduction

Irradiation of foods using ionizing radiation is of great importance with respect to Good Manufacturing Practice (GMP) and Good Hygienic Practice (GHP). International trade of fresh fruits has seen immense growth in recent time worldwide and orange has been found as one of the major items in this trade (USDA Foreign Agricultural Service, 2011). Growth is likely to continue over the next decade. However, the import of oranges from other countries is associated with the risk of migration of potentially damaging organisms such as insect pests to new areas. Food irradiation technology could be one of the potential solutions to this problem. However, the international legislation requires labeling

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http://dx.doi.org/10.1016/j.radphyschem.2015.02.024 0969-806X/© 2015 Published by Elsevier Ltd. of irradiated food to facilitate international trade and to meet consumer's rights of choice. Therefore, development of potential detection methods to classify marketed foods as irradiated or nonirradiated is of paramount interest (Arvanitoyannis, 2010).

Significant progress has been made in the development of detection methods for irradiated foods (Sanyal et al., 2009; Kwon et al., 2013; Sanyal et al., 2014; Jo et al., 2015). For routine detection of irradiated foods such as herbs, spices, fruits, vegetables, and shellfish (Beneitez et al., 1994; EN 1788, 2001; Sanyal et al., 2012), the standard validated methods have been based on thermoluminescence (TL). TL methods require a physical separation to extract minerals for calibrated analysis. Relatively lengthy and cumbersome laboratory preparation has limited the use of TL analysis. Therefore, there is a scope for development of faster methods requiring less sample preparation for routine commercial or enforcement testing. In this regard, photostimulated luminescence (PSL) technique is a nondestructive approach with simpler processing. PSL as a screening method (Sanderson et al., 2003) takes advantage of the fact that mineral debris which is found on many foods, store energy in charge carriers at structural, interstitial or impurity sites, when exposed to ionizing radiation. Optical stimulation of minerals release charge carriers, and part of the stored energy are emitted as light which can be measured by a photon counter. The PSL method is standardized by the European Committee for Standardization for the detection of irradiated foods, including shellfish, herbs, spices and seasonings (EN 13751, 2009). The technique comprises an initial measurement of PSL intensity, which may be used for screening purposes, and a calibration method to determine the PSL sensitivity to assist classification. The accumulated PSL signals (photon counts, PCs) are analyzed based on two thresholds, the lower threshold (T1 = 700counts/60 s) and upper threshold (T2 = 5000 counts/60 s). Samples with signals less than T1 are classified as nonirradiated and those with signals greater than T2 are classified as irradiated samples. However, PSL may be adversely affected by storage conditions of food samples (Bortolin et al., 2007). Samples can be measured more than once, but are subject to bleaching and therefore a controlled storage condition of food samples is required to identify irradiated samples after prolonged time. In addition, on repeated measurements, the PSL signals decrease and may give in special cases false positive and negative results (Bayram and Delincee, 2004). On the other hand, reliable identification of irradiated food after storage is one of the relevant requirements in the domain of commercial radiation processing of food.

In this study, the potential of the PSL technique as a screening method for oranges, irradiated with gamma, electron beam and X-ray for quarantine application, was verified at different temperature and under different light illuminations during 6 weeks of storage. Calibrated PSL measurements were also carried out to study the bleaching effect during storage at different conditions on photon counts (PCs) of minerals, irradiated with different doses, to understand the effectiveness of PSL as a potential detection method for irradiated oranges.

2. Materials and methods

2.1. Materials and irradiation treatment

Navel oranges from the United States were purchased from a local grocery in Daegu, Korea. Samples were exposed to 3 different types of allowed ionizing radiations in the dose range of 0-2 kGy. Feldspars and quartz were procured from Sigma as standard material to study the PSL responses after exposure to different ionizing radiations. Gamma irradiations were carried out using Co-60 gamma ray source (activity 100 kCi, dose rate 1.5 kGy/h, AECL, IR-79, MDS Nordion International Co. Ltd., Ottawa, Canada) in Korean Atomic Energy Research Institute, Korea. Electron beam and X-ray irradiations were carried out with electron accelerator (ELV-4, 10 MeV, Fujifilm, Tokyo, Japan) in EB-Tech, Korea. In case of electron beam the dose rates for 0.5 and 2 kGy accumulated doses were of the order of 1.11 and 3.14 kGy/s, respectively. For X-ray irradiation the dose rate was of the order of 0.42 kGy/s. Absorbed doses were measured using alanine dosimeter (EMS 104 EPR analyzer, Bruker Biospin, Rheinstetten, Germany). Samples were stored at 4 and 20 °C under the condition of a dark room (DR), artificial light (AL) and an indirect natural light (INL). Artificial light was in the range of about 200 ± 5 lx as measured by a Digital Lux Meter (1330A, TES, Taiwan).

2.2. PSL analysis

PSL measurements were performed as described by EN 13751 (2009) using a SURRC PPSL Irradiated Food Screening System (serial number 0021, Scottish Universities Research and Reactor Center, Glasgow, UK). The outer skin of the sample was carefully separated and cut into small size $(0.5 \times 0.5 \text{ cm}^2)$ using a sterile blade. The certain amount of samples $(5.0 \pm 0.2 \text{ g})$ and standard

Table 1

Lifets of post-inaulation storage temperatures on i se intensity of oranges before and after inaulation to unreferr foriging radiation	Effects of r	post-irradiation	storage tem	peratures on	PSL intensity	of oranges	before and	after irradiation	to different ionizing	g radiations
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Irradiation source	Irradiation dose (kGy)	Storage condition ^a	Storage period (week)					
			0	3		6		
			PCs ^b	PCs	I.R. ^c (%)	PCs	I.R. (%)	
None	0	D (4 °C) D (20 °C) FL (20 °C) INL	502 ± 97	$\begin{array}{c} 304 \pm 99 \\ 237 \pm 31 \\ 217 \pm 48 \\ 247 \pm 45 \end{array}$	- 39.44 - 52.78 - 56.77 - 50.70	$\begin{array}{c} 230 \pm 49 \\ 249 \pm 22 \\ 259 \pm 85 \\ 270 \pm 31 \end{array}$	- 54.18 - 50.39 - 48.40 - 46.21	
ү-гау	2	D (4 °C) D (20 °C) FL (20 °C) INL	3169 ± 262	$\begin{array}{c} 2562 \pm 187 \\ 2114 \pm 747 \\ 262 \pm 84 \\ 311 \pm 17 \end{array}$	- 19.15 - 33.29 - 91.73 - 90.18	$\begin{array}{c} 690 \pm 533 \\ 678 \pm 194 \\ 359 \pm 61 \\ 326 \pm 49 \end{array}$	- 78.22 - 78.60 - 88.67 - 89.71	
E-beam	2	D (4 °C) D (20 °C) FL (20 °C) INL	2771 ± 956	$\begin{array}{c} 1327 \pm 528 \\ 1683 \pm 286 \\ 1605 \pm 22 \\ 644 \pm 51 \end{array}$	- 52.11 - 39.26 - 42.07 - 76.75	$\begin{array}{c} 1,308 \pm 223 \\ 1,065 \pm 278 \\ 254 \pm 53 \\ 300 \pm 102 \end{array}$	- 52.79 - 61.56 - 90.83 - 89.17	
Х-гау	2	D (4 °C) D (20 °C) FL (20 °C) INL	2149 ± 641	$\begin{array}{c} 1659 \pm 427 \\ 1235 \pm 56 \\ 264 \pm 98 \\ 431 \pm 99 \end{array}$	- 22.80 - 42.53 - 87.71 - 79.94	$\begin{array}{c} 1,067 \pm 442 \\ 985 \pm 299 \\ 283 \pm 136 \\ 297 \pm 56 \end{array}$	- 50.34 - 54.16 - 86.83 - 86.17	

^a D: Dark, FL: Fluorescent light, INL: Indirect natural light (ambient).

^b N=negative: less than 700 PCs, P=positive: more than 5000 PCs, I=intermediate: between 700 and 5,000 PCs.

^c Increasing rate compared with the intensity immediately after irradiation using each irradiation source.

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