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Modification of LSC spectra of ^{125}I by high atomic number elements

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HIGHLIGHTS

- The influence of cocktail components on ^{125}I pulse-height spectra was studied.
- The spectra with Hionic-Fluor differed significantly from other cocktails.
- Modification by varying high atomic numbered elements in the cocktail was evaluated.
- Presence of photon interactions vary LSC spectra, depending upon composition.

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ABSTRACT

The ^{125}I pulse-height spectra via a liquid scintillation counter (LSC) displayed notable variations. The counting efficiencies of higher and lower energy peaks increased and decreased, respectively, with the enhancement of the amount of high atomic numbered elements within the cocktails. This tendency was ascribed to the increasing probability of the interaction of photons with the scintillation cocktail. Moreover, it was noted that the shape of a ^{125}I spectrum strongly depends on the amount of high atomic numbered elements.

1. Introduction

Radioactive compounds labeled with ^{125}I are widely used in biomedical research, because peptides and proteins can easily be labeled with ^{125}I by relatively simple procedures (Fraker and Speck, 1978; Bolton and Hunter, 1973; Greenwood et al., 1963). In addition, various types of biologically active ^{125}I-labeled compounds are commercially available. Tritium-labeled compounds are sometimes used in combination with ^{125}I-labeled compounds; for example, [^{3}H]dextran has been used as an impermeable reference in the measurement of brain-to-blood efflux transport of an ^{125}I-labeled peptide (Akanuma et al., 2008). Although the counting of ^{125}I will be performed using an NaI (Tl) scintillation counter, the counting of ^{3}H will require the use of a liquid scintillation counter (LSC).

The pulse height spectrum of ^{125}I measured by LSC contains two peaks, with the lower energy peak of ^{125}I almost overlapping with that of ^{3}H as shown in Fig. 1. In simultaneous counting of ^{3}H and ^{125}I using the spillover method (Ishikawa, 1975; Hetenyi and Reynolds, 1967) by LSC, the activity of ^{3}H is derived from subtracting the counts of ^{125}I counted in ^{3}H window from the total counts in ^{3}H window. The counts of ^{125}I counted in ^{3}H window is calculated from the counts of ^{125}I counted in ^{125}I window using a correction curve of the ratio of ^{125}I counts in ^{3}H window to those in ^{125}I window expressed as a function of quenching. It is necessary to prepare a correction curve for the peak ratio accurately for the precise calculation of ^{3}H activity.

It is generally known that correction curves with different scintillation cocktails are not always identical in LSC. The cause is not clear. Thus quench-correction curves were generated based on the LSC analysis via the simultaneous counting of ^{3}H and ^{125}I by the spillover method using commercially available Clear-sol II, Ecoscint XR, Insta-Gel Plus, Ultima Gold, and Hionic-Fluor (Shoji and Kondo, 2013). In using Hionic-Fluor and Ultima Gold, the counting efficiency of ^{125}I and the proportion of ^{125}I counts counted in ^{125}I window were greater in comparison to other cocktails; however, the counting efficiencies for ^{3}H with these cocktails were virtually identical.

Hionic-Fluor and Ultima Gold are widely used in biomedical studies because they are capable of measuring high ionic strength samples and solubilized tissue samples in strong alkaline media (PerkinElmer Inc.). Hionic-Fluor and Ultima Gold include non-negligible amounts of chemicals containing high atomic numbered elements [e.g., di-(2-

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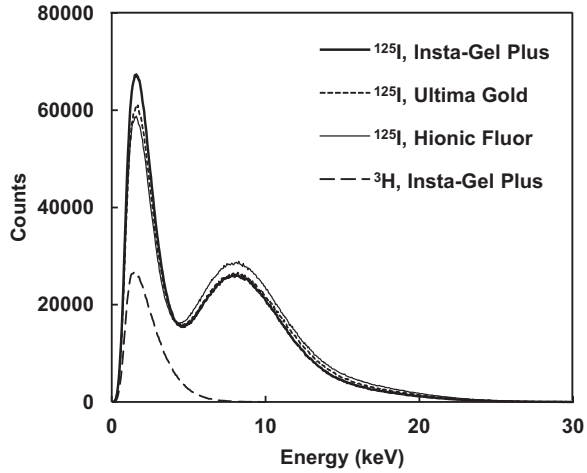

Fig. 1. Pulse height spectra of ^{125}I and ^{3}H. 0.1 mL of ^{125}I or ^{3}H was mixed with 10 mL of each scintillation cocktail and acquired for 30 min using an LSC-7400. The quenching index parameter, ESCR, was adjusted to 9.42 ± 0.05 by adding appropriate amounts of nitromethane.

ethylhexyl) phosphoric acid (DEHP), triethyl phosphate (TEP), and dioctyl sodium sulfosuccinate (DSS)], whereas Clear-sol II, Ecoscint XR, and Insta-Gel Plus do not include them (Table 1). Consequently, Hionic-Fluor and Ultima Gold contain the high atomic numbered elements sodium, sulfur, and phosphorus.

The decay of ^{125}I by electron capture (EC) emits a 35.5 keV γ-ray via the excited level of ^{125}Te into the ground state of ^{125}Te. Approximately, 93% of those γ-rays knock out an innershell electron by internal conversion (IC) with an IC electron ultimately ejected. Subsequently, the resultant vacancy of such an innershell is quickly filled with an electron by a downward transition from an outer shell. The energy gap between the shells is compensated for by the emission of X-ray or Auger electrons. There is large stochastic variability in the EC process owing to the combinations of energies for the emitted electrons from an Auger cascade. In addition, the X- and γ-rays from ^{125}I can interact with matter by the photoelectric effect and Compton scattering, thereby creating additional electrons. The contribution of these processes appears in the pulse height spectrum of the LSC analysis.

Because high atomic numbered elements interact with X-and γ-rays at a higher probability, the pulse height spectra by these scintillation cocktails will noticeably differ from ones with lower atomic numbers. Therefore the influence of high atomic numbered elements in sample cocktails should be known for facilitating the obtainment of reliable data.

The purpose of this study is to demonstrate that the peak ratio in the pulse height spectrum of ^{125}I by LSC varies depending on scintillator composition, and to clarify the mechanism. The pulse-height spectra of ^{125}I and ^{3}H were obtained from five separate types of commercially available cocktails (see Table 1). Using Insta-Gel Plus, spectrum changes via the addition of DEHP, TEP, and DSS were also observed to clarify the mechanism of the spectrum change. The change in peak ratio for ^{125}I was also evaluated by a Monte Carlo simulation based on Geant4 (Agostinelli et al., 2003). The simultaneous standardization of ^{3}H and ^{125}I radioactivity by the spillover method will be performed more precisely by quantitatively expressing the influence of high atomic numbered elements in the cocktails. Findings obtained in this study will improve the methodology for the pharmacokinetic study simultaneously using ^{3}H- and ^{125}I-labeled compounds.

2. Materials and methods

2.1. Scintillation cocktails and additives

Clear-sol II and Ecoscint XR were obtained from Nacalai Tesque, Inc., (Kyoto, Japan) and National Diagnostics Inc., (Atlanta, GA 30336), respectively. Insta-Gel Plus, Hionic-Fluor, and Ultima Gold were purchased from PerkinElmer, Inc. (Waltham, MA 02451). The ingredients and proportions in the utilized scintillation cocktails are summarized in Table 1 according to their material safety data sheets. The elemental compositions of Insta-Gel Plus, Ultima Gold, and Hionic-Fluor are listed in Table 2 (PerkinElmer Inc.). To ascertain the influence of high atomic numbered elements for the pulse height spectra, predetermined amounts of DEHP (Nacalai Tesque), TEP (Tokyo Chemical Industry Co., Ltd., Tokyo, Japan), or DSS (Tokyo Chemical Industry Co., Ltd.) were mixed with Insta-Gel Plus to produce a total cocktail volume 10 mL.

2.2. ^{3}H and ^{125}I solutions

A solution of 1-deoxyribosyl-5-([^{3}H]methyl)uracil ([methyl-^{3}H]-thymidine) (GE Healthcare, 74 GBq/mmol, 37 MBq/mL) was diluted with an aqueous solution of 1-mM thymidine. The activity was determined using an LSC (LSC-7400, Hitachi, Ltd., Tokyo, Japan) equipped with an external standard method. LSC-7400 employs an external standard channel ratio, ESCR, as the quenching index parameter. A solution of Na^{125}I (PerkinElmer Inc., carrier-free, 13.11 GBq/mL) was diluted with an aqueous solution containing 0.3-mM sodium iodide, 0.1-mM sodium thiosulfate, and 0.8-mM lithium hydroxide. The radioactivity of the^{125}I sample was then determined using an^{125}I standard solution (Japan Radioisotope Association, Tokyo, Japan). The activity was also calibrated by the sum-peak method (Pomme et al.,

Table 1
Ingredients of commercial scintillation cocktails.

Cocktail	Clear-sol II	Ecoscint XR	Hionic-Fluor	Ultima Gold	Insta-Gel Plus
Solvent	Xylene 39% 1,4-Dioxane 27%	Phenyl xylyl ethane 50–70%	1,2,4-Trimethylbenzene 40–60%	Di-isopropylnaphthalene 60–80%	1,2,4-Trimethylbenzene 40–60%
Surfactant	Polyethylene glycol nonylphenyl ether 25%	Polyethylene glycol nonylphenyl ether 20–30% Primary alcohol ethoxylate 5–10%	Polyethylene glycol nonylphenyl ether 10–20%	Polyethylene glycol nonylphenyl ether 10–20%	Polyethylene glycol nonylphenyl ether 40–60%
Others	N,N-Dimethyldodecyl-amine N-oxide 7%	Butoxy ethanol 5–10%	Di-(2-ethylhexyl)-phosphoric acid 10–20% Dioctyl sodium sulfosuccinate 2.5–10% Triethyl phosphate 2.5–10%	Di-(2-ethylhexyl)- phosphoric acid 10–20% Dioctyl sodium sulfosuccinate 1–2.5% Triethyl phosphate 1–2.5%	
Fluorescent	PPO[a] 1% Bis-MSB[b] 1%	Not noted	PPO[a] 0–1% Bis-MSB[b] 0–1%	PPO[a] 0–1% Bis-MSB[b] 0–1%	PPO[a] 0–1% Bis-MSB[b] 0–1%

[a] PPO: 2,5-Diphenyloxazole.
[b] Bis-MSB: 1,4-Bis(2-methylstylyl) benzene.