RADIONUCLIDIC SEPARATION OF RADIOACTIVE INDIUM FOR MEDICAL AND BIOLOGICAL RESEARCH APPLICATIONS FROM TARGET MATRIX BASED ON NUCLEAR REACTION OF ^{NAT}Cd (n,γ) ^{115}Cd ➔ ^{115m}In

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ABSTRACT

RADIONUCLIDIC SEPARATION OF RADIOACTIVE INDIUM FOR MEDICAL AND BIOLOGICAL RESEARCH APPLICATIONS FROM TARGET MATRIX BASED ON NUCLEAR REACTION OF ^{NAT}Cd (n,γ) ^{115}Cd ➔ ^{115m}In. Radioisotope ^{115m}In has been considered to be a very potential radioisotope for medical purposes and biological researches. Its physical properties are comparable to those of the radioisotope ^{99m}Tc. Although ^{115m}In is very potential for application in nuclear medicine and biological researches, it is not widely explored for domestic use due to domestic limitations on its production technology. Accordingly, the objective of the present works is to master a production processing technology of ^{115m}In for medical and biological research applications. As the daughter of ^{115}Cd, ^{115m}In is produced by neutron activation on cadmium target followed by separation in a radioisotope generator based on nuclear reaction of ^{114}Cd (n,γ) ^{115}Cd $\rightarrow$ ^{115m}In. In this study, natural CdO was used as a target while the irradiation was carried out in the G.A. Siwabessy reactor. The separation of radioisotope ^{115m}In from the irradiated target was carried out by means of solvent extraction and anion exchange column chromatography. In terms of solvent extraction, the post-irradiated target solution was extracted using two extractants namely 8-hydroxy-quinoline in chloroform and 2-ethylhexyl-phosphate in toluene. The resulting radioindium(III)-organo-complex was then stripped from the organic phase to release the radioisotope ^{115m}In. Meanwhile in anion exchange column chromatography, the cadmium fraction in the post-irradiated target solution was conditioned to form anion complex, CdI_4^{2-}, which was then bound on AG 1X8 (Cl^-, 100 – 200 mesh) resin column. The formed ^{115m}In, the daughter of ^{115}Cd, in the form of $^{115m}In^{3+}$ was then eluted from the column using 0.05 M HCl. It was found that the radioactive indium obtained from the solvent extraction using 8-hydroxyquinoline in chloroform was chemically contaminated by the extractant, while that obtained from the solvent extraction using 2-ethylhexyl-phosphate in toluene was significantly contaminated by ^{115}Cd. The anion exchange column chromatography was found to be the best method for separation of ^{115m}In from post-irradiated target solution because this method produced pure ^{115m}In. This was indicated by the resulting ^{115m}In fraction that gave a mono-energetic γ-ray spectrum peaking at 336 keV and a half-life of 4.486 hours which were related to ^{115m}In. The quantitative aspect which was regarded as a radioactivity of the produced ^{115}Cd was found to give a fluctuated result. This result was suspected to be inflicted by irradiation parameters such as inaccuracy in irradiation time, the changes of reactor power and neutron flux as well as inter-irradiation-position load, which varied from one irradiation to another irradiation.

Key words : Radioisotope ^{115m}In, ^{115}Cd – ^{115m}In generator, Radioisotope separation, Anion complex CdI_4^{2-}, ^{114}Cd (n,γ) ^{115}Cd → ^{115m} In nuclear reaction

ABSTRAK

PEMISAHAN RADIONUKLIDA INDIUM RADIOAKTIF UNTUK APLIKASI MEDIS DAN PENELITIAN BIOLOGIS DARI MATRIK TARGET BERBASIS REAKSI LNTI ^{NAT}Cd (n,γ) ^{115}Cd ➔ ^{115m}In. Radioisotop ^{115m}In merupakan satu radioisotop medis yang sangat komparatif terhadap radioisotop ^{99m}Tc dan dalam beberapa hal lebih menjadi pilihan dibandingkan radioisotop sejenisnya yaitu ^{111}In ataupun ^{113m}In. Walaupun mempunyai potensi penggunaan yang besar untuk pemanfaatan dalam bidang biologi dan kedokteran nuklir namun penggunaan di lingkungan domestik belum dapat ditawarkan kepada pihak pemakai karena teknologi prosesnya belum dikuasai. Dihasilkan sebagai radioisotop hasil peluruhan ^{115}Cd, radioisotop ^{115m}In berpotensi dibuat melalui iradiasi bahan sasaran kadmium di reaktor G.A. Siwabessy diikuti dengan pemisahan dalam sistem generator radioisotop berdasarkan reaksi inti ^{114}Cd (n,γ) ^{115}Cd → ^{115m}In. Sehubungan dengan itu, dalam penelitian ini teknik pemisahan radioindium dari matrik hasil iradiasi kadmium alam dalam bentuk CdO dipelajari sebagai satu tahapan dari upaya penguasaan teknologi proses radioisotop ^{115m}In untuk aplikasi di bidang biologi dan kedokteran nuklir. Percobaan pemisahan fraksi radioindium dengan teknik ekstraksi pelarut dilakukan melalui pembentukan komplek organo-indium(III) dan dengan teknik kromatografi kolom penukaran anion melalui pembentukan komplek anion tetraiodokadmium(II). Proses ekstraksi dengan ekstraktan 8-hidroksikuinolin dalam kloroform menghasilkan fraksi radioindium-^{115m}In dengan kemurnian radionuklida yang baik tetapi secara kimia terkontaminasi oleh spesi ligan organiknya. Ekstraksi menggunakan ekstrantan 2-etilheksil-fosfat dalam toluene menghasilkan fraksi radioindium-^{115m}In yang terkontaminasi secara signifikan oleh radionuklida induknya. Pemisahan menggunakan kolom kromatografi penukaran anion dengan resin AG 1X8 (Cl^-, 100 – 200 mesh) memberikan produk pemisahan yang lebih memuaskan. Secara kualitatif, fraksi radioindium hasil pemisahan diperiksa dengan spektrometri radiasi γ dan pengukuran waktu paruh. Dihasilkan spektrum radiasi γ dengan puncak tunggal pada 336 keV serta waktu paruh sekitar 4,486 jam, sesuai dengan karakteristika radioisotop ^{115m}In. Aspek kuantitatif yang dipertimbangkan terhadap radioisotop induknya menunjukkan bahwa keradioaktivan ^{115}Cd yang dihasilkan dipengaruhi oleh berbagai variabel teknis selama iradiasi, antara lain ketidaktepatan waktu iradiasi, perubahan daya reaktor dan fluks neutron termal selama proses iradiasi serta variabel beban sasaran antar posisi iradiasi yang berbeda antara satu proses iradiasi dengan proses iradiasi lainnya.

Kata kunci : Radioisotop ^{115m}In, Generator ^{115}Cd – ^{115m}In, Pemisahan radioisotop, Komplek anion CdI_4^{2-}, Reaksi inti ^{114}Cd (n,γ) ^{115}Cd → ^{115m} In.

INTRODUCTION

In-115m, an artificial radioisotope, is considered to be very potential for application in nuclear medicine and biological researches [1,2,3]. It has a half-life of about 4.5 hours and decays by isomeric transition emitting γ-ray of 366 keV (~95 %) and by β decay emitting β-ray of 0.86 MeV (~5 %). The emitted β-ray has low contribution on radiation dose but it is strong enough to kill cancer cells so that it can be used for cancer therapy [1,2]. Radioisotope ^{115m}In is also more favourable compared to other In radioisotopes due to its physical properties. ^{111}In for example, a cyclotron produced radioisotope, has multi-γ-rays (245.40 keV, 171.28 keV and 150.81 keV) and a longer half-life (2.8047 days), while ^{113m}In has a shorter half-life (1.658 hours) but higher energy of γ-ray (392 keV). ^{115m}In is also comparable to the most popular medical diagnostic radioisotope ^{99m}Tc but with a simpler chemical property as it has only one prominent oxidation state in aqueous solution [2]. In general, all substrates those are suitable to be labelled with ^{99m}Tc are also suitable to be labelled with ^{115m}In. The labelled substrates are potentially used for diagnosis of various tumours, cancers, intravascular infections and haematological damage detection due to its γ-ray emission as well as for therapeutic treatment due to its β-ray emission.

In relation to its potential use for research and medical purposes, the production technology for ^{115m}In is necessary to be mastered accordingly. The present works aim to study the separation method of radioactive indium from a matrix of post-irradiated natural cadmium oxide (CdO) as the target material. This study was a preliminary effort to master a production technology of ^{115m}In by means of $^{115}Cd/^{115m}In$ generator system based on nuclear reaction of $^{114}Cd\ (n,\gamma)\ ^{115}Cd \rightarrow\ ^{115m}In$ where the ^{115}Cd was produced by thermal neutron activation. The resulted radioactive ^{115m}In will hopefully be useful in molecule labelling in order to obtain suitable preparation for nuclear biology and medical applications, either for diagnosis or for therapeutic purposes.

In a radioisotope generator system, the ^{115m}In is separated from the matrix of its parent, ^{115}Cd ($T_{1/2}$ = 53.46 h, $E_{\beta\ max}$ = 1,1 MeV, E_γ = 527.9 keV, 492.3 keV, 260.9 keV and 231.4 keV). The parent, ^{115}Cd, can be produced by either thermal neutron activation in a nuclear reactor based on nuclear reaction of $^{114}Cd\ (n,\gamma)\ ^{115}Cd$ [1,4] or bremstrahlung radiation activation in an electron accelerator based on nuclear reaction of ^{116}Cd (γ,n) ^{115}Cd [3]. The G.A. Siwabessy research reactor located at BATAN Serpong is capable for production of ^{115}Cd based on nuclear reaction of ^{114}Cd (n,γ) ^{115}Cd by using enriched ^{114}Cd as a target material. However, in a study on the separation of ^{115m}In from the matrix of post-irradiated cadmium target, the use of enriched ^{114}Cd target is uneconomic because it is very expensive and has to be imported. The use of natural cadmium, in the form of Cd-metal, $Cd(NO_3)_2$ or CdO, is therefore preferred [1,2,3,4].

The separation of radioactive indium from the matrix of neutron irradiated natural CdO was carried out using two separation methods i.e. solvent extraction and anion exchange column chromatography. In solvent extraction, the separation of indium from cadmium is based on the ability of indium at a certain condition to form an In(III)-organoligand complex which is more soluble in organic solvent compared to in aqueous solution. In contrary with In(III), Cd(II) is not expected to form an organoligand complex at the above-mentioned condition, therefore it is expected to remain in the aqueous solution. Whereas the separation of indium from cadmium using anion exchange column chromatography is based on the ability of Cd(II) in forming an inorganic anion complex of Cd(II) at a certain condition. The anion complex of Cd(II) is then bound to anion exchanger resin. As In(III) is expected not to form an anion complex at the above-mentioned condition, therefore it is not bound to anion exchanger resin and it can be easily eluted from the column. The result of the present study is expected to be applicable in the production of ^{115m}In based on nuclear reaction of ^{114}Cd (n,γ) ^{115}Cd → ^{115m}In [1,2,4] as well as in the production of ^{111}In based on nuclear reaction of ^{112}Cd (p,2n) ^{111}In [5,6].

EXPERIMENTAL

Material and equipment

Unless otherwise mentioned, all materials were p.a. grade and used without further purifications. Cadmium oxide powder, CdO, 8-hydroxyquinoline, chloroform, 2-ethylhexyl-phosphate, toluene and all other chemicals used in the presented experiments were produced by Merck. Quartz tube of irradiation grade was used as irradiation ampoule, as well as inner and outer capsules for irradiation, which were made of highly pure aluminium, were produced by local company. Irradiation of the target was carried out in the Central Irradiation Position (CIP) of the G.A. Siwabessy reactor, BATAN Serpong. AG 1X8 (Cl^-, 100 – 200 mesh) resin was used as stationary phase in column chromatographic separation. De-mineralized water (aqua DM) was available from Water Purification Facility at the Center of Multi Purpose Reactor, BATAN, Serpong.

Separation of indium radioisotope from irradiated cadmium target by means of solvent extraction was carried out using vacuum-pulled extraction system, whereas by means of ion exchange column chromatography was performed using Econo-column 737-1010 (Bio-Rad Laboratories, USA). A gamma-spectrometry system equipped with a multi channel analyzer (Canberra 1000) and an HP-Ge detector (Canberra Industries) was used for radionuclidic analysis. Prior to its use, the spectrometer was calibrated using standard sealed-sources of ^{133}Ba (302.85 and 356.01 keV), ^{137}Cs (661.64 keV) and ^{60}Co (1173.23 and 1332.51 keV) from Du Pont. A micro pipette (5 μL, Ependorf) was used for sampling the radioactive sample of interest. Absorption spectra in ultra violet region were prepared using a UV/Vis Spectrophotometer model V-550 (JASCO).

Target preparation and post-irradiation treatment

The scheme of target preparation and irradiation followed by the treatment of the post-irradiated target is presented in Figure 1 [7]. This procedure was adopted from a procedure previously reported by Ehrhardt *et al.* [2]. An amount of CdO powder, weighed between 75 – 200 g, was carefully placed in a quartz ampoule specifically designed for solid target irradiation. The ampoule was then closed by glass welding technique and placed into an inner aluminium tube. The inner tube was closed by welding and then put into an outer tube for irradiation. The irradiation was carried out in the CIP with an average neutron flux of about $1.26 \times 10^{14} n.\ cm^{-2.}\ sec^{-1}$ [8]. After ending the irradiation, the irradiated target was then transported into a hot cell and kept for 8 – 10 hours in order to allow the short-lived radioisotope produced to decay before further processing.

The quartz ampoule was taken out of the irradiation capsule and broken at the tip to transfer its content into a 100 mL glass beaker. The irradiated CdO was then dissolved in concentrate HNO_3 (10 mL). The resulting solution was heated to dryness, and then the residue was re-dissolved with concentrate HCl (10 mL). This procedure was repeated for three times, and the residue was then re-dissolved with water (15 mL). The solution was again heated to dryness and re-dissolved with water (repeated for three times). The residue was finally re-dissolved in water (20 mL). An amount of 8.5 g KI powder was added to the solution with stirring. The resulting solution was named as Solution I and used for further experiment.

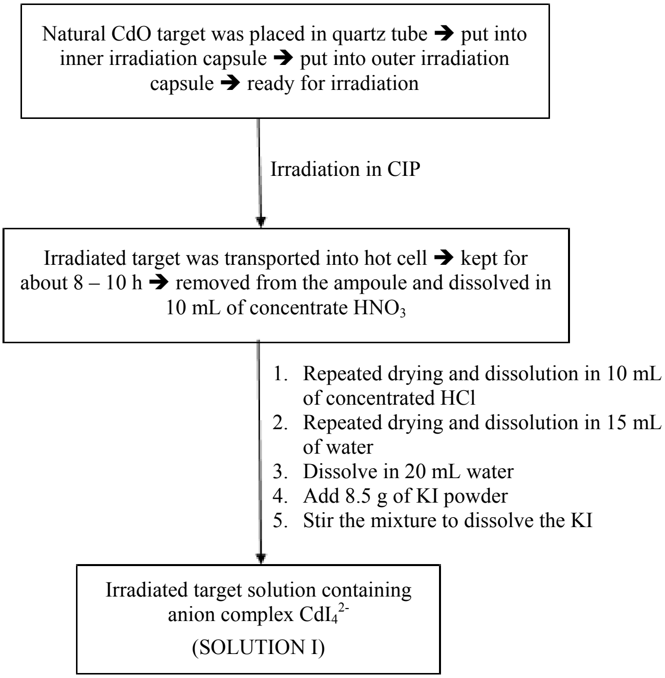

Figure 1. Target preparation and treatment of post-irradiated target.

Separation of radioactive indium by solvent extraction

The separation procedure of radioactive indium from Solution I by solvent extraction is presented in Figure 2. The extractions were conducted using two ligands as extractant, namely 8-hydroxyquinoline in chloroform and 2-ethylhexyl-phosphate in toluene. Each extractant was expected to form the corresponding organo-complex with In(III), namely In(III)-8-hydroxyquinoline and In(III)-2-ethylhexyl-phosphate respectively, which was extractable into the organic phase, while Cd(II) remained in the form of CdI_4^{2-} and stayed in the aqueous phase.

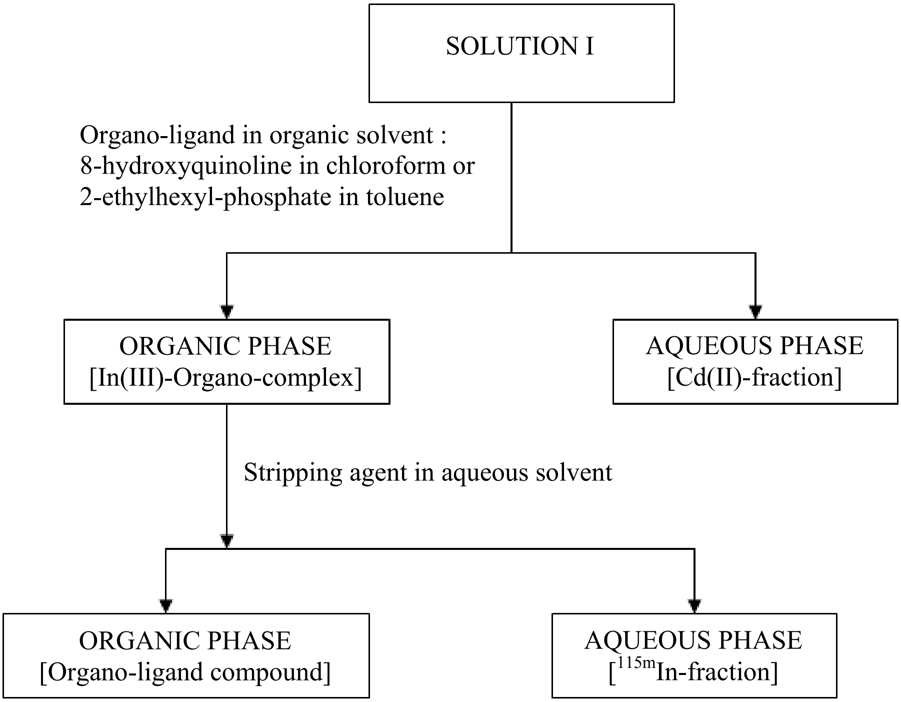

Figure 2. Scheme of extraction solvent separation.

The radioactive indium was then freed from the complex and recovered from organic phase by using stripping solutions. There were 0.1 M HCl for stripping In(III) from In(III)-8-hydroxyquinoline [9] and 0.2 M H_2SO_4 for stripping In(III) from In(III)-2-ethylhexyl-phosphate [10]. In all extraction, the volume ratio of organic to aqueous phase was 1 : 1 and the concentration of extractant was 0.01 M.

Separation of radioactive indium by anion exchange column chromatography

The separation of radioactive indium from Solution I by means of anion exchange column chromatography is shown in Figure 3 [7]. The resin used for separation was pre-conditioned by washing it with water (in batch). The washed resin was then packed into an Econo-column and primed with 0.05 M HCl and then with water.

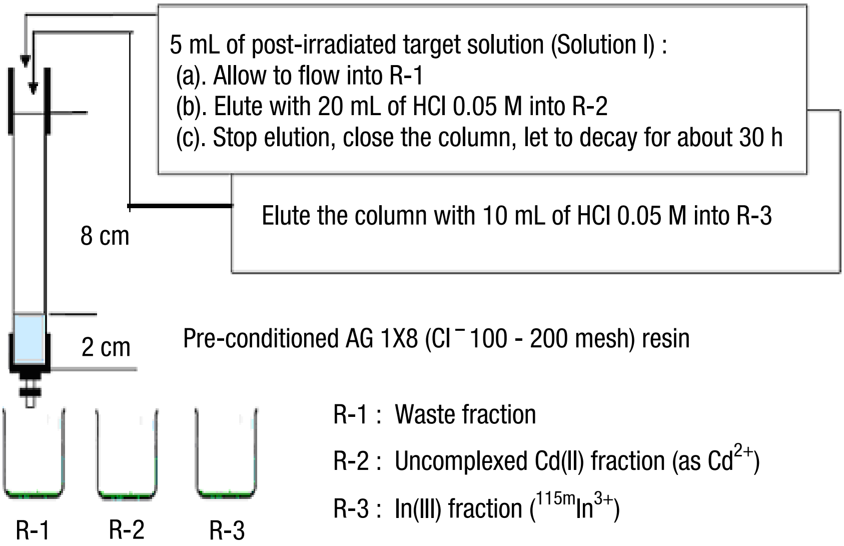

Figure 3. Separation of radioactive indium by anion exchange column chromatography.
(The radioactive indium fraction was collected in R-3)

Radionuclidic analysis

Radionuclidic analysis was conducted to the Solution I, the organic phase (extraction steps), the aqueous phase (stripping steps) and the effluents (R-2 and R-3). A 5 μL of sample was pipetted out, spotted and absorbed on 2-cm-diameter filter paper. The filter paper was dried and put into a small plastic bag and then counted by using calibrated γ-spectrometer to gain the γ-ray spectra. The half-life of the ^{115m}In sample was determined by repeated counting and the resulting counts were then plotted as a function of decay time. The resulting decay curve was compared to the theoretical calculation [7,11] using a half-life value of 4.486 h [12].

Quantitative aspect

The quantitative aspect was regarded as the radioactivity of radioactive cadmium fraction assuming that all the radioactive cadmium was in the form of ^{115}Cd. For this reason the quantitative measurement was performed at the E_γ of about 527.9 keV, which is the highest intensity of γ-ray emitted by ^{115}Cd. The measurement of efficiency as a function of E_γ was calibrated at 5 different rack positions using point-sealed sources namely ^{133}Ba (302.85 and 356.01 keV), ^{137}Cs (661.64 keV) and ^{60}Co (1173.23 and 1332.51 keV) [13]. The sample measurement was carried out at the first rack that gave the highest efficiency.

A sample of Solution I (5 μL) was spotted on a 2-cm diameter filter paper as mentioned before and counted at about 527.9 keV (the highest intensity of E_γ of ^{115}Cd). The radioactivity of ^{115}Cd at the end of irradiation (EOI) was calculated by applying correction factors of time, sample volume as well as detector efficiency. The results were then compared to the theoretical values which were obtained from an internally-improved calculation program [14].

RESULT AND DISCUSSION

Natural cadmium comprises eight isotopes. Their natural abundance and (n,γ)-nuclear reaction products are shown in Table 1 [12,15]. There are nine radioisotopes and three stable isotopes of cadmium those can be produced by (n,γ)-nuclear reaction on natural cadmium target. However, based on the combination of natural abundance (τ), thermal neutron cross section (σ) and half-life of the corresponding (n,γ)-produced radioisotopes ($T_{1/2}$), the ^{115}Cd radioisotope will be the most dominant product compared to the others.

The production of cadmium radioisotopes of longer half-life (^{113}Cd, ^{113m}Cd and ^{109}Cd) will be insignificant in the applied duration time of irradiation, whereas the shorter half-life of cadmium radioisotopes (^{107}Cd, ^{117m}Cd, ^{117}Cd and ^{111m}Cd) will quickly decay and almost none of them will be left by the end of the process. The half-life of ^{115m}Cd (44.6 days) is almost 20 times higher than that of ^{115}Cd (53.46 hours), whereas the (n,γ)-cross section for the formation of ^{115m}Cd is about one-tenth to that of ^{115}Cd (0.036 barn and 0.300 barn respectively), so it can be expected that the ^{115m}Cd radioisotope produced from a (n,γ) reaction on natural cadmium target will be negligible compared to the resulting ^{115}Cd.

Tabel 1. Products of (n,γ)-nuclear reaction on natural cadmium.

NATURAL ISOTOPES OF Cd			(n,γ)-NUCLEAR REACTION PRODUCT			
ISOTOPE	τ [%] [15]	σ (n,γ) (barn) [15]	ISOTOPE	$T_{1/2}$ [12]	E_γ (keV) [12]	I_γ (%) [12]
Cd-106	1.25	1.0	Cd-107	65 h	828.9	0.17
Cd-108	0.89	1.1	Cd-109	462.6 d	---	---
Cd-110	12.49	0.14	Cd-111m	48.54 min	245.395	94.5
					150.824	29.1
		11	Cd-111	Stable	---	---
Cd-111	12.80	24	Cd-112	Stable	---	---
Cd-112	24.13	0.04	Cd-113m	14.1 y	263.7	0.023
		2.2	Cd-113	7.7×10^{15} y	---	---
Cd-113	12.22	2.06×10^4	Cd-114	Stable	---	---
Cd-114	28.73	0.036	Cd-115m	44.6 d	933.8	2.0
		0.300	Cd-115	53.46 h	527.9	27.45
					492.3	8.03
					260.89	1.94
					231.44	0.74
Cd-116	7.49	0.025	Cd-117m	3.36 h	564.397	14.7
					860.41	7.9
					748.05	4.5
		0.050	Cd-117	2.49 h	273.349	28
					344.459	17.9
					434.19	9.8

From the separation of radioactive indium by means of solvent extraction method using 8-hydroxyquinoline in chloroform, it was observed that the extracted fraction only showed a single peak of γ-ray at 336 keV which agreed with the peak of ^{115m}In. There were no γ-ray peaks of any radioactive cadmium observed. These phenomena indicated that only In(III), as previously predicted, formed an organic complex with 8-hydroxyquinoline. The ^{115m}In was then recovered from the organic phase by stripping into 0.1 M HCl followed by UV-spectrophotometric observation on the hydrochloric acid phase. The UV-absorption spectrum of the hydrochloric acid phase is shown in Figure 4.A., appeared to be very similar to that of 8-hydroxyquinoline in 1 M HCl (Figure 4.B), indicating that 8-hydroxyquinoline was also extracted into the hydrochloric acid phase. The peak of 8-hydroxyquinoline (365 nm) was also observed from a mixture of Cd^{2+} and 8-hydroxyquinoline in 1 M HCl, whereas the other 8-hydroxyquinoline's peak was covered by the peak of the Cd^{2+} [9] (Figure 4.C). The Cd^{2+} solution has its own single-absorption peak at 301 nm [9,16] as shown in Figure 4.D.

When ^{115m}In in the organic phase was stripped out with saline solution, it was found that no absorption peak of 8-hydroxyquinoline was observed on the UV-absorption spectra of the saline fraction. The resulting absorption curve (Figure 4.E) was very similar to that of In^{3+} in saline solution (Figure 4.F) indicating that the 8-hydroxyquinoline was not re-extracted into saline fraction. But the observation on γ-ray spectra of the saline fraction did not show the existence of ^{115m}In in the saline fraction. The above-mentioned data indicated that the system of 8-hydroxyquinoline - chloroform was not suitable to be used for separating ^{115m}In from the post-irradiated cadmium matrix (Solution I). Based on Figure 4.E and Figure 4.F, it was also found that In^{3+} did not give any absorption peak in the UV-region [9]. Thus, the ^{115m}In content in saline fraction had to be detected by its mono-energetic γ-ray with E_γ of 336 keV.

The solvent extraction for the separation of Zn(II) - Cu(II) matrix by using 2-ethylhexyl-phosphate in toluene had been reported to give satisfied results [10]. In this work, however, both ^{115m}In and ^{115}Cd were extracted into the organic phase. When the organic phase was stripped with 0.2 M H_2SO_4, the ^{115m}In was recovered in the acid phase but a significant amount of ^{115}Cd was also found as a contaminant in the final product of ^{115m}In. This was presumably due to the reaction between Cd(II) and 2-ethylhexyl-phosphate to form Cd(II)-2-ethylhexyl-phosphate, which was then co-extracted into the organic phase. Based on the above results, it can be confirmed that 2-ethylhexyl-phosphate–toluene system was not suitable for extracting ^{115m}In from the post-irradiated cadmium matrix (Solution I).

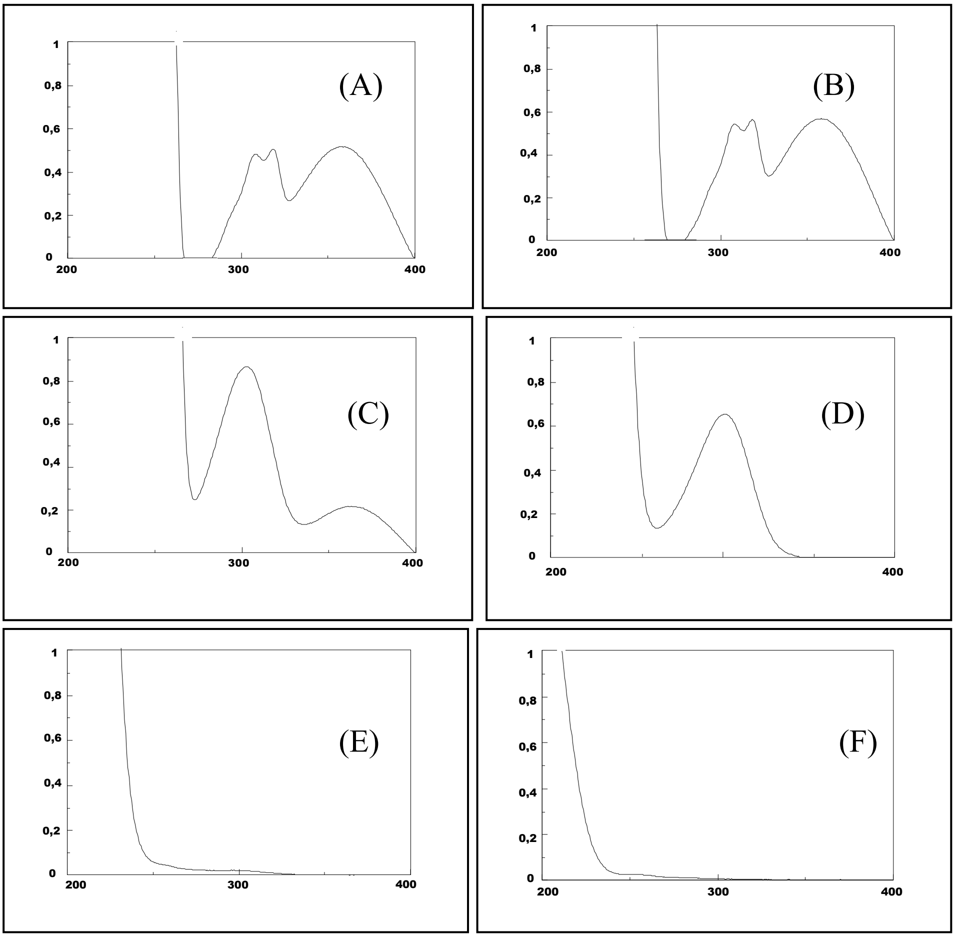

Figure 4. The UV-absorption spectra of six solution related to the solvent extraction of ^{115m}In from ^{115}Cd using 8-hydroxyquinoline-$CHCl_3$ system.

(A). HCl 0.1 M fraction of stripping step.
(B). Solution of 8-hydroxyquinoline in HCl 1 M.
(C). Solution of (8-hydroxyquinoline + Cd^{2+}) mixture.
(D). Solution of Cd^{2+} standard.
(E). Saline fraction of stripping step.
(F). Solution of In^{3+} standard.

Separation procedure by means of anion exchange column chromatography was adopted from the method reported by Ehrhardt [2]. This separation is based on the chemical reactions shown in Equation 1 and 2 [7].

$$[Cd/Cd^*]^{2+}{}_{(aq)} \quad + \quad 4\ KI_{(aq)} \rightarrow [Cd/Cd^*]I_4{}^{2-}{}_{(aq)} \quad + \quad 4K^+{}_{(aq)} \ \ldots\ldots\ (Eq.\ 1)$$

$$2\ [AG\ 1X8]Cl_{(s)} + [Cd/Cd^*]I_4{}^{2-}{}_{(aq)} \rightarrow [AG\ 1X8]_2[Cd/Cd^*]I_{4\,(s)} + 2Cl^-{}_{(aq)} \ \ldots\ldots\ (Eq.\ 2)$$

The radioactive indium was maintained in the form of trivalent cation In^{3+} whereas the radioactive cadmium was conditioned in an anion $CdI_4{}^{2-}$ form that can be bound onto the anion exchanger resin. The radioactive anion complex of $^{115}CdI_4{}^{2-}$ was then allowed to decay in column to give ^{115m}In, in the form of $^{115m}In^{3+}$ that could be eluted from the column using 0.05 M HCl. The γ-ray spectra of the resulting eluate was then measured in order to detect the existence of ^{115m}In as a final product and ^{115}Cd as a potential contaminant.

The γ-ray spectra of Solution I, R-2 and R-3 fractions (see Figure 3) are presented in Figure 5. The areas of γ-ray higher than 633 keV and that lower than 220 keV were ignored because no single peak was observed in those areas. It can be seen from spectrum 5.A (Solution I), several γ-ray energies are clearly identified and related to ^{115}Cd in agreement with the data presented in Table 1, *i.e* 528, 492, 261 and 231.5 keV. The peak of 336 keV is emitted from radioactive ^{115m}In as the daughter of ^{115}Cd, so that it will always be observed together with the peaks of ^{115}Cd. This result was as expected, that is no other radioactive cadmium other than ^{115}Cd was found in irradiated natural cadmium matrix.

The γ-ray spectra of R-2 fraction is presented in Figure 5.B. This spectrum was found to be very similar to the spectrum of Solution I (Figure 5.A). This result indicated that [Cd/Cd*](II) in Solution I was not completely in the form of anion complex of tetraiodocadmium-(II), $^{115}CdI_4{}^{2-}$. A part of it was still in the form of $^{115}Cd^{2+}$ that was not bound on the resin but eluted out of the column [7]. This agreed with the observed ^{115}Cd in the organic phase in the extraction using 2-ethylhexyl-phosphate–toluene system. The reason was that the formation of anion complex $^{115}CdI_4{}^{2-}$ was incomplete. In the case of chromatographic separation, the unchanged $^{115}Cd^{2+}$ was not bound by the anion exchanger resin, whereas in the case of extraction separation it reacted with 2-ethylhexyl-phosphate to form Cd(II)-2-ethylhexyl-phosphate and was extracted into the organic phase. The incompleteness of $^{115}CdI_4^{2-}$ formation perhaps was caused by unsuitable pH and mole ratio of KI to Cd^{2+} in the formation of the anion complex [7].

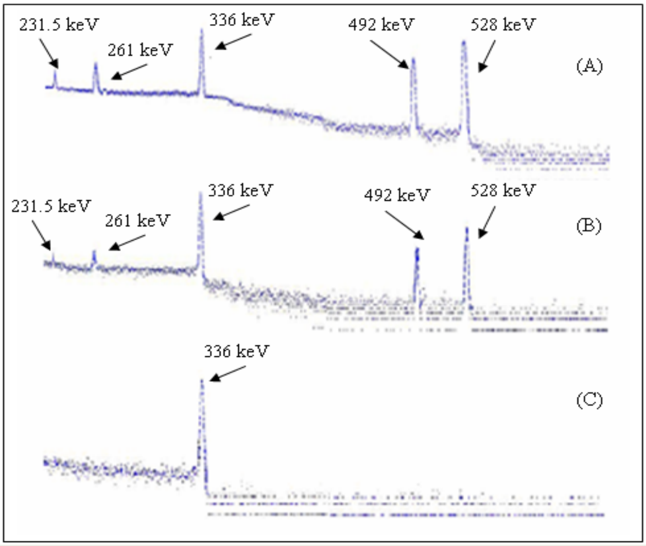

Figure 5. The γ-ray spectra of fraction separated by anion exchange column chromatography.

(A). Solution of post-irradiated target (Solution I).
(B). Solution of R-2 fraction.
(C). Solution of R-3 fraction.

The γ-ray spectrum of R-3 fraction is presented in Figure 5-C. As expected, this spectrum only gave a single γ-ray energy of 336 keV which was related to ^{115m}In. This γ-ray spectrum did not show any γ-ray energies that related to that of ^{115}Cd. This result indicated that the R-3 fraction contains high radionuclidic purity of ^{115m}In.

In order to confirm that the 336 keV peak was a peak which belong to ^{115m}In, the decay curves for determining half-life were then prepared. The decay curves of three samples of R-3 fraction containing ^{115m}In from three different processes namely R-3(A), R-3(B) and R-3(C) had been prepared according to Equation 3.

$$\ln A(t) = \ln A(0) - \lambda.t \text{ ....(Eq. 3)}$$

where A = radioactivity (counts), λ = decay constant and t = decay time

The resulting decay curves are presented in Figure 6. It can be seen from Figure 6 that the decay curves of the resulting ^{115m}In fraction of samples R-3(A), R-3(B) and R-3(C) gave a linear relationship [ln A(t) vs. t]. The slope and linearity coefficient values of these lines were close to that of theoretical ^{115m}In decay line assuming that the half-life of ^{115m}In was 4.486 h [12]. The conformity of the obtained γ-ray energy (mono-energetic of 336 keV) and the half-life (4.486 hours) confirms that the radioactive indium separated by anion exchange column chromatography was pure ^{115m}In.

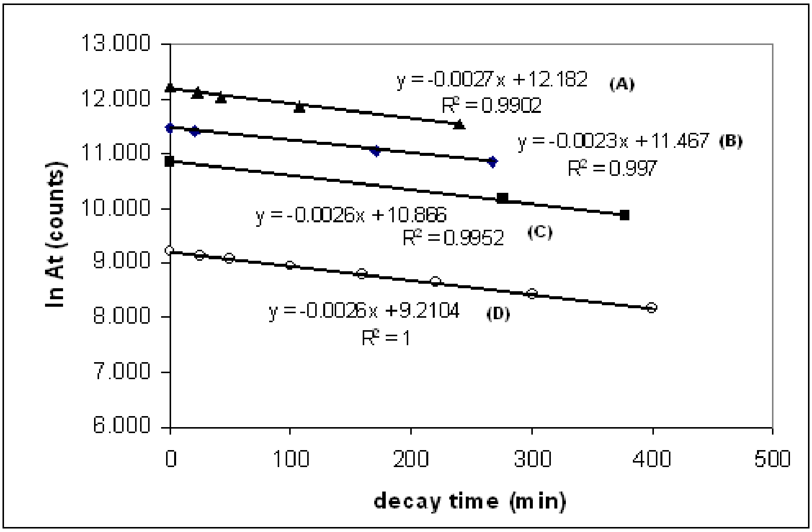

Figure 6. Decay line of radioactive indium.

(A), (B) and (C) : samples of separating-^{115m}In fractions.
(D) : theoretical decay line of ^{115m}In assuming the half-life of 4.486 h [12]

The quantitative aspect of experimental result was regarded as radioactive cadmium fraction assuming that the irradiation primary product was only ^{115}Cd. The radioactivity of ^{115}Cd produced from three processes are summarised in Table 2 [7].

Tabel 2. Radioactivity of the resulting ^{115}Cd.

VARIABLE OF EXPERIMENT	PROCESS-1	PROCESS-2	PROCESS-3
Weight of CdO target (g)	0.200	0.075	0.100
Natural Cd content (g)	0.1758	0.0657	0.0875
Iradiation time (hours)	198	256	104
Radioactivity of resulting ^{115}Cd at EOI (Ci)	0.1703	0.0849	0.0733
Specific activity of ^{115}Cd (Ci/g)	0.851	1.132	0.733
Theoretical radioactivity of ^{115}Cd (Ci)	0.2517	0.0982	0.1005
Radioactivity yield (%)	67.7	85.8	73.0

It can be seen on Table 2 that the yield of ^{115}Cd from the three different processes fluctuated significantly. This results were suspected to be inflicted by irradiation parameters applied which, in general, varied from one irradiation cycle to another and, moreover, the parameters fluctuated during a cycle of irradiation. Such parameters included the change of reactor power from time to time and the deviation of the neutron flux from the assumed value of 1.26×10^{14} n.cm^{-2}.sec^{-1} [8]. On the other hand, the average value of neutron flux on a certain irradiation position is significantly influenced by the target burden at the other position that may be different from one process to another process [17].

CONCLUSION

Neutron irradiation on natural CdO target was able to produce radioactive ^{115}Cd containing neither radioisotopic nor radionuclidic impurities. The radioactive ^{115m}In fraction, as the daughter of ^{115}Cd, could be separated from the matrix of post-irradiated target by means of anion exchange column chromatography based on the formation of anion complex CdI_4^{2-}. The column chromatographic separation method was more satisfactory compared to separation by solvent extraction using either 2-ethylhexyl-phosphate in toluene or 8-hydroxyquinoline in chloroform.

The separated radioactive indium was found to have a single peak of its γ-ray at 336 keV and a half-life of 4.486 hours which were in agreement with the γ-ray energy and the half-life of ^{115m}In. Therefore, the natural CdO can be used as a target material to produce ^{115m}In satisfactorily in terms of purity.

From three repeated experiments, the radioactivity yields of ^{115}Cd fraction were found to fluctuate between 67.7 to 85.8 %. This fluctuation was suspected as a result of irradiation parameters applied (irradiation time, the changes of reactor power and neutron flux during the irradiation, as well as the inter-irradiation-position load), which varied from one to another irradiation cycle.

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