STUDY ON SEPARATION OF ¹³⁷Cs FROM ²³⁵U FISSION PROCESS WASTE. UTILIZATION OF SILICA GEL-SUPPORTED FERROCYANIDE COMPLEX SALT FOR ¹³⁷Cs PICKING

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ABSTRACT

STUDY ON SEPARATION OF ¹³⁷Cs FROM ²³⁵U FISSION PROCESS WASTE. UTILIZATION OF SILICA GEL-SUPPORTED FERROCYANIDE COMPLEX SALT FOR ¹³⁷Cs PICKING. In connection with the potential domestic demand especially in the fields of industry and nuclear medicine, the separation of 137 Cs from 235 U fission process waste is to be of interest although its economic value could be a polemic. A preliminary study on the separation of ¹³⁷Cs from the ²³⁵U fission process waste generated in the production of ⁹⁹Mo in P.T. BATAN Teknologi, Serpong, was performed through experiments on ¹³⁷Cs picking from sample solution of the radioactive fission waste (RFW). The presented study is aimed to gain experimental data supporting utilization of the matrix of silica gel-supported ferrocyanide complex salt for the separation of ¹³⁷Cs from RFW. Subsequent step would be the recovery and purification of ¹³⁷Cs as part of production technology of ¹³⁷Cs. The RFW sample was batch-treated with the matrix of silica gel-supported ferrocyanide complex salt which was synthesized from silica gel, potassium hexacyanoferrate(II) and copper(II) chloride. The binding of radioisotopes in RFW on the matrix was observed by γ -spectrometry of the RFW solution before and during the treatment. The results showed that approximately 85% of ¹³⁷Cs could be picked from the RFW sample into the matrix. Less amount of 95 Zr and 95 Nb was bound into the matrix. 103 Ru was slightly bound into the matrix whereas $^{141/144}$ Ce and 129m Te were not. It was observed that by using 0.2 and 0.4 g of matrix for 10 ml of RFW, the amount of matrix influenced the binding quantity of 95 Zr and 95 Nb but not that of ¹³⁷Cs.

FREE TERMS: Separation of 137 Cs, 235 U fission process, Ferrocyanide complex salt, Radioactive fission waste (RFW), γ -spectrometry

ABSTRAK

STUDI PEMISAHAN ¹³⁷Cs DARI LIMBAH PROSES FISI ²³⁵U. PENGGUNAAN MATRIK SILIKA GEL-GARAM KOMPLEK FEROSIANIDA UNTUK PEMUNGUTAN ¹³⁷Cs. Berkaitan dengan potensi kebutuhan domestik, terutama di bidang industri dan kedokteran nuklir, pemisahan ¹³⁷Cs dari limbah proses fisi ²³⁵U menjadi hal yang menarik walaupun mungkin masih menjadi perdebatan apakah ekonomis atau tidak. Sebagai studi awal untuk pemisahan ¹³⁷Cs dari limbah proses fisi ²³⁵U, telah dilakukan percobaan pemungutan ¹³⁷Cs dari cuplikan limbah radioaktif proses fisi (radioactive fission waste, RFW) yang dihasilkan pada proses produksi ⁹⁹Mo di P.T. BATAN Teknologi, Serpong. Percobaan ini bertujuan untuk mendapatkan data eksperiment yang mendukung penggunaan matrik silika gel-garam komplek ferosianida untuk memisahkan ¹³⁷Cs dari RFW. Tahapan selanjutnya adalah pemungutan dan pemurnian ¹³⁷Cs sebagai bagian dari teknologi produksi ¹³⁷Cs. Cuplikan RFW diperlakukan dalam proses batch dengan matrik silika gel-garam komplek ferosianida yang dibuat dari silika gel, kalium heksasianoferat(II) dan tembaga(II) klorida.

Pengikatan radioisotop dalam RFW pada matrik diamati dengan teknik spektrometri- γ terhadap larutan RFW sebelum dan selama perlakuan dengan matrik. Hasil percobaan menunjukkan bahwa sekitar 85%¹³⁷Cs dapat terambil dari larutan RFW dan terikat pada matrik. Dalam kuantitas yang lebih rendah, radionuklida ⁹⁵Zr dan ⁹⁵Nb juga dapat terikat pada matrik. Radionuklida ¹⁰³Ru terikat pada matrik dalam jumlah yang relatif kecil sedangkan radionuklida ^{141/144}Ce dan ^{129m}Te tidak terikat. Dengan menggunakan 0,2 dan 0,4 gram matrik untuk perlakuan terhadap 10 ml larutan RFW, dapat diamati adanya pengaruh jumlah matrik terhadap kuantisasi pengikatan ⁹⁵Zr dan ⁹⁵Nb tetapi tidak terhadap pengikatan ¹³⁷Cs.

KATA KUNCI: Pemisahan ¹³⁷Cs, Proses fisi ²³⁵U, Garam komplek ferosianida, Limbah radioaktif proses fisi (RFW), Spektrometri- γ

I. INTRODUCTION

Cesium 137 (¹³⁷Cs) is one of radioisotopes having multi-purpose utilization, especially in industry and medical fields^[1]. ¹³⁷Cs is not a natural radioisotope but can be obtained as a fission product of ²³⁵U with a fission yield of about 6%^[2] meaning that about 6 atoms of ¹³⁷Cs are produced in 100 fissions of ²³⁵U. The separation of ¹³⁷Cs from fuel element waste is important due to the risk of increase of its temperature on decay being possible to give further risk on radiation safety^[3]. By agreement, however, BATAN is not allowed to process the spent fuels. On the other side, radioisotope production process by ²³⁵U fission generates radioactive waste fraction containing various radioisotopes including ¹³⁷Cs. Considering the potential domestic demand, the separation of ¹³⁷Cs from ²³⁵U fission process waste is to be of interest, although its economic value could be a polemic.

P.T. BATAN Teknologi, a domestic government factory having activities related to commercial application of nuclear techniques, has routinely performed fission process of ²³⁵U to produce radioisotopes ⁹⁹Mo and ¹³¹I. The process produces 4 fractions of fission products^[4,5], namely radioiodine fraction, radiomolybdenum fraction, radioxenon (noble gas) fraction and post-irradiated uranium fraction. The first two fission products are further processed to produce respectively ¹³¹I as Na¹³¹I solution and ⁹⁹Mo as Na₂⁹⁹MoO₄ solution respectively. Meanwhile, the other two fission products are handled as radioactive waste in which the radioxenon fraction is kept in the gas form while the post-irradiated uranium fraction, which contains some other fission products besides the unfissioned uranium, including ¹³⁷Cs, is kept in the concentrated-liquid form called radioactive fission waste (RFW).

The removal of ¹³⁷Cs from the RFW can be performed by numerous methods based on solvent extraction^[6,7], adsorption^[8], ion exchange^[9] as well as membrane techniques^[10]. Most of the reported methods are not easily applied due to economic and technical problems such as complexity and high cost of the equipment especially for small-scale experiment, and rarity of the required chemicals that it is necessary to synthesize, which is neither a simple nor cheap process. On the other side, some ion exchange resins which are known to have specificity for cesium binding mostly have very fine particle size and need other supporting agent^[11].

As a preliminary study on the separation of ¹³⁷Cs from the ²³⁵U-fission process waste, experiment on ¹³⁷Cs picking from sample solution of the RFW generated from ⁹⁹Mo production in P.T. BATAN Teknologi, Serpong, was performed. The sample of water-diluted RFW was batch-treated on a matrix of silica gel-supported ferrocyanide complex salt, which was

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synthesized from silica gel, potassium hexacyanoferrate(II) and copper(II) chloride according to method for the determination of ¹³⁷Cs in seawater reported by Chih Chieh Su et al.^[12]. The use of silica gel-supported ferrocyanide complex salt is of interest as it can be prepared easily and was claimed to show satisfactory result in the pre-concentration and determination of cesium content in natural water^[12,13].

The presented study is aimed to gain experimental data supporting utilization of silica gel-supported ferrocyanide complex salt for the separation of ¹³⁷Cs from RFW. Subsequent step would be the recovery and purification of ¹³⁷Cs as part of production technology of ¹³⁷Cs radioisotope.

II. EXPERIMENTAL

2.1. Materials and Equipment

Sample solution of RFW from Radioisotope Production Laboratory, P.T. BATAN Teknologi, Serpong, was diluted with demineralized water to obtain lower radioactivities as necessary. The demineralized water was from water purification facility, Multi Purpose Reactor Center, BATAN. The main chemical materials were p.a. reagent grade, and from Merck, unless mentioned otherwise, silica gel, copper(II) chloride and potassium hexacyanoferrate(II) trihydrates (p.a. Fluka).

The main equipment was γ -spectrometer with multi-channel analyzer Canberra 1000 complete with HP-Ge detector and software of MCA Genie 2000 VDM. The system was calibrated using point sources of ⁵⁷Co (122 keV), ¹³⁷Cs (661 keV) and ⁶⁰Co (1332 keV) from Oxford Instruments Inc. (Gamma Source Set Model S-13). Magnetic stirrer plate from Thermoline Nouva was used for stirring and mixing. Eppendorf pipette of 5 μ l was used for sampling the solution to be measured.

2.2. Preparation of Matrix of Silica Gel-Supported Ferrocyanide Complex Salt

The matrix of silica gel-supported ferrocyanide complex salt was prepared according to method used by Chih Chieh Su et al.^[12]. Approximately 5 g of silica gel was placed in a 250-ml beaker and mixed with 100 ml of 0.1 M potassium hexacyanoferrate(II) trihydrates, then stirred for about 1 hour. The mixture was then decanted or filtered. The solid product was dried in an oven at approximately 80 °C for about 24 hours.

The dried matrix of silica gel-potassium hexacyanoferrate(II) was mixed with 100 ml of 0.2 M copper(II) chloride and stirred again for about 1 hour. The mixture was then decanted or filtered and the resulting solid product was water-washed and dried in an oven at about 80 °C for not less than 24 hours to obtain the matrix of ready-used silica gel-supported ferrocyanide complex salt.

2.3. Batch-Treatment of RFW Sample with the Matrix and γ-spectrometry Analysis

10 ml of RFW sample was placed in a 100-ml beaker containing 0.2 g of the matrix. The mixture was combined and stirred at room temperature. With an interval of 1 hour each within 5 hours of stirring, the stirring was paused and the supernatant was cited carefully with 5- μ L Eppendorf pipette and sorbed on Whatman filter paper (d = 3.5 cm). The filter paper was dried in air and then placed in a plastic purse specially provided for γ -spectrometry analysis. The samples were counted at a setting position for 600 sec. The results were compared to the count data from 5 μ l of initial RFW sample that was measured using separated filter paper. The experiment was repeated using an amount of 0.4 g of the matrix.

III. RESULTS AND DISCUSSION

Some supporting or binding materials are required in preparation of resins having very fine particle size. It does not influence the ion exchanger behaviour of the resins but makes the resin more suitable for column preparation^[11,13,14]. According to Chih Chieh Su^[12] and Bellomo^[15], the ion exchanger behaviour of potassium hexacyanoferrate(II)-copper(II) chloride matrix is due to the formation of complex salt formulated as $K_2Cu_3[Fe(CN)_6]_2$, which provides the K⁺ ions to be replaced by other cations. The chemical reaction of the matrix mixture and the ion exchange capabilities for monovalent cations (M⁺), for example, can be illustrated below:

$$2K_4Fe(CN)_{6(aq)} + 3CuCl_{2(aq)} \rightarrow K_2Cu_3[Fe(CN)_6]_{2(s)} + 6KCl_{(aq)}$$
(1)
$$K_2Cu_3[Fe(CN)_3]_{-1} + 2M^+ \qquad (2)$$

$$K_{2}Cu_{3}[Fe(CN)_{6}]_{2} + 2M^{T}_{(aq)} \rightarrow Cu_{3}[Fe(CN)_{6}]_{2}^{2} \{2M^{T}\} + 2K^{T}_{(aq)}$$
(2)

The presence of silica gel in the matrix mixture is for supporting material that binds the $K_2Cu_3[Fe(CN)_6]_2$ molecules to form coarser particles as stated by Terada et al.^[13]. The mentioned function of the silica gel is comparable to the function of asbestos material in the matrix of zirconium phosphate-ammonium phosphotungstic acid complex inorganic ion exchanger as reported by Reddy et al.^[14].

The picking of radionuclides in the RFW by the matrix of silica gel-supported ferrocyanide complex salt can be observed by comparing the radionuclides and their radioactivity in the RFW before and after or during treatment with the matrix. It should be noted that in this study, it is restricted to γ -emitter radionuclides and α or β emitters are ignored. The RFW sample used in this study was originally received from the Radioisotope Production Laboratory, P.T. BATAN Teknologi, Serpong as the post-irradiated uranium fraction generated by the process schemed in Figure1.



Figure 1. Scheme of process generating radioactive fission waste (RFW) (compiled from literatures^[5,6,16,17])

According to Figure 1, it is easily understood that there are numerous radionuclides other than ¹³⁷Cs which can be found in RFW fraction. Nevertheless, some radionuclides could not be detected because the fission yields (Y) are too low, or the half-lives $(T_{1/2})$ are too short. Some fission products were not detected by γ -spectrometry because they do not emit γ -rays. It can be seen in Table 1 that various radionuclides were significantly detected by γ -spectrometry of the RFW sample before being treated with the matrix of ferrocyanide complex salt. For each of the radionuclides, the count number on energy with highest intensity (I) was further used as calculation base for the fraction picked on the matrix.

E_{γ} (keV)	Radionuclide	Number of counts	Reference data ^[18]			
			E_{γ} (keV)	I (%)	Y (%)	T _{1/2} ^(a)
80.5	Ce-144	10788	80.1	1.6	5.47	284.6 d
133.6	Ce-144	124938	133.5	11.1		
145.8	Ce-141	21938	145.4	48.2	5.80	32.5 d
497.2	Ru-103	15390	497.1	90.9	3.04	39.27 d
610.5	Ru-103	720	610.3	5.6		
512.0	Ru-106 ^(b)	4180	511.9	20.4	0.403	1.02 y
622.5	Ru-106	1472	621.8	10.7		
1050.6	Ru-106	154	1050.5	1.7		
661.9	Cs-137	13208	661.7	85.2	6.22	30.17 y
696.8	Te-129m ^(c)	3502	695.9	3.3	0.127	33.6 d
			695.7	3.1		
724.5	Zr-95	48460	724.2	44.1	6.49	64 d
757.0	Zr-95	56563	756.7	54.5		
766.1	Nb-95 ^(d)	203537	765.8	99.8	6.49	34.97 d

Table 1 Typical resume of γ -spectrometry of RFW before being treated with the matrix of ferrocyanide complex salt

Notes:

d = days, y = yearsa)

According to other references^[19,20] ¹⁰⁶Ru is a β -emitter emitting no γ -rays. The observing γ -rays are emitted by the daughter of ¹⁰⁶Ru, i.e. ¹⁰⁶Rh. The counts at 696.8 keV as stated from ^{129m}Te probably overlap with the counts of activity at b)

c) 697 keV springing from ¹⁴⁴Pr, i.e. the daughter of ¹⁴⁴Ce by β -decay.

The counts of activity at 766.1 keV constitute accumulation of fission-produced ⁹⁵Nb and ⁹⁵Nb d) originated by β -decay of ⁹⁵Zr.

When the RFW solution was mixed and stirred with the matrix of silica gel-supported ferrocyanide complex salt, some radioisotopes appeared to be unbound on the matrix. This phenomenon was indicated by the fact that the count number of the cited RFW sample from the mixture at the concerning energy remained relatively unchanged as compared to that of equivolume initial RFW sample. By treating 10 ml of RFW solution on 0.2 g of matrix, it can be seen from Figure 2(a) that neither radioactivity of 144 Ce nor 141 Ce in the solution decreased, indicating that cerium was not picked into the matrix.

In the same system, it is shown in Figure 2(b) that like the cerium, tellurium is not picked into the matrix as indicated by the radioactivity of the ^{129m}Te in the solution which remains relatively unchanged. On the contrary, the radioactivity of ¹³⁷Cs in the RFW solution sharply dropped indicating that the cesium was bound and picked into the matrix. However, the capability of the matrix to bind radioisotopes from the RFW was observed not to be specific for cesium. It can be seen in Figure 2(c) that both ⁹⁵Zr and ⁹⁵Nb were significantly bound and picked into the matrix.

There was a slight difference in radioactivity profile between ¹⁰³Ru and ¹⁰⁶Ru which ought not to happen because the interaction between radioisotopes and the matrix is a chemical interaction. The observing data shown in Figure 2(d) indicates that the radioactivity of ¹⁰³Ru represents the binding of ruthenium while that of ¹⁰⁶Ru represents the binding of rhodium rather than ruthenium as ¹⁰⁶Ru decays by β -ray emission to ¹⁰⁶Rh, a non-fission product radioisotope emitting γ -rays of 511.9, 621.8 and 1050.5 keV (see notes below Table 1). Based on this reason, it can be stated that ruthenium was slightly bound on the matrix but not rhodium.

In order to see whether the quantity of binding would be affected by the amount of matrix, the RFW samples were treated with the matrix in the amounts of 0.2 and 0.4 g for a similar volume of RFW, i.e. 10 ml respectively. The reason to utilize such amounts of matrix is based on expectation that doubling the amount of matrix can give significant information whether the amount of matrix will affect the binding of radioisotopes or not. Meanwhile those amounts of matrix can be completely submerged in 10 ml of RFW solution. The evaluation was performed for cesium (¹³⁷Cs), zirconium (⁹⁵Zr) and niobium (⁹⁵Nb) which were significantly bound on the matrix by means of cation exchange mechanism based on reaction (2), as follow:

$$m \operatorname{K}_{2}\operatorname{Cu}_{3}[\operatorname{Fe}(\operatorname{CN})_{6}]_{2} + 2\operatorname{M}^{m_{+}}_{(aq)} \to \{\operatorname{Cu}_{3}[\operatorname{Fe}(\operatorname{CN})_{6}]_{2}^{2^{-}}\}_{m} \{2\operatorname{M}^{m_{+}}\} + 2m \operatorname{K}^{+}_{(aq)}$$
(3)

where M^{m+} represents ions of ¹³⁷Cs, ⁹⁵Zr and ⁹⁵Nb respectively.





Figure 2. Radioactivity of some radionuclides in RFW sample before and during the treatment with 0.2 g of matrix

The results presented in Figure 3 show that the binding of ¹³⁷Cs was not affected by the amount of the matrix, whether 0.2 or 0.4 g, but the binding quantity of ⁹⁵Zr or ⁹⁵Nb was significantly affected. The data indicates that the exchange between K⁺ in the solid matrix with Cs⁺ in RFW solution occurs more easily than with cations of zirconium and niobium which are usually in the forms of Zr⁴⁺ and Nb³⁺ or Nb^{5+[21]}. The Cs⁺ and K⁺ ions not only have similarity in charge but also they are members of the same group in the periodic table, so they resemble each other in chemical behaviour.





Figure 3. The influence of radioisotope binding on the amount of the matrix by using 10 ml of RFW solution (Dotted line: measured radioactivity remaining in the solution. Solid line: calculated radioactivity bound in the matrix)

In general, the equilibrium of radioisotope binding has been achieved in 2 hours of mixing and stirring the matrix with the RFW solution. Approximately 85% of the ¹³⁷Cs was picked into the matrix, while the ⁹⁵Zr and ⁹⁵Nb were bound in the amount less than that of ¹³⁷Cs. If the resulting data is further considered, a selective stripping method will be necessary to pick the ¹³⁷Cs and thus to separate the ¹³⁷Cs from ⁹⁵Zr and ⁹⁵Nb. Perhaps it can be performed by loading the matrix into a chromatographic column followed by elution with suitable solution in which the ¹³⁷Cs⁺ will be eluted first before the other two. This experiment is now still in progress.

IV. CONCLUSIONS

The matrix synthesized from silica gel, potassium hexacyanoferrate(II) and copper(II) chloride was supposed to be a complex salt, i.e. $K_2Cu_3[Fe(CN)_6]_2$, supported by silica gel to form coarser particles. The matrix shows capability to bind some ²³⁵U fission-produced radioisotopes from RFW solution.

The 137 Cs from the RFW sample can be bound on the matrix in a quantity of about 85%, while the 95 Zr and 95 Nb was bound in an amount less than that of 137 Cs. Radioisotope of ruthenium, represented by 103 Ru, was slightly bound on the matrix, while ${}^{144/141}$ Ce and 129m Te were not.

The amounts of 0.2 and 0.4 g of the matrix significantly affected the quantities of the 95 Zr and 95 Nb picked from the RFW into the matrix but not the 137 Cs. As the capability of the matrix to bind radioisotopes is not specific for cesium, the picking of 137 Cs from the matrix will need a specific stripping method to separate the 137 Cs from other radioisotopes bound in the matrix.

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