Solvent Extraction of Cesium Using DtBuDB18C6 into Various Organic Solvents

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High concentration of Cs is present in high-level radioactive waste. It is well-known that Cs is an alkali element and difficult to extract completely into an organic phase. Crown ether compounds are widely available for Cs extractants; DtBuDB18C6 (di-t-butyl-dibenzo-18crown6), was used in this study. Organic solvents used for the industrial applications, such as n-dodecane and 1-octanol, have low solubility concerning the compound; other solvents were employed and tested. In this study, ketone-, ether-, and ester-type solvents showed high solubility for DtBuDB18C6 and DtBuDB18C6, when dissolved in ketones and alcohols, exhibited relatively high Cs distribution ratios (D(Cs)), closely to 10.

1. Introduction

Cs is one of the elements that is required to be removed from high-level radioactive waste (HLW) [1-5] because of its relatively high yield in fission products, 30-year half-life of 137Cs, emission of strong gamma (661.7 keV) and beta rays (512 keV), and strong heat generation in all fission products and actinides. The heat generated by Cs is problematic when producing vitrified waste or MA fuels. Cs easily enters the environment because of the relatively low boiling point of Cs metals; therefore, radioactivity from Cs becomes a serious environmental problem in case of the nuclear accidents. Hence, recovery of Cs from HLW and its introduction into interim storage is an important task for the atomic energy field. In addition, the development of a chemical recovery method for Cs and transmutation to 135Cs, which shows very long half-life (T1/2 = 2.3 × 106 y), is currently one of the most important tasks [3]. Solid-liquid separation methods, such as adsorption on zeolites and ferrocyanides, have been widely applied to recover Cs up to the present day [6-10]; however, because of the strong bond of Cs to the solid absorbents, this technique requires large volumes of effluent.

Liquid-liquid extraction methods are also useful for Cs recovery. In this case, crown ether compounds and calix-crown compounds for Cs and Sr extraction are promising [11-15]. The normal crown compounds are generally water-miscible; dibenzo-18crown6 and di-t-butyl-dibenzo-18crown6 are available for Cs extraction because of their lipophilicity and dicyclohexyl-18crown6 and di-t-butyl-dicyclohexyl-18crown6 (DtBuDCH18C6) are also useful for Sr extraction [14,15]. In previous studies, chlorinated solvents, namely 1,2-dichloroethane and chloroform, were used generally for the Cs extraction. However, these solvents are harmful to the environment, so organic solvents without Cl are preferable. Solvents, n-dodecane and 1-octanol, are used for reprocessing and partitioning processes, however, the solubility of crown compounds.
in these solvents is limited. Therefore, alternative solvents should be examined for the preparation of Cs extraction solvents.

In this study, ketone-, ether-, ester-, and alcohol-type solvents were studied for solubility of crown compounds and distribution ratios \(D(\text{Cs})\) of Cs; these diluents are composed of carbon, hydrogen, and oxygen atoms that are less of an environmental burden. The crown compound employed here was DtBuDB18C6 (see Figure 1), which is more available for solvent extraction due to its lipophilicity. Detailed \(D(\text{Cs})\) into extraction solvents, extraction capacities, and back-extraction conditions were also investigated.

![Figure 1. Structure of DtBuDB18C6.](image)

2. Experimental

2.1 Reagents

DtBuDB18C6 was purchased from Wako Pure Chemical Industries. Extractants of more than 95% purity were used for solvent extraction without further purification. Standard solutions (1,000 ppm concentration) of Li, Na, K, Rb, and Cs (Wako Pure Chemical Industries) were used for solvent extraction. Other reagents, i.e., all organic solvents and nitric acid, were of analytical grade.

2.2 Solvent extraction methods

The crown compounds were dissolved in solvents at room temperature and prepared to the desired concentration. A standard solution of each metal was diluted with 0.5 M HNO\(_3\) to an appropriate concentration. The two phases were contacted and mechanically shaken at 1,500 strokes/min for 20 min, at 25 ± 0.1°C. After centrifugation, the metal concentration in the aqueous phase was measured using either ICP-OES (SPS 3100; Seiko Instruments Inc.) or ICP-MS (SPQ 9200; Seiko Instruments Inc.). Metal concentration \([M]\) in the organic phase was calculated by subtracting the metal concentration in the aqueous phase from the initial concentration. The \(D\) values were defined as the ratio of \([M]\) in the organic phase to \([M]\) in the aqueous phase.

The back-extraction method was almost similar to the forward-extraction; the difference was the starting organic phase that included the metals. Back-extraction efficiency was obtained from the results of Cs concentration in the aqueous phase after stripping. Measurement of extraction capacity was achieved by taking an aliquot of the organic phase. After the extraction of high concentration of Cs into the organic phase, the sample was evaporated by heating. The residue after evaporation was added to 1–2 mL HNO\(_3\)–HClO\(_4\), and the organic sample was decomposed by heating. After completely decomposing, the residue was dissolved in 0.5 M HNO\(_3\) and prepared to an appropriate concentration by diluting it with 0.5 M HNO\(_3\) for ICP-MS measurement.

2.3 Measurements for physical properties of diluents

The solubility of the organic diluent in water was measured by total organic carbon analyzer (TOC-V CSN; Shimadzu Corp.). After equilibration of the sample in water, C in the organic content was measured and its percentage in the water was calculated. In order to determine the vaporization ratio of diluents, the sample solution was put into 20 mL vials (15 mmφ) without a cap. Kept at 35°C for the longest 7 h, the weight of solution was measured before and after experiments, and weight-loss ratios were obtained against
times (g/h). Dielectric constants and viscosities for all the organic solvents were measured by a dielectric constant meter (BI-870; Brookhaven Instruments Corp.) and a viscometer (SV-A/H; A&D INSTRUMENTS).

3. Results and Discussion

3.1 Basic properties of diluents

The melting points and the flash points for diluents are found in Handbook of Fine Chemicals, Sigma-Aldrich (2009-2010), and the results were plotted against their carbon numbers in structure (Figure 2(a) and (b)). Figure 2(a) shows the relation between melting points and carbon numbers, and Figure 2(b) shows that of the flash points. Here, the diluents are limited to the structures having normal alkyl groups without double bonds and phenyl groups, because the complicated chemical structures, with the functional groups having the oxygen atoms and phenyl groups, may exhibit against the rule of regular melting and flash points.

From Figure 2, each diluent group has an individual trend. Compared to the same carbon numbers, the order of melting points is alcohol > ketone > ester > hydrocarbon > ether, and that of flash points is alcohol > ketone > ester > hydrocarbon > ether. The melting points under 10°C and flash points over 70°C are preferable for the use of diluent, therefore, the ranges to use for carbon numbers in diluents are alcohol (8-12), ketone (9-12), ester (9-12), hydrocarbon (> 10), and ether (> 12). Information is available to show which structure is a useful diluent. Here, the small volume of ester may decompose due to hydrolysis during extraction in the presence of acid. No third phase formation is observed, no nasty smell can be perceived and experimental condition is 20 min shaking at 25°C, thus this effect on Cs extraction is low enough.

Figure 3(a) and (b) show the relationship between solubility in water (a) and vaporization ratio (b) against carbon numbers. Probably, numbers of carbon exhibit their lipophilicities, thus increase of carbon numbers decrease solubility in water (Figure 3(a)). Molecular weight increases with carbon numbers, so the vaporization ratio may decrease with molecular weight of organic solvent as shown in Figure 3(b). Here, solubilities of water and vaporization ratio are 0.004% and 0.0092 g/h for n-dodecane and 0.039% and 0.0062 g/h for 1-octanol, these values should be as the references.

![Figure 2](image-url)

Figure 2. Relationship between melting (a) and flash (b) points against carbon numbers of diluents. The data were obtained from Sigma-Aldrich (2009-2010).
3.2 Extraction of Cs to various solvents

Prior to the extraction experiments, the solubility of DtBuDB18C6 in 15 diluents (see in Table 1) was investigated at a room temperature. A white precipitate of the extractant, due to the low solubility in hydrocarbons, e.g., \( n \)-dodecane, was observed after mixing the sample solution. Most of the alcohol diluents exhibited the highest concentrations, limited to 0.1 M. Other solvents showed sufficient solubility, enabled to prepare 0.5 M concentration; with no precipitate was found after dissolution.

Solvent extraction of Cs into various solvents was roughly performed and the solvents used were alcohols, ethers, esters, aldehyde, and ketones. The extractant concentrations were 0.1 M for alcohol and 0.2 M for all other solvents, and 3 M HNO\(_3\) was used as the aqueous phase set-up. The experimental results are shown in Table 1; 1,2-dichloroethane and nitrobenzene showed relatively high \( D \)(Cs), 3.48 and 5.50, respectively. For all the other solvents, the \( D \)(Cs) was dependent on the solvent type. The \( D \) values in Table 1 show that alcohol- and ketone-type solvents had higher values (\( \geq 0.94 \)) than those for ester- and ether-type solvents (\( \leq 0.33 \)). Although alcohol had limited solubility, alcohols and ketones exhibited relatively high \( D \)(Cs), and were consequently chosen for the following study and examined in detail.

![Graph (a)](image-a.png)

![Graph (b)](image-b.png)

Figure 3. Relationship between solubility in water (a) and vaporization ratio (b) against carbon numbers of diluents.

| type of solvent | \( D \)(Cs) |
|-----------------|-------------|
| DtBuDB18C6 from 3 M HNO\(_3\) into various solvents. | type of solvent | DtBuDB18C6 concentration |
| 1-octanol | alcohol | 0.1 M | 1.19 |
| n-dodecane | hydrocarbon | - | - |
| 1,2-dichloroethane | chlorinated | 0.2 | 3.48 |
| nitrobenzene | aromatic | 0.2 | 5.5 |
| 1-hexanol | alcohol | 0.1 | 3.48 |
| 1-heptanol | alcohol | 0.1 | 2.92 |
| dipentylether | ether | 0.2 | 0.15 |
| dihexylether | ether | 0.2 | 0.33 |
| methyl octanoate | ester | 0.2 | 0.19 |
| methyl nonanoate | ester | 0.2 | 0.24 |
| methyl decanoate | ester | 0.2 | 0.09 |
| ethyl nonanoate | ester | 0.2 | 0.27 |
| 1-octanal | aldehyde | 0.2 | 2.12 |
| 2-octanone | ketone | 0.2 | 6.22 |
| 1-nonanal | aldehyde | 0.2 | 0.94 |
3.3 Extraction behavior of alkali metals by DtBuDB18C6 into alcohol

Alkali metals with and other than Cs were also used to elucidate extraction behavior in 1-octanol by DtBuDB18C6. The relationship between log $D$ vs. log [DtBuDB18C6] and log $D$ vs. log [HNO$_3$] is shown in Figure 4(a) and (b), respectively. Here, we use 3 M HNO$_3$, because the composition of HLW is similar to this condition. The slope values for the metals were 1.35 for Na, 1.4 for K, 1.45 for Rb, and 1.72 for Cs (Figure 4(a)). As shown in Figure 4(b), the $D$ values gradually increased up to 3 M HNO$_3$ then decreased, indicating ion-pair extraction with NO$_3^-$ for the reason of increase, and probably the competition reaction with H$^+$ for the decrease. This behavior has been previously reported [15], using an experimental condition of HNO$_3$-nitrobenzene. In the current study, the extraction trend (Figure 4) was K $>$ Cs $>$ Na $>$ Rb $>$ Li, which is different from the ionic radii trend (Li: 74, Na: 102, K: 138, Rb: 149, and Cs: 170 pm). Crown compounds can exhibit a size-fitting effect, but this does not explain the results. This trend was also different from former studies [13,16,17] that indicated the order of $D$ as K $>$ Rb $>$ Cs $>$ Na, wherein the authors stated that the highest extraction was ~149 pm of Rb. Their experimental conditions (DB18C6 in 1,2-dichloroethane with picrate ion) were different from the current study. Takeda [17] stated that different experimental conditions, especially different diluents, produce other trends for metal extraction even with the same crown compound.

The extraction of Cs into the following alcohols was investigated: 1-heptanol, 1-octanol, 2-octanol, 3-octanol, and 1-nonanol. The results of the relationship between log $D$ vs. log [DtBuDB18C6] are shown in Figure 5. The slopes in this figure were 2.37 for 1-heptanol, 1.72 for 1-octanol, 1.68 for 2-octanol, 1.81 for 3-octanol, and 2.23 for 1-nonanol. A 1 : 2 complex of Cs:DtBuDB18C6 was formed in these diluents. Note that [18] reported a slope of 0.84 with a highest $D$(Cs) of 3.5 using DB18C6 in nitrobenzene; although different slope values were obtained, and the highest $D$(Cs) (~1) under these conditions was similar that obtained by Kumar et al. The physical properties of the alcohols in the current study were different; the dielectric constants for these alcohols decreased in the following order: 1-heptanol (11.7) $>$ 1-octanol (10.66)
> 1-nonanol (8.89), and 1-octanol (10.66) > 2-octanol (8.03) > 3-octanol (5.75). In addition, the alcohol solvents had, overall, high viscosity (1-heptanol 4.98; 1-octanol 7.03; and 1-nonanol 9.96 mPa·s); other solvents are usually equal or lower than 1 mPa·s. The high viscosity of extraction solvents may be a problem for running mixer-settler experiments.

3.4 Extraction behavior of Cs by DtBuDB18C6 into ketone and aldehyde

In order to determine the extraction behavior of Cs in ketones, the same composition of ketone but with different structures (namely structural isomers) was used: 1-nonanal (nonanal), 2-nonanone, 3-nonanone, 4-nonanone, and 5-nonanone. The results (Figure 6) indicated the relationship between log $D$ vs. log [DtBuDB18C6]. Apart from the $D$ values in nonanal, almost the same $D$(Cs) and extraction behavior were observed; there is currently no explanation for this difference between nonanal and other nonanones. A $D > 1$ was necessary, at least, to set-up the separation process condition; therefore, nonanal, 2, 3, 4, and 5-nonanone should be chosen for this purpose. The specific difference of physical properties of these diluents is its solubility in water: 0.0819% for nonanal, 0.077% for 2-nonanone, 0.198% for 3-nonanone, 0.172% for 4-nonanone, and 0.211% for 5-nonanone. Other properties, i.e., viscosity, dielectric constant, and volatilization were almost the same. The solubility of the extractant in water was significantly determined by its extractability. The diluents showing high solubility in water have the potential to introduce the extractant into the aqueous phase. Here, 1-nonanal is an aldehyde and may produce carboxylic acid by self-oxidation in the presence of HNO₃. From the present work, no third phase formation and no nasty smell can be perceived after extraction, thus this effect on Cs extraction is considered to be low. The results of 1-nonanal should be taken as a reference value.

2, 3, 4, and 5-Nonanone had similar $D$(Cs) values (Figure 6). 2-Nonanone was selected because it is
easily and cheaply obtained, and $D$ were investigated mainly using different ketones with the same position of the oxygen atom as the 2-nonanone. Figure 7(a) indicates the dependence of $D_{Cs}$ on $DtBuDB18C6$ concentration in 2- or 4-ketones (2-heptanone, 4-heptanone, 2-octanone, 2-nonanone and 2-decanone). It appeared to obtain a $D_{Cs} > 10$ for 0.2 M $DtBuDB18C6$; this indicated that satisfactory extraction of Cs into the organic phase by mixer-settler extractions could be expected. The highest $D_{Cs}$ was observed in 4-heptanone; the other 2-ketones showed almost similar $D$ as the others. Figure 7(b) shows the dependence of $D_{Cs}$ on HNO$_3$ concentration for different ketones (4-heptanone, 2-nonanone, 4-nonanone, 5-nonanone and 2-decanone). Similar behavior for Cs extraction is shown in Figure 4(b); a gradual increase of $D_{Cs}$ with HNO$_3$ and a decrease from the highest value of 3 M HNO$_3$. The order of $D_{Cs}$ at 3 M HNO$_3$ was 4-heptanone > 5-nonanone > 4-nonanone > 2-nonanone > 2-decanone (Figure 7(b)). 4-Heptanone had the highest $D_{Cs}$, but it is highly soluble in water (0.267%) and is highly volatile due to its low molecular weight. In the future, concerning industrial availability, the physical properties of all ketones should be thoroughly investigated.

3.5 Extraction behavior of alkali metals by $DtBuDB18C6$ into ketone

Alkali metal extraction by $DtBuDB18C6$ in 2-nonanone was studied, and the results are shown in Figure 8. Log-log relations (Figure 8(a)) indicated slopes of 1.07 for Na, 0.944 for K, 1.89 for Rb, and 1.94 for Cs; the different slope values, or extraction reactions, can be seen in the Na-K extraction for slope 1 and the Rb-Cs extraction for slope 2. These results determined that the different extraction reactions were a 1:1 complex for Na and K and a 1:2 complex for Rb and Cs. Figure 8(a) indicates that the highest $D_{Cs}$ was near to 10 under the condition of 0.2 M $DtBuDB18C6$ in 2-nonanone from 3 M HNO$_3$; this value is greater than that reported by [15], $D_{Cs} = 3.5$. It has already been mentioned that $DtBuDB18C6$ concentration higher than 0.2 M in 2-nonanone can be prepared, so a $D_{Cs} > 10$ can be obtained under such conditions. Figure 8(b) indicates the dependence of $D$ on HNO$_3$ concentration.
A continuous increase of $D$ with HNO$_3$ concentration, without peak, in the range of 0.2–6 M HNO$_3$, was observed. There was a small difference in $D$ from those shown in Figure 4(b) because of the different organic solvents used (1-octanol and 2-nonanone).

To obtain more information about extraction in ketones, 2-decanone and 2-undecanone were used because they have heavy molecular weights, are expected to result in low volatilization; low solubility in water; low aromatic and high flash points. The experimental results using alkali metals are shown in Figure 9, which were almost the same as the results shown in Figure 8(a). Less effect from the different solvents, using 2-nonanone, 2-decanone, and 2-undecanone, was found.

Figure 8. Extraction behavior of alkali metals by DtBuDB18C6 into 2-nonanone. (a): dependence on DtBuDB18C6 concentration at 3 M HNO$_3$, (b): dependence on HNO$_3$ concentration at 0.1 M (Cs), 0.2 M (Li, Na, K and Rb) DtBuDB18C6.

Figure 9. Extraction behavior of alkali metals by DtBuDB18C6 at 3 M HNO$_3$ into 2-decanone (a) and 2-undecanone (b).
3.6 Effect of Cs extraction into organic solvent with high di-electric constant

Effect of Cs extraction on organic solvent with lipophilicity is investigated. Here, the lipophilicity in the organic phase can be obtained from their di-electric constants. The di-electric constants depend strongly on the ratio of carbon/oxygen (C/O) atom ratios. One example is that di-electric constants for 2-octanone, 2-nonanone, 2-decanone, and 2-undecanone are measured to 11.1, 9.86, 8.85, and 8.07, the increase of C/O ratios give the decrease of their di-electric constant. In addition, Cs extraction by DtBuDB18C6 is based on the ion-pair extraction, which may lead the high $D(Cs)$ into solvent having high di-electric constant. The relationship between $D(Cs)$ and $D(Sr)$ as a reference, and di-electric constants for all organic solvents are shown in Figures 10 and 11. Here, the $D(Sr)$ values are obtained using extractant, DtBuDCH18C6. A positive relationship can be seen in both figures, the organic solvents with high di-electric constants may give the high $D(Cs)$ and $D(Sr)$.

3.7 Maximum extraction concentration of Cs into organic solvent

The highest concentration of Cs in the organic phase was investigated in the diluent, 2-nonanone. The results of the extraction of high Cs concentrations are shown in Figure 12. The experimental condition of Cs concentration in the aqueous phase increased from 20 mM to a maximum of 100 mM with the fixed DtBuDB18C6 concentration (0.1 M) in the organic phase. Although Cs concentration in organic phase after extraction increased, the percentage of Cs extraction decreased with the Cs concentration in the initial aqueous one (Figure 12). The maximum Cs concentration, namely loading capacity, could not be determined because the Cs concentration in the organic phase did not stop increasing as shown in Figure 12. The theoretical value for the maximum Cs concentration is 50 mM if 100 mM DtBuDB18C6 is used, due to the molar ratio of Cs: crown to be 1:2 from the slope analysis. The highest Cs concentration in the current study was 24 mM, which did not reach the theoretical value.

![Figure 10. Relationship between $D(Cs)$ and di-electric constant for all solvents examined. Conditions, 0.1 M DtBuDB18C6 in alcohol and 0.2 M DtBuDB18C6 in other solvents, and 3 M HNO$_3$.](image1)

![Figure 11. Relationship between $D(Sr)$ and di-electric constant for all solvents examined. Conditions, 0.2 M DtBuDCH18C6 in other solvents, and 3 M HNO$_3$.](image2)
3.8 Back extraction of Cs into aqueous phase

As indicated in Figures 4(b), 7(b), and 8(b) for dependence of $D$(Cs) on HNO$_3$ concentration, the $D$ values increased with HNO$_3$ concentration because of ion-pair extraction. This means that low HNO$_3$ condition leads low $D$(Cs) value, and the condition, diluted HNO$_3$, could be used for back-extraction. Then, 0.01 M HNO$_3$ was used as the back-extraction condition in this study. At first, Cs was extracted into the organic phase with 0.1 M DtBuDB18C6 in 2-nonanone as the organic phase and 0.1 M [Cs] in 3 M HNO$_3$ as the aqueous phase. After extraction, ~24 mM Cs was present in the organic phase, then 0.01 M HNO$_3$ was added to the organic phase, and the mixture was shaken. After shaking and removing the aqueous phase, fresh 0.01 M HNO$_3$ was introduced and shaken again. This procedure was repeated five times and the Cs concentration in the aqueous phase was then determined by ICP-MS. The results are shown in Figure 13 with the number of back-extractions shown on the x-axis and back-extraction efficiency shown on the y-axis. In the current study, over 60% of Cs was stripped by one back-extraction and approximately 98% was stripped after five back-extractions. This result indicates the ease of stripping Cs from an organic phase compared with the solid-liquid separation method. From this study, it is evident that HNO$_3$ (0.01 M) could be used as the back-extraction condition.

4. Conclusion

In conclusion, extraction of Cs by DtBuDB18C6 was studied using alcohols, ketones, ethers, and esters as diluents of crown ethers because of their environment friendly C, H, and O compositions. The solubility of DtBuDB18C6 in ketones, ethers, and esters was higher compared to alcohols and hydrocarbons. The $D$ values in ethers and esters were small, however, those in ketones and alcohols were observed as ≥
0.94. Using available solvents, i.e., 1-octanol and 2-nonanone, $D$(Cs) increased with HNO$_3$ concentration, which indicated an ion-pair extraction. Slope analysis gave a 1 : 1 complex of Na and K and a 1 : 2 complex of Rb and Cs with DtBuDB18C6. A $D$(Cs) of almost ten or higher was obtained when 0.2 M or higher concentrations of DtBuDB18C6 in 2-nonanone and 3 M HNO$_3$ were used in the experimental condition. The $D$(Cs) into the organic solvent with high di-electric constant seems to be high, due to the ion-pair extraction. High concentrations of Cs (up to 24 mM) were extracted in 2-nonanone and diluted HNO$_3$ (0.01M) was used as the stripping solution; approximately 98% by stripping five times could be removed from the organic phase.

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References

1) B. C. Russell, P. E. Warwick, I. W. Croudace, Anal. Chem., 86, 11890-11896 (2014).
2) D. R Raut, P. K. Mohapatra, M. K. Choudhary, S. K. Nayak, J. Membr. Sci., 429, 197-205 (2013).
3) Y. Sasaki, K. Morita, S. Suzuki, H. Shiwaku, K. Ito, Y. Takahashi, M. Kaneko, Solvent Extr. Res. Dev., Jpn., 24, 113-122 (2017).
4) R. S. Herbst, J. D. Law, T. A. Todd, V. N. Romanovskiy, I. V. Smirnov, V. A. Babain, V. N. Esmiantovskiy, B. N. Zaitsev, Sep. Sci. Technol., 38, 2685-2708 (2003).
5) Y. Morita, I. Yamaguchi, T. Fujiwara, K. Mizoguchi, M. Kubota, JAERI-Res., 2000-024 (2000).
6) E. H. Borai, R. Harjula, L. Malinen, A. Paajanen, J. Hazard. Mater., 172, 416-422 (2009).
7) Y. Wu, X. X. Zhang, S. Y. Kim, Y. Z. Wei, J. Nucl. Sci. Technol., 53, 1968-1977 (2016).
8) L. T. X. Thuy, M. Yasuzawa, T. Yabutani, Bull. Chem. Soc. Jpn., 86, 958-962 (2013).
9) S. Ayrault, B. Jimenez, E. Garnier, M. Fedoroff, D. J. Jones, C. Loos-Neskovic, J. Solid State Chem., 141, 475-485 (1998).
10) K. Kawatake, N. Shigemoto, J. Nucl. Sci. Technol., 49, 1048-1056 (2012).
11) Y. Kudo, S. Katsuta, Y. Takeda, Anal. Sci., 25, 563-566 (2009).
12) T. G. Levitskaia, L. Maya, G. J. Van Berkel, B. A. Moyer, Inorg. Chem., 46, 261-272 (2007).
13) Y. Kikuchi, Y. Sakamoto, Anal. Chim. Acta, 403, 325-332 (2000).
14) K. K. Gupta, P. V. Achuthan, A. Ramanujam, J. N. Mathur, Solvent Extr. Ion Exch., 21, 53-71 (2003).
15) D. Saha, J. Vithya, R. Kumar, C. R. V. Subramani, P. R. V. Rao, Radiochim. Acta, 104, 195-204 (2016).
16) Y. Kikuchi, Y. Sakamoto, Anal. Chim. Acta, 370, 173-179 (1998).
17) Y. Takeda, Bunseki Kagaku, 51, 515-525 (2002).
18) A. Kumar, P. K. Mohapatra, V. K. Manchanda, J. Radioanal. Nucl. Chem., 229, 169-172 (1998).