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DEVELOPMENT OF A NOVEL SOLVENT FOR THE SIMULTANEOUS SEPARATION OF STRONTIUM AND CESIUM FROM DISSOLVED SPENT NUCLEAR FUEL SOLUTIONS

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ABSTRACT

The recovery of Cs and Sr from acidic solutions by solvent extraction has been investigated. The goal of this project was to develop an extraction process to remove Cs and Sr from high-level waste in an effort to reduce the heat loading in storage. Solvents for the extraction of Cs and Sr separately have been used on both caustic and acidic spent nuclear fuel waste in the past. The objective of this research was to find a suitable solvent for the extraction of both Cs and Sr simultaneously from acidic nitrate media.

The solvents selected for this research possess good stability and extraction behavior when mixed together. The extraction experiments were performed with 4′,4,(5′)-Di-(t-butyldicyclohexano)-18-crown-6 {DtBuCH18C6}, Calix[4]arene-bis-(tert-octylbenzo-crown-6) {BOBCalixC6} and 1-(2,2,3,3-tetrafluoropropoxy)-3-(4-*sec*-butylphenoxy)-2-propanol {Cs-7SB modifier} in a branched aliphatic kerosene {Isopar$^®$ L}. The BOBCalixC6 and Cs-7SB modifier were developed at Oak Ridge National Laboratory (ORNL) by Bonnesen et al. [1].

The values obtained from the SREX solvent for D_{Sr} in 1 $\underline{M}$ nitric acid ranged from 0.7 to 2.2 at 25°C and 10°C respectively. The values for D_{Cs} in 1 $\underline{M}$ nitric acid with the CSSX solvent ranged from 8.0 to 46.0 at 25°C and 10°C respectively. A new mixed solvent, developed at the Idaho National Engineering and Environmental Laboratory (INEEL) by Riddle et al. [2], showed distributions for Sr ranging from 8.8 to 17.4 in 1 $\underline{M}$ nitric acid at 25°C and 10°C respectively.   The D_{Cs} for the mixed solvent ranged from 7.7 to 20.2 in 1 $\underline{M}$ nitric acid at 25°C to 10°C respectively. The unexpectedly high distributions for Sr at both 25°C and 10°C show a synergy in the mixed solvent. The D_{Cs}, although lower than with CSSX solvent, still showed good extraction behavior.

INTRODUCTION

A significant amount of the radioactivity in high-level liquid waste from spent nuclear fuel recycling comes from the fission products ^{137}Cs and [90]Sr. Together ^{137}Cs and ^{90}Sr account for over 99% of the relative radioactivity of dissolved nuclear fuel solutions once the actinides have been removed [3]. Removal of ^{137}Cs with a halflife ($t_{1/2}$) of 30 years, and ^{90}Sr with a $t_{1/2}$ = 29 years would increase the safety of handling of the majority of the liquid waste and reduce the storage and disposal costs of these radioactive components. The ability to remove and recover Cs and Sr from spent nuclear fuel waste is also an important issue regarding short-term heat loading in a geological repository [4]. Solvents

for the extraction of cesium and strontium have been developed separately for both alkaline and acidic liquid radioactive waste reprocessing at the INEEL and at other facilities [4,5,6]. Removal of ^{137}Cs ($t_{1/2}$ = 30 y) and ^{90}Sr ($t_{1/2}$ = 29 y) from spent nuclear fuel would allow these isotopes to be stored in a short-term waste facility. The removal of cesium and strontium, and some actinides, would allow the long-term storage facility to store waste closer together by eliminating these major sources of the heat load. The goal is to improve the long-term storage facility's storage capacity and to create less need for new facilities. To do this, the extraction technology must be highly selective and cost effective.

In previous work, cesium and strontium have been removed from nuclear acidic waste solutions and related alkaline wastes using separate solvents designed specifically for these elements [1,4,5,6]. In general, crown ethers have been selected for the removal of strontium [6], and calixarenes [1], have been selected for separations of cesium. This study investigates the combination of these two types of extractants to create a solvent that will extract both cesium and strontium from acidic media, simultaneously. The solvent components were selected for this research based on previous work, which showed these compounds to have favorable selectivity for cesium and strontium [5,7,8]. These include a common extractant used in the selective separation of strontium from acidic solutions which is a mixture of DtBuCH18C6, TBP and Isopar® L (Strontium Extractant, SREX process). For the extraction of cesium, a solvent containing BOBCalixC6, Cs-7SB modifier, TOA (trioctylamine) and Isopar® L (Cesium Extractant, CSSX process) was selected. The BOBCalixC6 and the Cs-7SB modifier were both developed at Oak Ridge National Laboratory (ORNL) by Bonnesen et al. [1]. Further work with BOBCalixC6 to develop a Cs extraction process (CSSX) was reported by Leonard et al. [5]. These compounds possess good phase stability when mixed together and exhibit excellent simultaneous cesium-strontium extraction behavior.

EXPERIMENTAL

All diluents were reagent grade and were used as received. Deionized water was used to prepare all aqueous acid solutions. The nitric acid was reagent grade from Aldrich Chemical Co. (Milwaukee, WI). Isopar® L isoparaffinic diluent was obtained from Exxon Chemical Company (Houston, TX). The ^{85}Sr and ^{137}Cs radiotracers used for spiking the simulants were obtained as ^{85}SrCl$_2$ in 1 $\underline{M}$ HCl and ^{137}CsCl in 1 $\underline{M}$ HCl from Isotope Products (Burbank, CA). The DtBuCH18C6 crown ether was purchased from Eichrom Industries, Inc. (Darien, IL). The BOBCalixC6 calixarene crown ether and Cs-7SB modifier, obtained from Oak Ridge National Laboratory, were originally received from IBC Advanced Technologies, Inc. (American Fork, UT) and Boulder Scientific Company (Mead, CO), respectively, and were used as received [9,10]. Figure 1 shows the structures of the individual components.

BOBCalixC6
Calix[4]arene-bis-(tert-octylbenzo-crown-6)

DtBuCH18C6
4′,4′,(5′)-Di-(t-butyldicyclo-hexano)-18-crown-6

Cs-7SB modifier
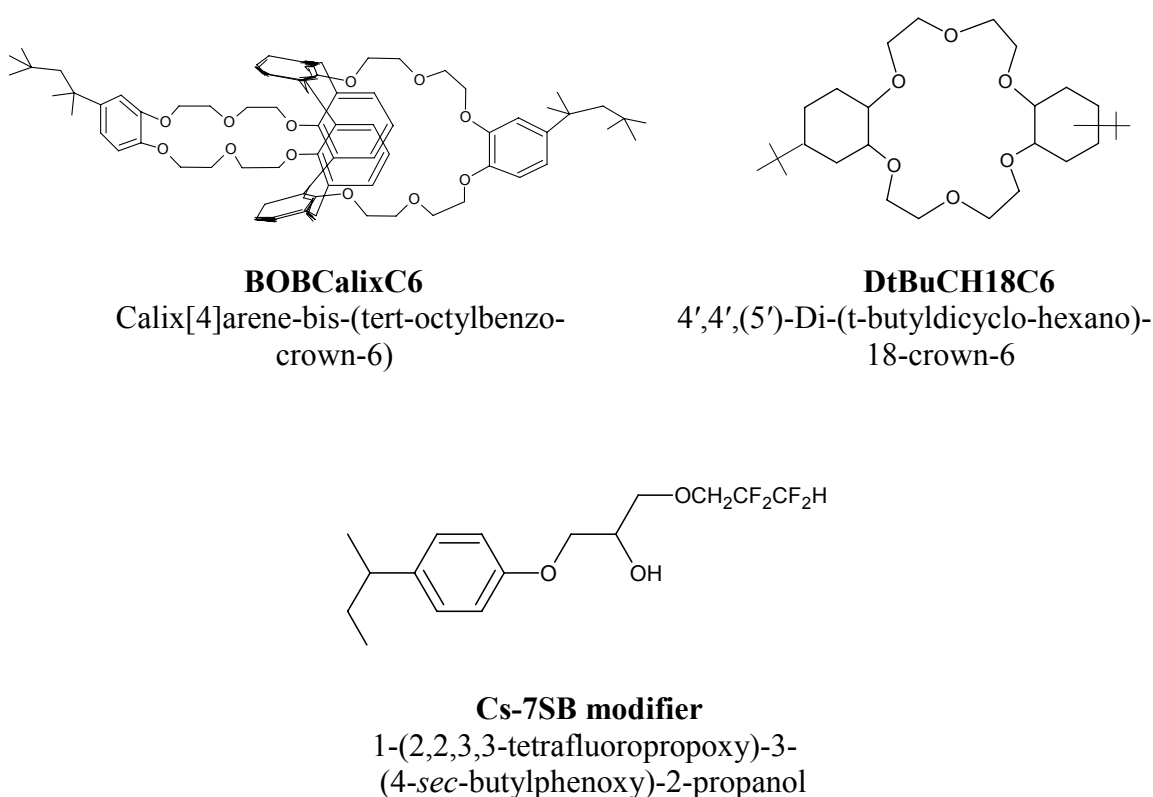1-(2,2,3,3-tetrafluoropropoxy)-3-(4-*sec*-butylphenoxy)-2-propanol

Figure 1. Molecular structures of the cesium and strontium extractants and the Cs-7SB modifier.

Initial experiments investigated the individual extractants with regard to their Cs and Sr extraction efficiency. The SREX solvent was prepared using a mixture of 0.15 $\underline{M}$ DtBuCH18C6, 1.2 $\underline{M}$ TBP and Isopar® L as reported in Horwitz et al [6]. The mixture was stirred until the components went into solution after approximately 1 hour. The ^{137}Cs extraction solvent, CSSX, was received already prepared and consisted of 0.007 $\underline{M}$ BOBCalixC6, 0.75 $\underline{M}$ Cs-7SB modifier, 0.003 $\underline{M}$ trioctylamine (TOA) and Isopar® L [1]. Radiotracers, used in all test samples, were diluted and carriers were added that consisted of 0.001 $\underline{M}$ $Sr(NO_3)_2$ for Sr and 0.0001 $\underline{M}$ $CsNO_3$ for Cs, which was consistent with previous work [10,11]. The mixture was heated to incipient dryness and concentrated HNO_3 was added to finish the conversion of the tracers to the nitrate salts. After three such cycles, 10 mL of varying concentrations from 0.01 $\underline{M}$ to 10 $\underline{M}$ of HNO_3 were added to the tracers and carriers to re-dissolve the salts in preparation for the extraction studies. The activity of the radiotracer was monitored to qualitatively follow the process of extracted phases.

Nitric acid dependency tests used $^{85}Sr_{w/c}$ (with carrier) prepared in varying concentrations of HNO_3 extracted with SREX solvent in equal proportions. All test samples in this research were shaken by hand for 1 minute, centrifuged for 1 minute, the organic and aqueous fractions were separated and the two phases analyzed. The organic and aqueous aliquots were γ-ray counted using an HPGe detector. The 661 keV line of

[137]Cs and the 514 keV line of [85]Sr were used to measure these elements. Nitric acid dependency tests were run for [137]Cs$_{w/c}$ as above and extracted with CSSX solvent.

Mixed solvents were prepared by adding neat DtBuCH18C6 to produce concentrations from 0.045 $\underline{M}$ to 0.358 $\underline{M}$ in 0.007 $\underline{M}$ BOBCalixC6, 0.75 $\underline{M}$ Cs-7SB modifier, TOA and Isopar L. The Cs/Sr extraction solvent used in the rest of this study was a mixture of 0.15$\underline{M}$ DtBuCH18C6, 0.007 $\underline{M}$ BOBCalixC6 and 0.75 $\underline{M}$ Cs-7SB modifier in Isopar L. The extractant was prepared by adding neat DtBuCH18C6, BOBCalixC6, and Cs-7SB modifier to a mixing vessel. Approximately 10% of the required final Isopar L volume is added to lower the viscosity and the mixture was left to stir overnight. The remainder of the Isopar L was then added the next morning. Ambient temperature throughout the experiments was 23°C ± 3°C unless otherwise specified.

Acid dependency tests used [85]Sr$_{w/c}$ (with carrier) prepared in varying concentrations of HNO$_3$ extracted with SREX solvent at equal volume ratios. Acid dependency tests were also run for [137]Cs$_{w/c}$ in the same varying concentrations of HNO$_3$ as above and extracted with CSSX solvent. Nitrate dependency tests were done using varied amounts of Al(NO$_3$)$_3$ in 0.5 $\underline{M}$ HNO$_3$ to yield NO$_3^-$ concentrations of 0.60, 0.70, 0.90, 1.0, 2.5, and 5.0 $\underline{M}$ total NO$_3^-$ [7]. To each of these concentrations of NO$_3^-$, ^{137}Cs and [85]Sr tracers were added. All organic solvents were pre-equilibrated with the appropriate aqueous phase prior to extraction. In the extraction step, organic solvent and radiotraced acidic solution were combined. The radiotracer/acid solution comprised the aqueous bottom layer of the two phase system while the organic solvents comprised the top organic phase. For the constant temperature portion of the experiment, a temperature controlled water bath was used and the samples were left to equilibrate in the bath for 15 minutes. All test samples in this research were shaken for 1 minute, centrifuged for 1 minute, the organic and aqueous fractions were separated by pipeting out equal volumes, 0.5 mL, of each phase and the two phases analyzed.

RESULTS AND DISCUSSION

Nitric acid dependency experiments showed that acidities > 3 $\underline{M}$ resulted in third phase formation. Therefore, all extractions discussed here were performed at < 3 $\underline{M}$ HNO$_3$. It was also found that in the SREX solvent, D$_{Cs}$=2.9E^{-3} (1 $\underline{M}$ HNO$_3$) in the CSSX solvent D$_{Sr}$=nd (1 $\underline{M}$ HNO$_3$, nd=none detected). The first nitric acid dependency tests, Figure 2, showed the SREX solvent to give a Sr distribution ratio of 0.70 and the CSSX solvent to give a Cs distribution ratio of 7.8 in 1 $\underline{M}$ HNO$_3$ at ambient temperature. The resulting slopes for plots of Log D versus [HNO$_3$] were 1.40 ±0.13 for Sr and 0.75 ±0.17 for Cs. The mean of these data (N= 3) has an experimental uncertainty of ± 5% and is consistent with previous work [11,12].
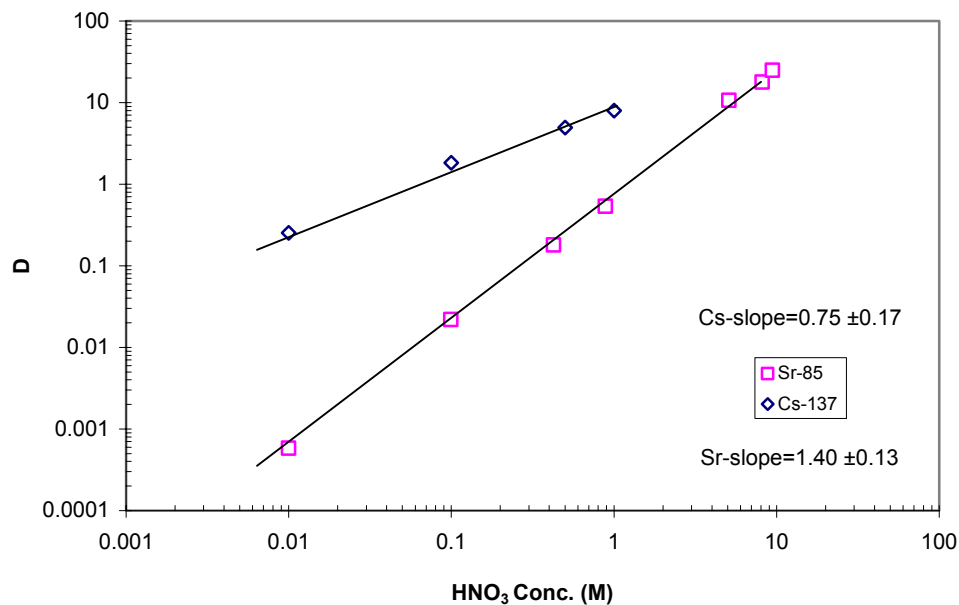
Figure 2. D_{Sr} versus nitric acid concentration for SREX and D_{Cs} versus nitric acid concentration for CSSX. The associated slope for Sr = 1.40 ± 0.13 and the slope for Cs = 0.75 ±0.17.

A nitric acid dependency test was performed on the new mixed extractant in the same manner as for the SREX and CSSX extractants shown in Figure 2. The results for the new mixed solvent, Figure 3, showed an increase in the Sr distribution compared to SREX and a Cs distribution ratio which remained approximately the same as with the CSSX solvent in 1 $\underline{M}$ HNO_3. In Figure 3, the Sr slope=1.4 ±0.1 and the Cs slope=0.7 ±0.03 for the new mixed solvent were approximately the same as previous tests using SREX and CSSX for the separation of Sr and Cs, respectively, suggesting that the reaction stoicheometry has not changed in the mixture.
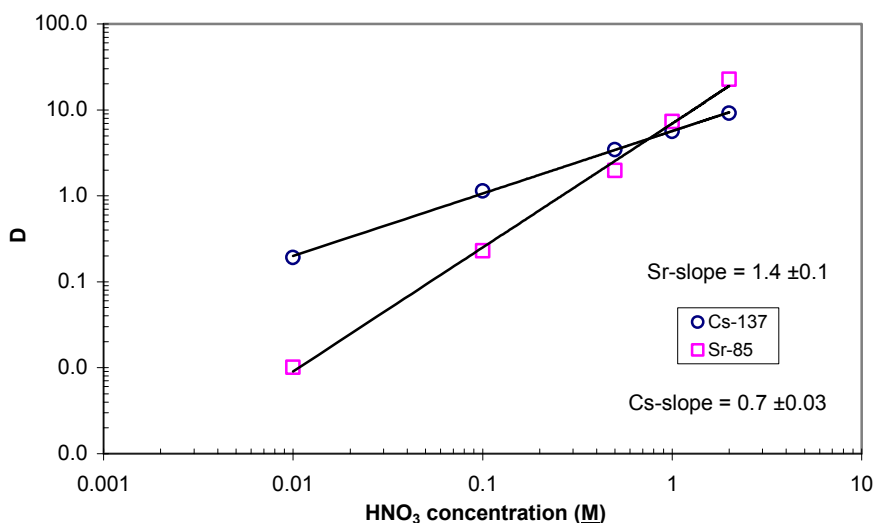
Figure 3. D_{Sr} and D_{Cs} versus nitric acid concentration for new mixed solvent. The associated slopes of the plots of Sr =1.4 ±0.1 and Cs =0.70 ±0.03.

Nitrate dependency tests were also performed using $Al(NO_3)_3$, with constant HNO_3 in an effort to elucidate the effect of nitrate ion on the forward distribution of strontium, in the absence of acidity. Figure 4 shows a Log/Log relationship between D and $[NO_3^-]$ with a slope for strontium of 2.2 ±0.3 and a slope for cesium of 1.0 ±0.4. The slopes are similar to previous work [7,13], and indicate that 2 NO_3^- anions contribute in the extraction of Sr as shown in equation 1 and one NO_3^- anion contributes in the extraction of Cs as shown in equation 2 [14]. The organic phase at >2.5 $\underline{M}$ NO_3^- concentrations showed possible extractant decomposition.
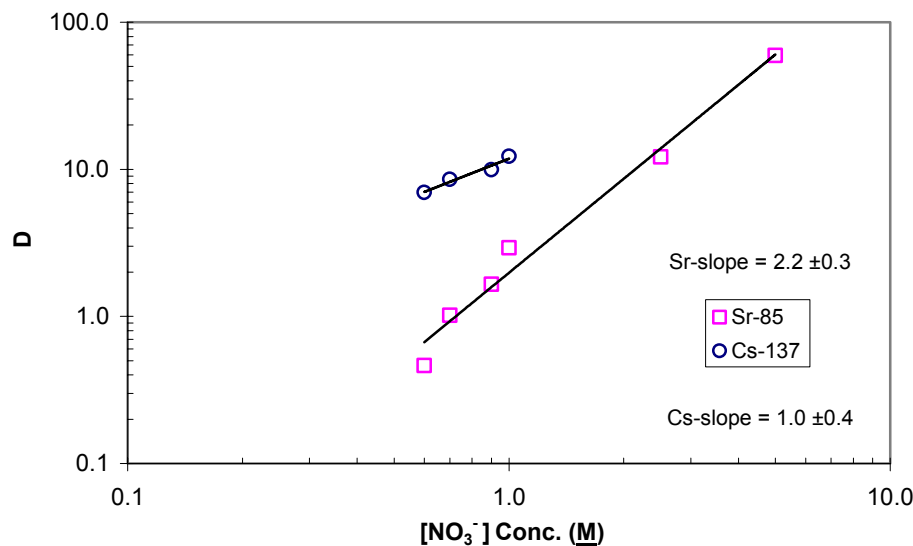
$$Sr^{2+} + 2NO_3^- + DtBuCH18C6 \rightleftharpoons DtBuCH18C6 \cdot Sr(NO_3)_2 \qquad \text{Eq. 1}$$

$$Cs^+ + NO_3^- + BOBCalixC6 \rightleftharpoons BOBCalixC6 \cdot Cs(NO_3) \qquad \text{Eq. 2}$$

Figure 4. D_{Sr} and D_{Cs} in 1 $\underline{M}$ HNO$_3$ versus NO$_3^-$ concentrations from SREX solvent for Sr and CSSX solvent for Cs. The associated slope for the plot of Sr = 2.2 ±0.3 and for the plot of Cs = 1.0 ±0.4.

Simply combining the solvents, however, was not effective for Cs and Sr co-extraction. When the two solvents were mixed in a 1:1 volume ratio, the distribution of Sr remained approximately the same, D_{Sr}=0.9, as it was with SREX, D_{Sr}=0.7, while the distribution of Cs decreased to D_{Cs}=0.2. However, when only neat DtBuCH18C6 was added in varying concentrations to the CSSX solvent an unexpectedly high forward distribution for Sr was achieved. In fact, the distribution of Sr increased to D_{Sr} = 7.8 while the distribution for Cs, D_{Cs} = 5.7, remained relatively high in comparison with the distributions for CSSX solvent alone.

When the DtBuCH18C6 concentration was increased in the mixture there was a corresponding increase in the distribution ratio of Sr. The mixture used for further experiments was 0.007 $\underline{M}$ BOBCalixC6, 0.15 $\underline{M}$ DtBuCH18C6, and 0.75 $\underline{M}$ Cs-7SB modifier in Isopar[®] L. The mixture was selected based on the plot of the Cs and Sr distribution coefficients as a function of the DtBuCH18C6 concentration in 0.007 $\underline{M}$ BOBCalixC6, 0.75 $\underline{M}$ Cs-7SB modifier and TOA in Isopar[®] L (CSSX solvent). Conditions were selected which gave good performance for Sr, while maintaining
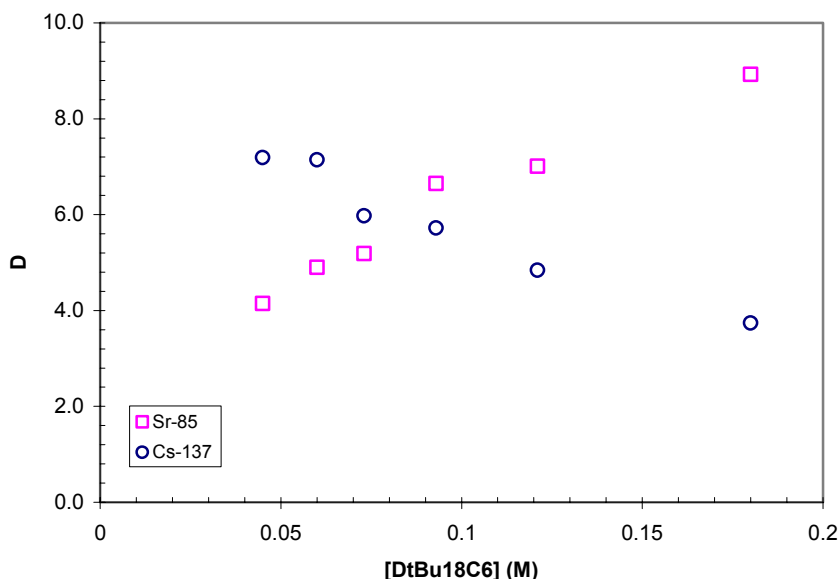
adequate extraction of Cs as shown in Figure 5.

Figure 5. D_{Cs} and D_{Sr} in 1 $\underline{M}$ HNO$_3$ versus DtBuCH18C6 concentration in 0.007 $\underline{M}$ BOBCalixC6, 0.75 $\underline{M}$ Cs-7SB modifier, and 0.003 $\underline{M}$ TOA in Isopar® L (CSSX solvent).

In order to determine which component of the mixed solvent was causing this synergy for Sr extraction, tests were conducted on various combinations of DtBuCH18C6, BOBCalixC6, Cs-7SB modifier, TBP, and TOA. Tests run on the SREX solvent with the addition of 0.75 $\underline{M}$ Cs-7SB modifier gave a D_{Sr} = 3.4 in 1 $\underline{M}$ HNO$_3$. This is a considerable drop in distribution from using neat DtBuCH18C6 mixed with Cs-7SB modifier alone which gives a D_{Sr} = 6.6. Therefore, TBP was not used in the new mixed solvent since it shows that it hinders forward extraction of Sr. In limited testing, TOA showed no effect on the distribution of Sr when neat DtBuCH18C6 was added to CSSX as well as when TOA was added to the SREX solvent. When a combination of 0.003 $\underline{M}$ TOA and 0.15 $\underline{M}$ DtBuCH18C6 in Isopar® L was tested the distribution for Sr showed almost no extraction with a D_{Sr} = 9.9E^{-3} in 1 $\underline{M}$ HNO$_3$. This test eliminates TOA as the possible synergistic component. Therefore, TOA was not investigated in experiments with the new mixed Cs/Sr extractant. In an effort to find out if the BOBCalixC6 was the synergistic component for the increased Sr distributions, a test was run using only DtBuCH18C6 and Cs-7SB modifier in Isopar® L. The resultant strontium distribution was 6.4, and the cesium distribution was 0.05 indicating no cesium extraction by the crown ether, but enhanced D_{Sr}.

Cs-7SB modifier alone in Isopar® L exhibited no extraction of Cs and Sr from 1 $\underline{M}$ nitric acid solutions. However, when its concentration was varied in the mixed solvent the results shown in Figure 6 were found. They show the Cs-7SB modifier to be the probable synergistic component in producing the elevated Sr distributions.
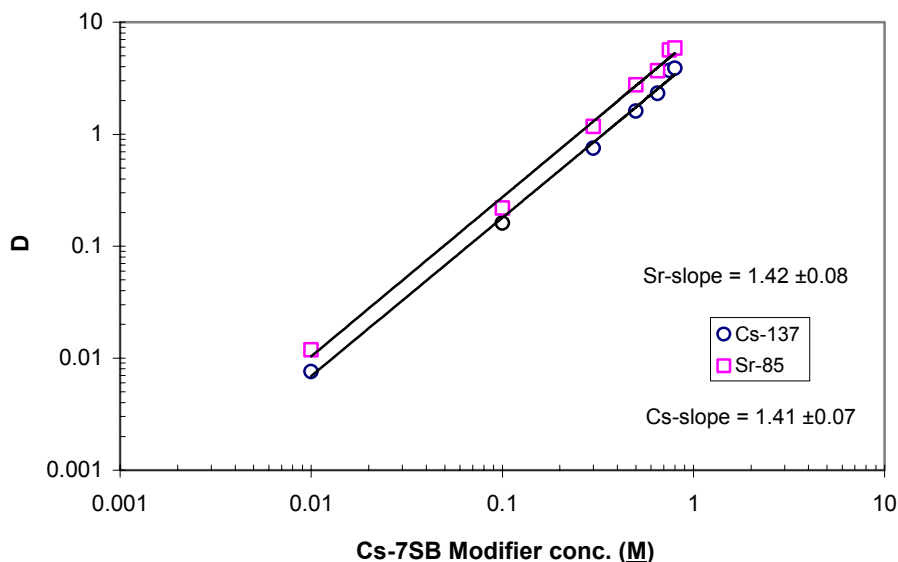
Figure 6. D_{Sr} and D_{Cs} in 1M HNO$_3$ versus Cs-7SB concentration in 0.15 M DtBuCH18C6, 0.007 M BOBCalixC6 and Isopar L. The associated slopes for the plots of Cs =1.41 ±0.07 and Sr =1.42 ±0.08.

Due to the increased viscosity associated with the modifier, concentrations >0.8 M were not studied. Addition of Cs-7SB to the SREX extractant significantly improves the Sr removal from acidic solutions. The positive effect of fluorinated modifiers is well documented for the extraction of cesium using crown ethers or calixarenes, [15] but these modifiers have not been previously reported as being applied to the SREX process for enhancing strontium extraction.

From previous literature it was noted that calixarene extractions are temperature dependent, giving higher distributions for cesium at lower temperatures [1,10,15]. This increase in distribution with decreasing temperature is of interest for the Sr extraction as well as the Cs. Temperature dependence tests on the SREX, CSSX and the new mixed solvent were conducted over a range of temperatures. Tests from 1 M HNO$_3$ at 10°C yielded D_{Sr} = 2.2 for the SREX solvent, D_{Cs} = 46.0 for the CSSX solvent, and D_{Sr} = 17.4, D_{Cs}= 20.2 for the new mixed solvent in separate studies. Temperature dependence testing on the new mixed solvent over a range of temperatures yielded increased distributions as shown in Figure 7.
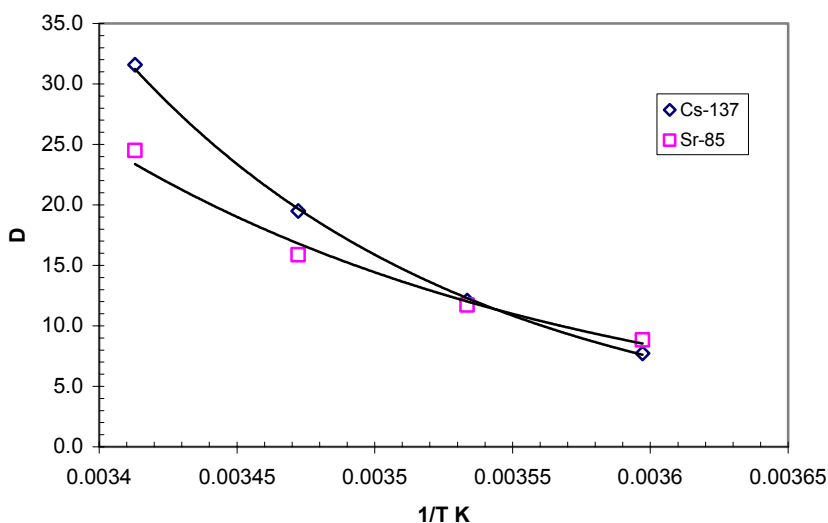
Figure 7. D_{Cs} and D_{Sr} in $1\underline{M}$ HNO_3 from the new mixed solvent at various temperatures.

To support development of a complete separation process, the ability to repetitively extract and back extract Cs and Sr to recover and reuse the new mixed solvent were studied. Repetitive forward extractions of Cs and Sr using fresh contacts of the mixed solvent gave the results shown in Table 1.

Contact #	D_{Cs}	D_{Sr}
1	7.2	7.3
2	6.7	7.4
3	5.5	6.6
4	2.6	7.0

Table 1. Repetitive extractions of Cs and Sr from mixed solvent.

These results indicate that Sr continues to be extracted with high distribution ratios during multiple contacts and the Cs distributions gradually decrease with the fourth contact dropping off by approximately a factor of 3 from the first contact. Currently it is not fully understood why D_{Cs} decreases in the fourth contact and further testing in this area is being done.

Efficient stripping of most of the Cs/Sr from the loaded new mixed solvent was demonstrated in four stages by repeatedly contacting equal volumes of loaded solvent with 0.01 $\underline{M}$ HNO_3. The results of four back extractions (strips) are shown in Table 2.

	D_{Cs}	D_{Sr}
Strip #1	0.45	0.04
Strip #2	0.24	0.01
Strip #3	0.21	nd
Strip #4	0.42	nd

Table 2. Back extraction of Cs and Sr from mixed solvent. nd = none detected.

By the third strip, the level of Sr was below detection limits. The D_{Cs} was consistent with previous literature in which TOA was not present [1,10,15]. The number of strips necessary to quantitatively extract the Cs out of the new mixed solvent may be decreased by the addition of a trialkylamine, such as TOA, which was found to assist in stripping [1,10,15].

In additional support of a viable separation process, the new mixed extractant was tested on a simulated dissolved spent nuclear fuel solution. The composition of the simulated dissolved spent nuclear fuel solution is shown in Table 1.

Table 1

HNO_3	3.0E-01	Ce	4.3E-03
Sr	2.0E-03	Nd	7.2E-03
Cs	4.1E-03	Sm	5.0E-03
Rb	1.0E-03	Gd	2.6E-04
Zr	1.1E-02	Eu	1.4E-04
Ba	3.7E-03	Y	1.4E-03
La	2.2E-03		

The nitric acid concentration was increased above that in the original simulant to 1$\underline{M}$ HNO_3 for comparison with the previous experimental data using acidic solutions alone. The temperature dependence testing showed distributions for Cs and Sr at 25°C of 3.0 and 4.5, respectively and at 10°C the distributions for Cs and Sr were 6.1 and 10.8, respectively as shown in Figure 8.
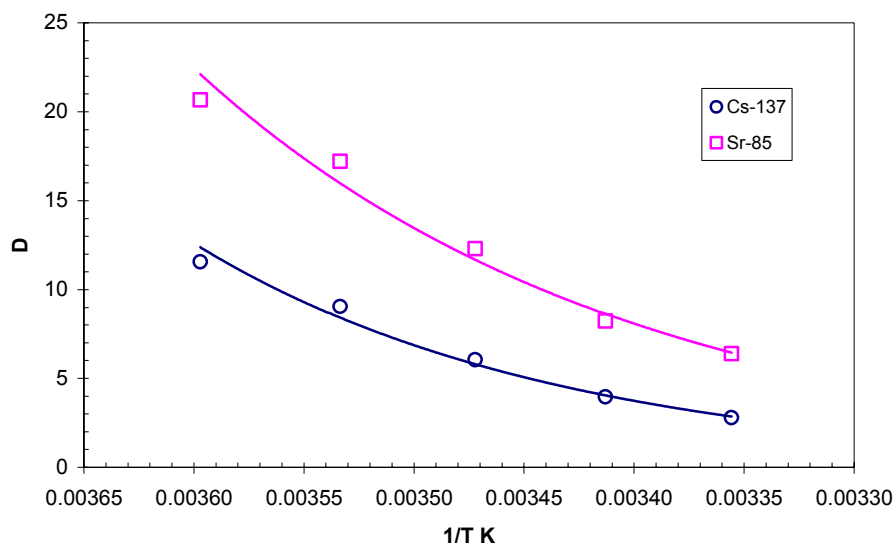
Figure 8. D_{Sr} and D_{Cs} in 1 $\underline{M}$ HNO$_3$/Dissolved Spent Nuclear Fuel Simulant versus the New Mixed Solvent

CONCLUSIONS

The ability to extract ^{137}Cs and ^{90}Sr from acidic nitrate solutions with the new mixed solvent demonstrates a potential for the removal and recycling of these radioisotopes from spent nuclear fuel solutions. The combination of DtBuCH18C6, BOBCalixC6, and Cs-7SB modifier in Isopar L has shown to be an effective mixed solvent for the extraction of Cs and Sr from acidic nitrate media. The synergistic properties for the extraction of Sr with this new mixed solvent show that not only can it be used for simultaneous extractions but also as a stand alone solvent for Sr removal. Distributions for Sr and Cs using the new mixed solvent (Figure 8) were 8.8 and 7.7 respectively, rather than the expected D_{Sr} and D_{Cs} value of 0.7 and 6.0 respectively, at 25°C in 1 $\underline{M}$ HNO$_3$. The combination of DtBuCH18C6, BOBCalixC6 and Cs-7SB modifier in Isopar$^{®}$ L shows synergistic extraction of Sr while maintaining high Cs extraction. This synergy is due to the Cs-7SB modifier.

The results of temperature dependence studies on the new mixed solvent show D_{Cs} and D_{Sr} increase with decreasing temperature, giving distributions for Sr between 7 and 28 at 20°C and 5°C, respectively. The results for Cs were also very good with D_{Cs} values of between 8 and 33 at 20°C and 5°C, respectively. The results of this study also demonstrate the new mixed solvent's superior extraction ability over SREX solvent with a D_{Sr} = 0.7 for SREX and a D_{Sr} = 7.7 for the new mixed solvent.

The results of repetitive extractions show the new mixed solvent to continue to remove Sr with high distribution ratios through the fourth contact, while the distribution of Cs decreased in the fourth contact. This decrease is currently being studied and further

testing will need to be done. The new mixed solvent demonstrated excellent back extraction for Sr with levels below detection limits by the third strip phase. The back extraction of Cs showed an increase in the distribution in the fourth strip which can not currently be accounted for and further research in this area needs to be done. The distribution ratios for Cs and Sr in a dissolved spent nuclear fuel simulant using the new mixed solvent were also very good with distributions of D_{Cs}=6.1 and a D_{Sr}=10.8 at 10^oC

Overall, the new mixed solvent shows promise as a simultaneous Cs and Sr extractant and this research is on going in an effort to optimize this single solvent extraction process.

ACKNOWLEDGMENTS

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