REMOVING Cs FROM NUCLEAR WASTE LIQUID BY CROWN ETHER AND HETEROPOLY ACID: SIMULATED TESTS

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(Received October 4, 1995)

The extraction behavior of DB21C7-HPW synergistic system for Cs has been investigated and compared with that of DB21C7. Two simulated radioactive waste have been examined. It shows that this synergistic system is not only effective for ordinary waste SAW1, but also for the waste of high concentration of nitrates (SAW3). An extraction percentage 99.1% of Cs of SAW3 was obtained by an extracting stage of 5 and scrubbing stage of 3. The stability of synergistic agents towards radiolysis is satisfied and the selectivity for Cs is quite good. Both the additive compound and the extracted Cs complex of DB21C7 and HPW have been separated and identified.

Nuclear waste solution produced by reprocessing of irradiated fuel elements, possesses high level-activity. If it is directly solidified by cementation or glasslike ceramics, the waste volume is going to be greatly raised, and then the following handling and final disposal of these solidified blocks will meet much trouble.

Seeking for the economic and effective ways to remove $^{137}$Cs from medium or high active waste solutions, becomes continuously the aim of international scientists.

Schulz$^1$ gave the technological review and evaluation to the methods of removing by-product $^{137}$Cs from nitric solution. Up to now, there exists two kinds of programs been regarded hopeful. The one is crown ether system and the other is dicarbolide system.

$$H^+[[\pi-(3)-1,2-B_9C_2H_{11}Cl_2]_{2}Co]^-$$

Kyrs$^2$ used dicarbolide in nitrobenzene as extractant. The later could preferentially extract Cs from 0.5M HNO$_3$. The shortage of this method lies in the potential corrosive danger caused by the radiolysis product Cl.

Davis$^3$ suggested a 4 component system of TBP-NNS-crown ether-kerosens. The chosen crown ether was bis(1-hydroxy-2-ethylhexyl) benzo-18-crown-6. Synthesizing of the appointed crown ether needs more efforts and hence the extractant price will greatly increase.

Blasius$^4$ put forward the method to extract Cs synergistically with DB21C7 and heteropoly acid. This paper reports some new results of removing Cs with the crown ether system on the basis of Blasius' paper.
Results and discussion

1. The extraction of DB21C7 for Cs

According to the requirement of a good fit of ionic diameter and the cavity size of crown ether, 21C7 and its derivatives will be the optimum candidates for the extraction of Cs. This prospect has been confirmed by several papers\(^4\). The coordination of 21C7 and DB21C7 with Cs exhibits high stability constants.

Owing to its slight solubility in water, DB21C7 was chosen as the neutral component of extractants. Considering the extractable compound being an ion-pair complex, we chose the polar agent nitrobenzene as the solvent for promoting the solubility of extractable complex in organic phase.

The effect of the concentration of HNO\(_3\) on the extraction of Cs is shown in Tab 1. Distribution behavior of Cs was investigated by using \(^{134}\)Cs as radiotracer. Distribution ratios of Cs were determined by measuring the γ-activity concentration of both the organic and aqueous phase.

Increase of the concentration of HNO\(_3\) enhanced the extraction of Cs. Here exists both the common ionic effect and the salt-out effect caused by the hydrous species formation.

<table>
<thead>
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<th>CHNO(_3), M</th>
<th>0.5</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
</tr>
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<tbody>
<tr>
<td>D</td>
<td>1.00</td>
<td>2.14</td>
<td>4.40</td>
<td>5.86</td>
<td>6.01</td>
</tr>
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</table>

Table 1
Effect of the concentration of HNO\(_3\)

extractant: 0.01M DB21C7 in nitrobenzene
aqueous phase: 0.5-4M HNO\(_3\), 40ppm Cs

Waste solution produced by post-processing of irradiated nuclear fuel contains high concentration of NaNO\(_3\). The later could be extracted in a small extent by DB21C7. This paper measured the effect of the concentration of NaNO\(_3\) in a range of 0.5-3.5M on the extraction of Cs (see Table 2).

<table>
<thead>
<tr>
<th>C(_{\text{NaNO}_3}), M</th>
<th>0.5</th>
<th>1</th>
<th>1.5</th>
<th>2</th>
<th>3</th>
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<tbody>
<tr>
<td>D</td>
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<td>0.93</td>
<td>0.89</td>
<td>0.82</td>
<td>0.76</td>
<td>0.52</td>
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</table>

Table 2
Effect of the concentration of NaNO\(_3\)

extractant: 0.012M DB21C7 in nitrobenzene
aqueous phase: 0.5M HNO\(_3\), various conc. of NaNO\(_3\), 40 ppm Cs

The results showed that the distribution ratio of Cs had not remarkable varience while the concentration of NaNO\(_3\) varied less than 1.5M. From NaNO\(_3\) concentration of 2M on, the distribution ratio decreased gradually. It reflects the effect of competition extraction of Na in high concentration to Cs in a concentration of rather low level.