Yttrium-90 separation in carbonate media by solvent extraction

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**Introduction**

Yttrium-90

$T_{1/2} = 64$ hours, $E_\beta = 2.28$ MeV

Daughter product of Strontium-90

Has radiotherapy application because of its practically pure $\beta^-$ particle emission.

Useful for Strontium-90 monitoring in the environment

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**Strontium-90**

$T_{1/2} = 28.8$ years, $E_\beta = 0.546$ MeV

One of the fission products of $^{235}\text{U}$

A part of radioactive waste and nuclear fallout from nuclear tests

Exhibits biochemical behavior similar to calcium and accumulates in bones

Used as a marker radionuclide for atropogenic radioactive contamination levels determination

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$^{90}\text{Y}/^{90}\text{Sr}$ separation is still a challenging task for radiochemists in both environmental and clinical application.
Introduction

Usual practice: $^{90}\text{Y}$ recovery from acidic media with crown ethers and D2EHPA (di-2-ethyl-hexyl-phosphoric acid) as extractants.

Our proposal: $^{90}\text{Y}/^{90}\text{Sr}$ separation and recovery from alkaline carbonate media

advantages – greener, faster, cheaper

Potential yttrium extractants

Diluents
Toluene
2-nitro-toluene
Butyl acetate

2,3-DHN
(2,3-dihydroxynaphthalene)

PAN
(pyridylazonaphthol)

8-HQ
(8-hydroxyquinoline)

PAR
(pyridylazoresorcinol)

chromotropic acid
Experimental

Step 1

Y₂O₃ solubility and dissolution kinetics study

Na₂CO₃, K₂CO₃ and (NH₄)₂CO₃ were used to study Y₂O₃ solubility at room temperature.

To study yttrium dissolution rate we prepared a disk with a constant surface area:

\[ 0.03 \text{ g Y}_2\text{O}_3 \rightarrow 6 \text{ mm diameter and 0.4 mm thickness disk} \]

with the surface \( S \), described as

\[ S = 2 \pi r^2 + 2 \pi rh = 2 \pi r (r + h) \]

The batch experiments were conducted with 20 mL of each alkaline agent in a 25 mL bottle.

<table>
<thead>
<tr>
<th>Alkaline agent</th>
<th>Solubility g/L</th>
<th>Dissolution rate V g·m²/s</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 M NaHCO₃</td>
<td>0.5 ± 0.035</td>
<td>0.071 ± 0.023</td>
</tr>
<tr>
<td>1 M Na₂CO₃</td>
<td>1.7 ± 0.017</td>
<td>0.213 ± 0.025</td>
</tr>
<tr>
<td>1 M K₂CO₃</td>
<td>0.9 ± 0.031</td>
<td>0.262 ± 0.021</td>
</tr>
<tr>
<td>1 M (NH₄)₂CO₃</td>
<td>2.8 ± 0.182</td>
<td>0.243 ± 0.026</td>
</tr>
<tr>
<td>2 M Na₂CO₃</td>
<td>4.9 ± 0.096</td>
<td>0.430 ± 0.013</td>
</tr>
<tr>
<td>2 M K₂CO₃</td>
<td>1.4 ± 0.029</td>
<td>0.458 ± 0.010</td>
</tr>
<tr>
<td>2 M (NH₄)₂CO₃</td>
<td>8.2 ± 1.01</td>
<td>0.622 ± 0.018</td>
</tr>
</tbody>
</table>

Based on the high Y₂O₃ solubility and dissolution rate, we studied yttrium extraction from Na₂CO₃, K₂CO₃ and (NH₄)₂CO₃ solutions.
Experimental

Step 2

Yttrium extraction and Y/Sr separation in alkaline carbonate media

Despite $(\text{NH}_4)_2\text{CO}_3$ has the highest solubility and dissolution rate, it possess a poor yttrium extraction and low distribution ratio (D) (with third phase formation).

$\text{K}_2\text{CO}_3$ demonstrates the highest yttrium distribution ratio (D) with 8-HQ, followed by $\text{Na}_2\text{CO}_3 \rightarrow \text{K}_2\text{CO}_3$ was chosen for further extractions and separations studies.

Only 8-HQ and 2,3-DHN demonstrated sufficient values of yttrium distribution ratios.

For extraction experiments the following system was chosen:

“0.01 M of L (8-HQ/2,3-DHN) in org. diluent (BuAc/2-nitrotoluene) - 0.001 M [Y] in 0.5 M $\text{K}_2\text{CO}_3$ aqueous solution”.

For $^{90}\text{Y}/^{90}\text{Sr}$ separation the following system was chosen:

“0.001 M of L (8-HQ/2,3-DHN) in org. diluent (BuAc/2-nitrotoluene) – 4*10^{-5} M [Y] and 1*10^{-6} M [Sr] in 0.5 M $\text{K}_2\text{CO}_3$ aqueous solution”.

Y and Sr concentrations are measured spectrophotometrically and by ICP.

$^{90}\text{Y}$ and $^{90}\text{Sr}$ distribution ratios were measured radiometrically.
Experimental
Yttrium extraction (8-HQ)

The effect of initial pH of the aqueous solution

Extraction with 8-HQ demonstrates that the value of yttrium distribution ratio $D$ depends on initial pH of the aqueous phase, gradually increasing from pH = 10 to pH = 13.5.

Organic phase saturation shows the limit ratio $[L]:[M] = 4.1$, which one can interpret as solvate composition ML4.
Experimental
Yttrium/Strontium separation with 8-HQ

Extraction and separation experiments, carried out for stable Y and Sr demonstrated the principal possibility of Y/Sr pair separation by solvent extraction. Separation factor SF for extraction systems «0.001 M of 8-HQ in org. diluent – 4*10^{-5} M [Y] and 1*10^{-6} M [Sr] in 0.5 M K_{2}CO_{3} aqueous solution» increases with pH and reaches its maximum values at pH = 13.5 both for BuAc and 2-nitrotoluene. Y is extracted much better (D_{max} = 3.89) in 2-nitrotoluene with practically the same Sr extraction for two organic diluents. Maximum separation is observed in 2-nitrotoluene: SF = 195.
Extraction and separation experiments, carried out for radioactive $^{90}$Y and $^{90}$Sr confirmed the principal possibility of Y/Sr pair separation by solvent extraction with some features for trace amounts of radioactive isotopes.

The trend of SF dependence on pH is the same: it reaches maximum values at pH = 13.5 both for BuAc and 2-nitrotoluene.

In contrast with the stable Y and Sr, $^{90}$Y is extracted much better ($D_{\text{max}} > 65$) in BuAc. Maximum separation is observed in BuAc: SF $\sim$ 200.
Experimental
Yttrium/Strontium separation

<table>
<thead>
<tr>
<th>Diluent</th>
<th>8-HQ pH = 13.5</th>
<th>2,3-DHN pH = 13.0</th>
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</thead>
<tbody>
<tr>
<td></td>
<td>D&lt;sub&gt;Y&lt;/sub&gt;</td>
<td>D&lt;sub&gt;Sr&lt;/sub&gt;</td>
</tr>
<tr>
<td>BuAc</td>
<td>0.177</td>
<td>0.177</td>
</tr>
<tr>
<td></td>
<td>65.7*</td>
<td>0.33*</td>
</tr>
<tr>
<td>2-nitrotoluene</td>
<td>3.89</td>
<td>0.02</td>
</tr>
<tr>
<td></td>
<td>9.59*</td>
<td>0.1*</td>
</tr>
</tbody>
</table>

* - measured radiometrically

The similarity of extraction and separation data, obtained for stable and radioactive isotopes with 8-HQ, is evident for 2-nitrotoluene. Taking into account the different experimental methods, the values are very close. This confirms the principal possibility of Y/Sr separation in carbonate media. The opposite situation for BuAc requires further investigation.

Conclusion

Y/Sr extraction and separation in carbonate media with several ligands showed, that only 8-HQ and 2,3-DHN can be regarded as perspective compounds for further studies.

The data obtained for stable and radioactive isotopes separation with 8-HQ revealed differences in the behavior of the extracted elements, which can be explained by differences in their concentrations (trace amounts for ⁹⁰Y and ⁹⁰Sr with the background of stable Y).

2,3-DHN demonstrated high solubility in aqueous phase with low values of distribution ratios, but it is still regarded as perspective after addition of modifier, preventing its transfer into aqueous phase.

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