A Method for the Replacement of $^{137}\text{Cs}$ with $^{40}\text{K}$ for Open-Source Ion Exchange Research Applications in Hot Cells.

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Presentation Outline

• Background,
• Connection to Hot Cells and Radiation Protection,
• Potassium and Ion Exchange,
• Method Development of $^{40}$K as an Analogue of $^{137}$Cs,
• Hot Cell Context & Conclusions.
Background

$^{137}\text{Cs}$ is one of the principal contaminants in many nuclear facilities being decommissioned, e.g. FGMSP, concrete lined Hot Cells.

Weathering and erosion of external facilities causes the leaching of radioactive material into the concrete structure. **External surface at 9 Sv/h!**

*First generation Magnox storage pond, (FGMSP)*

The remediation is to take place through the development of a “**nuclear elastoplast**” using ion exchange materials and electrokinetic methods.
Hot Cells and Worker Protection

Caesium cations:
- Two radioactive Cs isotopes,
- Significant safety management,
- Highly soluble,
- Migrates readily with water,
- Difficult to immobilise.

Post Irradiation Examination facility at PIED, Bhaba Atomic Research Centre, India.

Insoluble salts and carbonates on external pond-concrete surfaces require analytical study.
Potassium Calibration

Our current laboratory facilities are not sufficient to be able to handle open-source radioactive caesium. So we have used potassium.

Established a linear relationship between the concentration of KCl in solution and the associated count detected.

Proposal Issues - Isotope Selection

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Half-Life</th>
<th>Specific Activity (Ci/l)</th>
<th>Decay Mode</th>
<th>Decay Energy (MeV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cs-137</td>
<td>30 Yrs</td>
<td>88</td>
<td>Beta</td>
<td>0.19</td>
</tr>
<tr>
<td>Ba-137m</td>
<td>133 Seconds</td>
<td>540 Million</td>
<td>Gamma</td>
<td>0.662</td>
</tr>
</tbody>
</table>

Not practical or prudent to use an open Cs-137 aqueous source to develop the proposed technology described by the project description.

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<tbody>
<tr>
<td>K-40</td>
<td>1.3 Billion Yrs</td>
<td>0.0000071</td>
<td>Beta, Gamma</td>
<td>0.52, 0.146</td>
</tr>
</tbody>
</table>

Potassium 40 displays very similar physical and chemical properties to that of Cs 137.

[2] Argonne National Laboratory, Human Health Fact Sheets, August 2005

Our current laboratory facilities are not sufficient to be able to handle open-source radioactive caesium. So we have used potassium.
Detection Apparatus

Radiometric measurement provides a versatile and consistent means of quantifying concentrations and masses of radionuclides, both \textit{in situ} and in the laboratory.
Ion Exchange Approach

\[ M^-A^+ + B^+ \leftrightarrow M^-B^+ + A^+ \]  \[1\]

Where M is the ion exchange material carrying the A\(^+\) ions and B\(^+\) are the counter ions in an aqueous phase. It should be noted that this reaction can be reversible.

Potassium Adsorption Studies

Determining the plausibility of measuring an ion exchange reaction radiometrically using $^{40}$K.

**Batch Experiments**

- 5.62g of KCl
- 20mL of water
- Lewatit resin added [2]
- Stirred for 2 hours
- 7mL aliquots taken
- Detected for 20 hours
- 0g – 20g resin used

The linear relationship between number of counts, $N$, and mass of IX is given by $N = A - B[IX]$. $A = 6103 \pm 151$, $B = -199 \pm 12$

$I^-H^+ + K^+ \leftrightarrow M^-K^+ + H^+$

Potassium Adsorption Isotherms

Linear Isotherms

(a) \[ \frac{1}{q_E} = \left( \frac{1}{bQ^0} \right) \frac{1}{C_A} + \frac{1}{Q^0} \]

Langmuir

(b) \[ \log(q_E) = \log(K_f) + \frac{1}{n} \log(C_A) \]

Freundlich

\( K_f, \ 1/n = \) Freundlich Constants

\( b, \ Q^0 = \) Langmuir Constants

\( q_E = \) Grams of cation adsorbed per gram of resin

\( C_A = \) Adsorbed Concentration
## Potassium Adsorption Isotherms

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<th>Langmuir Model</th>
<th>Resin</th>
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<tr>
<td>$K_f$</td>
<td>$1/n$</td>
<td>$Q^0$</td>
</tr>
<tr>
<td>$\chi^2$</td>
<td>$b$</td>
<td>$\chi^2$</td>
</tr>
<tr>
<td>Empirical</td>
<td>0.286</td>
<td>0.109</td>
</tr>
<tr>
<td>0.371</td>
<td>120.039</td>
<td>1.525</td>
</tr>
<tr>
<td>0.491</td>
<td>0.1</td>
<td>0.1</td>
</tr>
</tbody>
</table>

Non-Linear Freundlich isotherm plotted against experimental data.

**Freundlich**

$$q_E = K_f C_A^n$$

**Langmuir**

$$q_E = (C_0 - C_E) \frac{V}{W}$$
Caesium Adsorption

- Approximately 6 grams of washed and dried K⁺ form resin;
- 20 ml DW;
- Varying masses of non-active CsCl added;
- Mixed for two hours;
- Aliquot of solution taken and detected;
- New Cs⁺ form resin is washed, dried, and detected.

Counts detected: IXCs⁺ + Solution = Total Post Cs ≈ IXK⁺

M⁻K⁺ + Cs⁺ ↔ M⁻Cs⁺ + K⁺
Caesium Adsorption Isotherms

Using the detected count from the displaced potassium Freundlich and Langmuir relationships were derived describing the caesium adsorption.

\[ q_E = \text{grams of Cs adsorbed per gram of resin} \quad \text{and} \quad C_E = \text{Equilibrium Concentration} \]
Caesium Adsorption Isotherms

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<td>$\chi^2$</td>
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<tr>
<td>Empirical</td>
<td>0.472</td>
<td>0.442</td>
</tr>
<tr>
<td></td>
<td>0.228</td>
<td>16.879</td>
</tr>
<tr>
<td></td>
<td>0.04</td>
<td>3.872</td>
</tr>
<tr>
<td></td>
<td>0.35</td>
<td>Total Cs$^+$ Capacity</td>
</tr>
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Non-Linear Freundlich

$q_E = K_f C_A^{\frac{1}{n}}$

The experimental data from the caesium adsorption study is consistent with the previous potassium adsorption study and the resin manufacturers data.
The use of a K$^+$ form ion exchange medium can be used in migration studies of non-active Caesium, as a marker.

- No radiation concerns,
- No special equipment required,
- Allows safe method development,
- Uses same technique as *in situ* radiation detection,
- Simulate real-world concentrations.
Context of Results II
Conclusions

• Demonstrated the novel use of $^{40}\text{K}$ as a radiometric source,
• This is shown in the development of an empirical adsorption isotherm from radiometric data,
• We can monitor non-active $\text{Cs}^+$ adsorption through the radiometric measurement of displaced $\text{K}^+$ in ion exchange media,
• This allows work to be carried out without the radiological concerns of more dangerous isotopes and the use of Hot Cells, thereby allowing potassium to be used as a non-active analogue of $^{137}\text{Cs}$.
Acknowledgements

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Thank you for your attention, any questions?