Combined technology for radioactive contaminated soil remediation based on application of hydroseparation, chemical leaching and addition of natural organic and mineral absorbers

ISTC Project # B-859

Valentina Toropova and Dmitri Davydov

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Technology for radioactive contaminated soil remediation based on application of hydroseparation, chemical leaching and addition of natural organic and mineral absorbers.
Goal of the research

Examine soil samples contaminated with $^{137}\text{Cs}$ and $^{90}\text{Sr}$ due to Chernobyl NPP accident:

– soils of agricultural regions,

– soils of the exclusion zone of Chernobyl NPP.

Great variety of soils characteristics:

– acidity,

– content of organic carbon and mineral part,

– type of humus,

– absorption capacity,

– sum of absorbed bases and $K^+$, $Ca^{2+}$, $Mg^{2+}$ etc.
Obtained results

Table 1 - $^{137}$Cs distribution throughout the uppermost layer (1 cm) fractions of the sandy and sandy-loam soils

<table>
<thead>
<tr>
<th>Size, mm</th>
<th>Relative mass, %</th>
<th>Specific activity, Bq/g</th>
<th>Relative activity, %</th>
</tr>
</thead>
<tbody>
<tr>
<td>&gt; 0.05</td>
<td>80.10</td>
<td>1.30</td>
<td>33.46</td>
</tr>
<tr>
<td>0.05 - 0.005</td>
<td>20.05</td>
<td>33.0</td>
<td>58.83</td>
</tr>
<tr>
<td>&lt; 0.005</td>
<td>0.9</td>
<td>18.80</td>
<td>6.9</td>
</tr>
</tbody>
</table>

Table 2 - $^{137}$Cs distribution throughout the deeper soil layers of the sandy and sandy-loam soils

<table>
<thead>
<tr>
<th>Depth, cm</th>
<th>Relative mass, %</th>
<th>Relative activity, %</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>&gt; 0.05 mm</td>
<td>0.05 - 0.005 mm</td>
</tr>
<tr>
<td>0-3</td>
<td>85.3</td>
<td>10.5</td>
</tr>
<tr>
<td>2-8</td>
<td>85.0</td>
<td>6.7</td>
</tr>
<tr>
<td></td>
<td>&gt; 0.05 mm</td>
<td>0.05 - 0.005 mm</td>
</tr>
<tr>
<td></td>
<td>27.4</td>
<td>66.6</td>
</tr>
<tr>
<td></td>
<td>22.5</td>
<td>39.3</td>
</tr>
<tr>
<td></td>
<td>&lt; 0.005 mm</td>
<td></td>
</tr>
<tr>
<td></td>
<td>9.5</td>
<td>36.4</td>
</tr>
</tbody>
</table>
### Table 3 - $^{137}$Cs content in silt fraction depending on soil type

<table>
<thead>
<tr>
<th>Soil</th>
<th>$^{137}$Cs content in silt fraction, %</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sand &amp; sod-podsol sand</td>
<td>25-50</td>
</tr>
<tr>
<td>Peat-clay highly-humus</td>
<td>&gt; 80</td>
</tr>
<tr>
<td>Turf-humus</td>
<td>&gt; 70</td>
</tr>
<tr>
<td>Peat-marsh</td>
<td>~ 80</td>
</tr>
<tr>
<td>Peat</td>
<td>&gt; 60</td>
</tr>
</tbody>
</table>
Conclusion

• Hydroseparation allows to extract soil fraction < 0.05 mm – fine fraction

• Fine soil fraction contains:
  - organic substances (~ 5 – 15 % of total soil mass)
  - radionuclides (~ 50 – 80 % of total radioactivity)

• Decontamination factor, $K_d \approx 3$

• For different soil types these values may vary significantly

• Hydroseparation can be effectively applied for soils with high concentration of fine soil fraction

• Preliminary ultra-sound treatment of soil suspension in water could be used to disaggregate soil particles.
2 – Chemical leaching

Kinetics of $^{137}$Cs desorption from silt soil fraction in the presence of K$^+$ and Cs$^+$

- **T = 20 °C**
  1. $[\text{K}^+, \text{Cs}^+_{\text{stable}}] = 10^{-5}$ mole/l
  2. $[\text{Cs}^+_{\text{stable}}] = 1 \cdot 10^{-3}$ mole/l

- **T = 60 °C**
  3. $[\text{K}^+] = 1 \cdot 10^{-5}$ mole/l
  4. $[\text{Cs}^+_{\text{stable}}] = 1 \cdot 10^{-5}$ mole/l
Desorption of $^{137}$Cs from sandy loam soil in the presence of Fe(III) hydroxo complexes

pH 3.5

1 – [Fe(III)] = $1 \cdot 10^{-4}$ mole/l

2 – [Fe(III)] = $5 \cdot 10^{-3}$ mole/l

Desorption, %

Desorption time, day
Desorption of $^{137}$Cs and $^{90}$Sr from sod-podsol and peat soils

Desorption solution - 1 mole/l KCl
1 – content of organic substance 1 - 5 %
2 – content of organic substance 10 - 20 %
Desorption of $^{137}$Cs from the mineral type soils by solutions containing inorganic cations

Solution composition:

- 0,01 mole/l FeCl$_3$
- 0,1 mole/L KNO$_3$, NaNO$_3$, CsNO$_3$, NH$_4$NO$_3$
The effect of multicharged cations on radionuclides leaching from soil

\[ \text{Me}^{z^+} + \text{H}_2\text{O} \rightarrow \text{MeOH}^{(z-1)^+} + \text{H}^+ \]
Conclusion

• Kinetics of $^{137}$Cs radionuclide extraction from soils and silt soil fraction is very slow. Notable result achieved after 7 days.

• Organic substance and clay content effects on efficiency of the radionuclide desorption. Maximum extraction of $^{137}$Cs can be reached for sandy soil ($\approx 40\%$).
Conclusion

• The presence of Fe(III) hydroxo complexes increases $^{137}$Cs extraction

• Degree of $^{137}$Cs extraction changes in the following sequence $\text{Fe}^{3+} > \text{Cs}^+ > \text{NH}_4^+ \approx \text{K}^+ \approx \text{Na}^+$ from 40 to 2 % respectively

• Higher temperatures increase $^{137}$Cs extraction by $\sim 50$ %

• Combination of hydroseparation and chemical leaching can give maximum decontamination factor $K_d \approx 4 – 5$
3 – Combined technology for radioactive contaminated soil remediation

Hydrosedation

High activity soil

Soil fractions requiring additional treatment

Low activity soil

Chemical leaching

Dewatering

Clean soil

Waste water treatment

Clean water

Solid phase

Conditioning Disposal

Disposal/Reuse

Clean water
1 – Investigation of radionuclides distribution in soil fractions
   • high activity soil
   • low activity soil – clean and requiring additional treatment

2 – Optimization of hydroseparation technologies:
   • membrane processes
   • filtration
   • centrifugation and gravitation

3 – Comparison of hydroseparation and dry fractioning methods

4 – Preliminary ultra-sound treatment of soil suspension in water could be used to disaggregate soil particles.
Chemical leaching

1 – Isotope exchange

$^{137}$Cs sorption in the presence of Cs$_{stable}$ is very low (< 20 %) → desorption into such solution should be high (> 80 %). In practice the desorption value was < 27 %.

Explanation – kinetics problems, e.g. interlayer localization of cesium.

One of the possible solution of this problem can be incorporation of hydroxo complexes into the interlayer space.
Chemical leaching

2 – Isomorphous exchange
• most simple and cheap reagents selection
• effect of temperature and pH

3 – Oxidation process
• chemical oxidation
• high temperature oxidation (to be determined)
Waste water treatment

1 – Phase separation technologies
   • Coagulation
   • Floculation
   • Flotation
   • Sedimentation
   • Mechanical filtration

2 – Sorption processes
   • Selective sorbents
   • Carbon
   • Ion-exchange

3 – Membrane technologies
   • Microfiltration
   • Ultrafiltration
   • Nanofiltration

4 – Red/Ox processes
   • Ultraviolet
   • Ozone
   • Electrooxidation
Dewatering

- Microfiltration, ultrafiltration
- Pressfiltration
- Centrifugation
Proposed steps for Fukushima

- Radionuclide distribution studies
- Comparison of hydroseparation and dry fractioning methods
- Isotope exchange
  - Isomorphous exchange
  - Oxidation process
- Chemical leaching studies
- Membrane processes
- Centrifugation
  - Gravitation
  - Filtration
  - Membrane processes
- Optimization of fractioning technologies
- Waste water treatment technologies
- Microfiltration
  - Ultrafiltration
  - Pressfiltration
  - Centrifugation
- Wet fraction treatment (dewatering)
- Phase separation
  - Absorbers
  - Membranes
  - Coagulation
  - Red/Ox processes
- Creation of pilot installation
Thank you for your attention!