Determination of pure beta emitters using LSC for characterization of waste from nuclear decommissioning

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• Closed nuclear facilities

- 100 mines;
- 90 commercial power reactor;
- 250 research reactors
- a number of fuel cycle facilities

A few of them were decommissioned, and most of them is on the way to be decommissioned.
Steps of decommissioning of nuclear facility

- **Preparation** (investigating the background radioactivity around NPP)
- **Plant Cleanout**
  - Removal of most radioactive component such as spend fuel elements, reactor internals, reactor vessel, etc. which is transferred for storage and disposal. (high level waste). (Evaluation of radioactivity before transferring)
- **Decontamination**
  - Removal of contamination from surfaces of facilities or equipment by chemical or mechanical methods, which can reduce the waste volume and active level in the waste. (Measurement of radioactivity to evaluation the decontamination, and estimation of radioactivity in the waste)
- **Dismantling**
  - Equipments within the facility are dismantled and classified by estimation of the radioactivity)
- **Demolition and site clearance**
  - Buildings demolished and radioactive wastes removed to storage or disposal facilities after estimation of the radioactivity in the waste.
- **Release of the site to alternative use** (measure the radioactivity level in the released area)
Main radionuclides in the nuclear reactor

• Long-lived fission products
  $^{137,135}$Cs, $^{106}$Ru, $^{90}$Sr, $^{99}$Tc, $^{129}$I, etc.

• Neutron activation products
  $^{58,60}$Co, $^{133}$Ba, $^{134}$Cs, $^{152,154,155}$Eu, $^3$H,
  $^{14}$C, $^{36}$Cl, $^{41}$Ca, $^{63,59}$Ni, $^{94}$Nb, $^{55,59}$Fe,
  $^{93}$Zr, $^{93}$Mo, $^{54}$Mn, $^{110m}$Ag, etc.
  $^{238-241}$Pu, $^{241}$Am, $^{243,244}$Cm, $^{237}$Np
Main Radionuclides in the nuclear waste and used materials in the view of measurement

- **Gamma radionuclides**
  - $^{60}$Co, $^{133}$Ba, $^{137}$Cs, $^{134}$Cs, $^{106}$Ru, $^{152,154,155}$Eu, $^{58}$Co, $^{54}$Mn, $^{59}$Fe, $^{110m}$Ag, $^{94}$Nb.

- **Beta Emitter**
  - $^{3}$H, $^{14}$C, $^{36}$Cl, $^{41}$Ca, $^{55}$Fe, $^{63}$, $^{59}$Ni, $^{93}$Zr, $^{93}$Mo, $^{90}$Sr, $^{99}$Tc, $^{129}$I.

- **Alpha emitter** (actinides)
  - $^{238-241}$Pu, $^{241}$Am, $^{243,244}$Cm, $^{237}$Np
Waste samples and the relevant critical radionuclides for decommissioning

- Graphite (reactor)
  - $^3\text{H}$, $^{14}\text{C}$, $^{55}\text{Fe}$, $^{63}$, $^{59}\text{Ni}$, $^{60}\text{Co}$, $^{152}\text{Eu}$

- Concrete (normal or heavy)
  - $^{41}\text{Ca}$, $^{60}\text{Co}$, $^{55}\text{Fe}$, $^{63}$, $^{59}\text{Ni}$, $^{133}\text{Ba}$, $^{152}\text{Eu}$

- Steel/stainless steel
  - $^{55}\text{Fe}$, $^{63}$, $^{59}\text{Ni}$, $^{36}\text{Cl}$, $^{93}\text{Zr}$, $^{93}\text{Mo}$, $^{94}\text{Nb}$, $^{60}\text{Co}$, $^{152}\text{Eu}$, transuranics

- Aluminium
  - $^{60}\text{Co}$, $^{36}\text{Cl}$, $^{63}\text{Ni}$, $^{55}\text{Fe}$

- Lead
  - $^{60}\text{Co}$, $^{63}\text{Ni}$, $^{55}\text{Fe}$

- Water
  - $^3\text{H}$, $^{14}\text{C}$, $^{63}\text{Ni}$, $^{99}\text{Tc}$, $^{36}\text{Cl}$, $^{129}\text{I}$, $^{90}\text{Sr}$, $^{60}\text{Co}$, $^{137}\text{Cs}$, transuranics

- Ion exchange resin
  - $^{55}\text{Fe}$, $^{63}$, $^{59}\text{Ni}$, $^{14}\text{C}$, $^{99}\text{Tc}$, $^{36}\text{Cl}$, $^{93}\text{Zr}$, $^{93}\text{Mo}$, $^{94}\text{Nb}$ $^{90}\text{Sr}$, $^{129}\text{I}$, $^{137}\text{Cs}$, $^{60}\text{Co}$, $^{135}\text{Cs}$, transuranics
Determination using chemical separation and LSC

- $^{55}\text{Fe}$
- $^{63}\text{Ni}$
- $^{41}\text{Ca}$, $^{90}\text{Sr}$
- $^{36}\text{Cl}$, $^{129}\text{I}$
Production of $^{63}\text{Ni}$ and $^{55}\text{Fe}$ in Nuclear Reactor (neutron activation)

- **$^{63}\text{Ni}$:**
  - $^{62}\text{Ni}(n, \gamma)^{63}\text{Ni}$ ($\sigma=14.5$ b; $\eta_{62\text{Ni}}=3.63\%$)
  - $^{63}\text{Cu}(n, p)^{63}\text{Ni}$, ($\eta_{63\text{Cu}}=69.17\%$)

- **$^{55}\text{Fe}$:**
  - $^{54}\text{Fe}(n, \gamma)^{55}\text{Fe}$ ($\sigma=2.3$ b; $\eta_{54\text{Fe}}=5.85\%$)
  - $^{56}\text{Fe}(n, 2n)^{55}\text{Fe}$, ($\eta_{56\text{Fe}}=91.75\%$)
Decay of $^{63}\text{Ni}$ and $^{55}\text{Fe}$

$^{55}\text{Fe}$ (2.73 y)
- EC (232 keV, 100%)
- No gamma ray

$^{55}\text{Mn}$ (stable)

$^{63}\text{Ni}$ (100.1 y)
- $\beta^-$ (66.95 keV, 100%)
- No gamma ray

$^{63}\text{Cu}$ (stable)

Fe-55 decays by electron capture emitting X-rays, conversion electrons and Auger electrons (5 - 6 keV)

X ray (5.89 keV, 25.4%)

No gamma ray
Analytical method for $^{63}\text{Ni}$ and $^{55}\text{Fe}$

Measurement methods

$^{55}\text{Fe}$: X-ray spectrometry ( <1%);
LSC (30-45%)

$^{63}\text{Ni}$: gass flow counting (anti-coincidence, <10-50%)
Ion implanted silicon detector (1-6%)
LSC (60-80%)

• Analytical procedure:
  • Decomposition of sample
  • Separation of Ni or Fe from matrix elements and all other radionuclides
  • Preparation of a suitable solution for LSC measurement.
Separation of Ni and Fe by anion exchange chromatography

<table>
<thead>
<tr>
<th>Element</th>
<th>Content, %</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Ni fraction</td>
</tr>
<tr>
<td>Fe$^{3+}$</td>
<td>&lt;0.001</td>
</tr>
<tr>
<td>Ni$^{2+}$</td>
<td>&gt;99.5</td>
</tr>
<tr>
<td>Co$^{2+}$</td>
<td>&lt;0.01</td>
</tr>
<tr>
<td>Ba$^{2+}$</td>
<td>&lt;7.5</td>
</tr>
<tr>
<td>Eu$^{3+}$</td>
<td>&gt;99.8</td>
</tr>
<tr>
<td>Cs$^+$</td>
<td>&gt;99.5</td>
</tr>
<tr>
<td>Sr$^{2+}$</td>
<td>&gt;99.5</td>
</tr>
</tbody>
</table>

- Ni can be completely separated from Fe, Co, Cu, Zn, U, Pu, etc.
- Fe can be separated from Ni, Cr, Mn, Th, etc.
Separation of Ni using Ni-DMG complex

• Ni can form a stable specific complex with dimethylglyoxime. By Ni-DMG precipitation or organic solvent extraction of Ni-DMG complex at low concentration, Ni can be separated from many other elements.

• While, some other metals, such as Co, Cu can also form a complex with DMG and interfering the separation of Ni.
Purification of Ni using Ni-Resin

The Nickel Resin contains the DMG inside the pores of a polymethacrylate resin. The nickel-DMG precipitate occurs on the resin, where it is held and readily separated from other elements in the supernatant.

1. Loading of solution
2. Washing with 0.2 M ammonium citrate to remove other elements
3. Eluting Ni using HNO3
4. Evaporte eluted Ni-DMG solution to 0.1-0.2 ml for LSC
Purification of Ni by specific Ni-extraction chromatography

<table>
<thead>
<tr>
<th>Element</th>
<th>Recovery or decontamination factor</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ni$^{2+}$</td>
<td>&gt; 98.5%</td>
</tr>
<tr>
<td>Fe$^{3+}$</td>
<td>$10^4$</td>
</tr>
<tr>
<td>Co$^{2+}$</td>
<td>$10^3$</td>
</tr>
<tr>
<td>Ba$^{2+}$</td>
<td>$10^4$</td>
</tr>
<tr>
<td>Eu$^{3+}$</td>
<td>$10^4$</td>
</tr>
<tr>
<td>Cs$^+$</td>
<td>$10^4$</td>
</tr>
<tr>
<td>Sr$^{2+}$</td>
<td>$10^4$</td>
</tr>
</tbody>
</table>

Ni specific extraction chromatography has a higher decontamination to most of elements, such as Fe, Co, Cu, Cr, Mn, Ba, Eu, transuranics, etc.

• A higher recovery of Ni can be obtained in the procedure.

Preparation of separated $^{55}$Fe for LSC

Content of Fe (mg)

Efficiency, %

FeCl₃ solution
Fe₃⁺ + H₃PO₃(2M)
FeSO₄
Quench correction for Fe-55
Quench correction for Ni-63

Graphite sample with 2 mg Ni carrier

Efficiency, %

Quench curve of Ni-63

$y = 9 \times 10^{-7}x^3 - 0.0015x^2 + 0.6991x + 95.98$
Recovery of Fe and Ni and decontamination factors for main interfering radionuclides and elements

<table>
<thead>
<tr>
<th>Interference</th>
<th>Recovery/decontamination factor</th>
<th>Interference</th>
<th>Recovery/decontamination factor</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Fe fraction</td>
<td>Ni fraction</td>
<td>Fe fraction</td>
</tr>
<tr>
<td>$^{55}$Fe</td>
<td>85-95%</td>
<td>$&gt;10^5$</td>
<td>$^{133}$Ba</td>
</tr>
<tr>
<td>$^{63}$Ni</td>
<td>$&gt;10^5$</td>
<td>80-95%</td>
<td>$^{134,137}$Cs</td>
</tr>
<tr>
<td>$^{58,60}$Co</td>
<td>$&gt;10^5$</td>
<td>$&gt;10^5$</td>
<td>$^{89,90}$Sr</td>
</tr>
<tr>
<td>$^{152,154}$Eu</td>
<td>$&gt;10^6$</td>
<td>$&gt;10^5$</td>
<td>$^{41,45}$Ca</td>
</tr>
<tr>
<td>$^{151}$Sm</td>
<td>$&gt;10^6$</td>
<td>$&gt;10^5$</td>
<td>$^{36}$Cl</td>
</tr>
<tr>
<td>$^{54}$Mn</td>
<td>$&gt;10^5$</td>
<td>$&gt;10^6$</td>
<td>$^3$H</td>
</tr>
<tr>
<td>$^{51}$Cr</td>
<td>$&gt;10^6$</td>
<td>$&gt;10^5$</td>
<td>$^{14}$C</td>
</tr>
</tbody>
</table>

For all interfering radionuclides, the decontamination factors higher than $10^5$. 
**41Ca in the concrete**

Activation products of calcium isotopes

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Target isotope Abundance %</th>
<th>Reaction</th>
<th>Cross section, bar</th>
<th>Half life</th>
<th>Decay</th>
</tr>
</thead>
<tbody>
<tr>
<td>41Ca</td>
<td>96.94</td>
<td>40Ca(n, γ)41Ca</td>
<td>0.41</td>
<td>1.03×10^5 y</td>
<td>EC</td>
</tr>
<tr>
<td>45Ca</td>
<td>2.086</td>
<td>44Ca(n, γ)45Ca</td>
<td>0.84</td>
<td>162.7 d</td>
<td>β-</td>
</tr>
<tr>
<td>47Ca</td>
<td>0.004</td>
<td>46Ca(n, γ)47Ca</td>
<td>0.7</td>
<td>4.54 d</td>
<td>β, γ</td>
</tr>
<tr>
<td>49Ca</td>
<td>0.187</td>
<td>48Ca(n, γ)49Ca</td>
<td>1.0</td>
<td>8.72 min.</td>
<td>β, γ</td>
</tr>
</tbody>
</table>

**Energy of X-rays and Auger electrons**: 0.3-3.6 keV  
Determination: X-ray spectrometry (<0.08%)  
LSC (10-20%)
Sr & Ca: Alkline earth element
Determination $^{41}$Ca in concrete

- Separation from matrix
  - Decomposition of heavy concrete by alkali fusion
  - Leaching Ca by acids
- Separation from active metals such as $^{60}$Co, $^{152}$Eu, $^{55}$Fe, $^{63}$Ni, $^{65}$Zn, $^{54}$Mn, $^{51}$Cr, etc.
  - Precipitation with Fe(OH)$_3$ by hydroxides at pH9
- Separation from other alkaline metals, such as $^{133}$Ba, $^{226}$Ra and $^{90}$Sr.
  - BaCrO$_4$ and SrCrO$_4$ precipitation
  - BaCl$_2$ and SrCl$_2$ precipitation in HCl solution
  - Ca(OH)$_2$ precipitation in NaOH solution
Decomposition of concrete for $^{41}$Ca and other radionuclides

- For ordinary concrete, silicates of calcium does not easily be decomposed by acids.
- For heavy concrete, some calcium exists as CaSO$_4$, which does not dissolved by acid.
- Alkaline fusion have to be used for the decomposition of concrete for the determination of calcium isotopes.
Separation of Ca and Sr from other metals by hydroxides precipitation

<table>
<thead>
<tr>
<th>Element</th>
<th>Recovery, %</th>
<th>Element</th>
<th>Decontamination factor</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ca</td>
<td>97.7±3.9</td>
<td>$^{137}\text{Cs}$</td>
<td>$(4.5±0.3)\times10^5$</td>
</tr>
<tr>
<td>$^{85}\text{Sr}$</td>
<td>97.9±2.1</td>
<td>$^{60}\text{Co}$</td>
<td>$(1.2±0.4)\times10^5$</td>
</tr>
<tr>
<td>$^{133}\text{Ba}$</td>
<td>97.3±2.8</td>
<td>$^{152}\text{Eu}$</td>
<td>$(8.5±0.5)\times10^5$</td>
</tr>
<tr>
<td></td>
<td></td>
<td>$^{59}\text{Fe}$</td>
<td>$(2.5±0.1)\times10^5$</td>
</tr>
<tr>
<td></td>
<td></td>
<td>$^{63}\text{Ni}$</td>
<td>$(2.5±0.2)\times10^5$</td>
</tr>
</tbody>
</table>
The new method for the separation of Ca from Sr and Ba

- Separation of Sr from Ca by Ca(OH)$_2$ precipitation
  - Ca(OH)$_2$: insoluble, $K_{sp} = 5.2 \times 10^{-6}$
  - Sr(OH)$_2$ and Ba(OH)$_2$: Soluble in alkine solution

Precipitate Ca as Ca(OH)$_2$ at 0.5 – 0.8 mol/l NaOH, repeat 3 times, 85% Ca can be recovered, and the decontamination factor for Sr and Ba are higher than $5 \times 10^4$
Spectra of $^{41}$Ca in heavy concrete from DR-2
Features of Method for $^{41}$Ca

- A separation of $^{41}$Ca from concrete is easy to operate
- Good decontamination from interfering radionuclides ($>10^4$)
- The chemical yields of the separation procedures for $^{41}$Ca is 80-90%.
- The detection limits for $^{41}$Ca is 0.020 Bq.

Hou X.L., Radiochim Acta, 2005
- $^{36}\text{Cl}$ is long-lived radionuclides ($3\times10^5$ yrs)
- $^{36}\text{Cl}$ decays mainly by pure beta emission of $E_{\text{max}}=708.6$ keV.
- $^{36}\text{Cl}$ measurement is normally carried out by LSC and AMS.
Determination of $^{36}$Cl and $^{129}$I in graphite

--- Sample decomposition

- Ashing at 900°C: iodine and part of Cl are lost.

- Decomposition at 900°C with $O_2$ and trapping iodine in NaOH solution: good recovery for iodine, but not good for chlorine.

- Leaching with acid (HNO$_3$) at heating: not complete remove iodine and Cl from graphite, and loss of the leached iodine.

- Digestion with HNO$_3$ and trapping iodine and chlorine with NaOH: Not complete removal of iodine and chlorine.

How to Do?
Determination of $^{36}\text{Cl}$ and $^{129}\text{I}$ in graphite
--- Sample decomposition

- Graphite can be completely dissolved in a mixture of acids: HNO$_3$+H$_2$SO$_4$ +HClO$_4$

- The optima ratio of mixture is:
  $$\text{H}_2\text{SO}_4:\text{HNO}_3:\text{HClO}_4 = 15:4.1$$

- A closed dissolution system is used for dissolve graphite in heating, Cl on the condenser tube and trap solution, while iodine mainly in the trap solution.
Schematic diagram of dissolution system of graphite for determination of $^{36}$Cl and $^{129}$I

1-Heating mantle; 2-three-necked flask; 3-sample in acid mixture; 4-bubbling tube; 5-separating funnel for adding acids; 6, 7-reflux condenser; 8-receiver; 9-wash bottle containing water; 10, 11-absorption bottles containing 0.4 mol/l NaOH
Determination of $^{36}\text{Cl}$ and $^{129}\text{I}$ in concrete
--- Sample decomposition

- Leaching with acid (HNO$_3$) at heating: --- not complete removal of iodine and Cl from graphite, and loss of the leached iodine.

- Digestion with HNO$_3$ and trapping iodine and chlorine with NaOH: ---- Not complete removal of iodine and chlorine

Alkalinal fussion using NaOH and Na$_2$CO$_3$, dissolution of fused cake in water, the supernatant is used for $^{129}\text{I}$ and $^{36}\text{Cl}$: ---- sample is completely decomposed and iodine and Cl are released. Iodine and Cl are not lost in alkaline medium
Determination of $^{36}\text{Cl}$ and $^{129}\text{I}$ in stainless steel

--- Sample decomposition

- Stainless steel is normally dissolved with HCl or HCl+$\text{HNO}_3$: --- could not be used for $^{36}\text{Cl}$ because of too much Cl in HCl is introduced.

- Single acid, HNO$_3$, could not dissolve stainless steel.

- 10M $\text{H}_2\text{SO}_4$ with $\text{H}_3\text{PO}_4$ is successfully used for dissolve stainless steel for $^{129}\text{I}$ and $^{36}\text{Cl}$: ---- sample is completely decomposed and iodine and Cl are released.
Separation of Cl from matrices and other radionuclides

✓ Specific precipitation of Cl$^-$ with Ag$^+$ (AgCl) can be used to selectively separation of Cl from matrix and other radionuclides (except iodine and bromine).

✓ Iodine ($^{129}$I) should be first separated from the solution before AgCl precipitation.

✓ No need to separate Br, since no long-lived radioisotopes of Br in the waste and environmental samples.

✓ The separated AgCl can be dissolved in NH$_4$OH and mixed with scintillation cocktail for LSC: But less AgCl can be used and high quench effect. How to improve?
Separation of Ag⁺ and Cl⁻ in anion exchange chromatography
Combined Analytical procedure for $^{36}$Cl and $^{129}$I

Graphite, steel
- Add stable Cl and I carriers, dissolve with acids, combine the absorption solutions and dissolved solution
- Add NaHSO$_3$, transfer to a separation funnels, add CCl$_4$, HNO$_3$, and NaNO$_2$ to extract, repeat the extraction
- Aqueous phase, $^{129}$I
  - LSC for $^{129}$I
  - ICP-MS for I
- Aqueous phase, $^{129}$I
  - Back-extract with NaHSO$_3$ solution, repeat extraction and back-extraction

Heavy Concrete
- Add stable Cl and I carriers and NaOH and Na$_2$CO$_3$, fuse at 500 °C, leaching with H$_2$O, centrifuge to separate the supernatant
- Add Ag$^+$, centrifuge, dissolve AgCl with NH$_3$, separate solution, add HNO$_3$ to re-precipitate AgCl, re-dissolve AgCl with NH$_3$, load to the column
- Aqueous phase
  - Washes, Ag$^+$ etc.
  - Effluent, $^{36}$Cl
  - Evaporate, dissolved with H$_2$O
  - ICP-MS for Cl
  - LSC for $^{36}$Cl
- AG 1x4 column
  - Wash with NH$_3$
  - Eluting with 0.1 M NH$_4$NO$_3$-0.6 M NH$_4$OH

Hou et al., Anal. Chem., 2007
Performance of the procedure for $^{36}\text{Cl}$

- Recovery of Cl: $>70\%$
- Decontamination factors for most of radionculides: $>10^6$
- Detection limit using LSC: 14 mBq
- Decommissioning samples, concrete, graphite, stainless steel, aluminum, lead, have been successfully analysed for $^{36}\text{Cl}$
Combined analytical procedure for \( ^{36}\text{Cl} \), \( ^{129}\text{I} \), \( ^{41}\text{Ca} \), \( ^{63}\text{Ni} \) and \( ^{55}\text{Fe} \)

- **Concrete**
  - Add Fe, Ni, Ca, Cl and I carriers and hold-back carriers, NaOH, NaCO₃, fuse at 500 °C, leaching with water, centrifuge, washing precipitate with 0.2 M NaCO₃ for 4 times
  - Dissolve with HCl, Add NaOH to pH9, centrifuge

- **Organic phase, I**
  - Add NaHSO₃ solution to back-extract iodine
  - Repeat extraction and back-extraction

- **Aqueous phase, I**
  - Add NaOH to pH10 and evaporate to 3 ml

- **Precipitate M(OH)ₓ**
  - Dissolve with HCl, Add NaOH to 0.5 mol/l, centrifuge, and repeat for 3 times

- **Precipitate Ca, Sr, Ba**
  - Evaporate to 1-7 ml

- **Supernatant**
  - Evaporate to dryness, dissolve with \( \text{H}_3\text{PO}_4 \)

- **Supematant, Ca, Sr, Ba**
  - Evaporate to 3 ml
  - Load to column
  - Dissolve with \( \text{HNO}_3 \)
  - Elute with 0.2 M NH₄Citr, NH₄OH to pH9, loading

- **Eluate, \(^{55}\text{Fe} \)**
  - Evaporate to 9 mol/l HCl, loading to anion exchange column
  - Eluting with 0.5 mol/l HCl

- **Effluent, \(^{63}\text{Ni} \)**
  - Evaporate, dissolved with HCl, add NH₄Citr, NH₄OH to pH9, loading

- **Eluate, \(^{55}\text{Fe} \)**
  - Evaporate to 6 M HCl, loading

- **Precipitate \(^{41}\text{Ca} \)**
  - Washing with 0.2 M NH₄Citr, Eluting with 3 M HNO₃

- **Eluate, \(^{63}\text{Ni} \)**
  - Evaporate to 9 mol/l HCl, loading

- **Precipitate, AgCl**
  - Add AgNO₃ to extract, repeat.
  - Add AgCl, add HNO₃ to pH2, centrifuge, washing precipitate with \( \text{H}_2\text{O} \), repeat precipitate

- **Solution, \( \text{Ag(NH}_3\text{)}_2^+ \), Cl⁻**
  - Load to column
  - Dissolve with \( \text{HNO}_3 \)
  - Elute with 0.2 M NH₄Citr, 0.6 M NH₄OH solution

- **Eluate \(^{36}\text{Cl} \)**
  - Evaporate to \( \text{H}_2\text{O} \)

**LSC measurement for \(^{36}\text{Cl}, \(^{129}\text{I}, \(^{41}\text{Ca}, \(^{63}\text{Ni}, \(^{55}\text{Fe} \text{}} \) and ICP-MS or ICP-AES measurement for chemical yield of Cl, I, Ca, Ni, Fe AES**

**Steps**
- Add CCl₄, HNO₃ to pH2, add NaNO₂ to extract, repeat.
- Add 25% \( \text{NH}_3 \) to dissolve AgCl, add HNO₃ to pH2, centrifuge, washing precipitate with \( \text{H}_2\text{O} \), repeat precipitate
- Add NaOH to pH10 and evaporate to 3 ml
- Add 25% \( \text{NH}_3 \) to dissolve
- Dissolve with \( \text{HNO}_3 \)
- Evaporate to dryness, dissolve with \( \text{H}_3\text{PO}_4 \)
Sampling of concrete and graphite from danish reactor, DR-2
Results of $^{55}\text{Fe}$, $^{63}\text{Ni}$, $^{36}\text{Cl}$ and $^{41}\text{Ca}$ in concrete from Aanish reactor, DR-2

$^{55}\text{Fe}$ and $^{63}\text{Ni}$ in graphite of DR-2

<table>
<thead>
<tr>
<th>Sample No</th>
<th>$^{55}\text{Fe}$</th>
<th>$^{63}\text{Ni}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>DR-3-T</td>
<td>92.2</td>
<td>94.63</td>
</tr>
<tr>
<td></td>
<td>545000</td>
<td>5552</td>
</tr>
<tr>
<td>IY7.5</td>
<td>90.4</td>
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<td>91.56</td>
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<tr>
<td></td>
<td>9.21</td>
<td>43.1</td>
</tr>
</tbody>
</table>

Free release criteria:

$^{63}\text{Ni} < 10$ Bq/g; $^{55}\text{Fe} < 1000$ Bq/g

- More then 65% of concrete shield was treated as non-radioactive waste