Determination of ^{36}Cl and ^{129}I in solid nuclear decommissioning materials

I. Llopart Babot[1,2,3], **M. Vasile**[1], **A. Dobney**[1], **S. Boden**[1], **M. Bruggeman**[1], **M. Leermakers**[2], **J. Qiao**[3]
[1]Belgian Nuclear Research Centre, SCK CEN, Mol, Belgium
[2] Vrije Universiteit Brussel, VUB, Brussels, Belgium
[3] Technical University of Denmark, DTU, Lyngby, Denmark
E-mail: ines.llopart@sckcen.be

Topic's presentation
PhD day, 13th November 2020

1. Introduction

Decommissioning materials need to be nuclide specific characterized due to legislation requirements

What is needed? [1]

DECOMMISIONING MATERIALS

Release: Unconditional; Conditional; Near Surface Disposal

Radioactive waste: Deep Geological Disposal

Radiological risk of waste

Quantification of the activity concentration of single radionuclides

Current approach

Scaling factors [2]

Activity α and β (DTM) ⟷ Activity γ-emitter (ETM)

- Use calculation codes
- Validated with real analysis

Current issues and needs

- **High uncertainty on the inventory**

Requirements nowadays

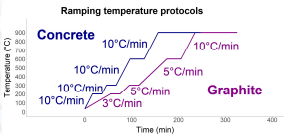
- Fast methodologies
- Reference materials
- Low Detection Limit (LD)

^{36}Cl and ^{129}I determination ➔ DTM radionuclides[3]

^{36}Cl	^{129}I
β-particle emitter	Low β-particle emitter
E_{max} = 709.6 keV	E_{max} = 154 keV
$T_{1/2}$ = 3.02 · 10^5 years	$T_{1/2}$ = 1.57 · 10^7 years

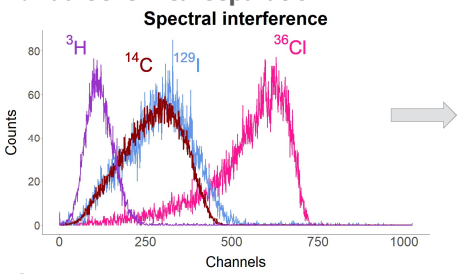
2. Materials and methods

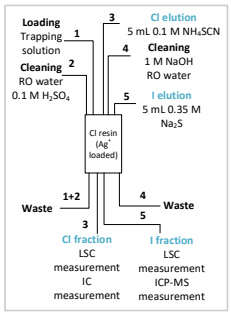
A. Sample combustion



- Pyrolysis up to 900°C [4]
- Flush-gas flow-rate: 200 mL/min
- No catalyst/quartz beats
- Glass connections and quartz tubes and sample boats
- ↓ temperature ramp rate ↑ matrix organic content

B. Radiochemical separation [5]





C. Measurement

Massic activity → **Chemical recovery quantification (η)**

Liquid Scintillation Counting (LSC)	Ion chromatography (IC)	Inductively Couple Plasma Mass Spectrometry (ICP-MS)
Low level LS counter	Stable Cl	Stable I

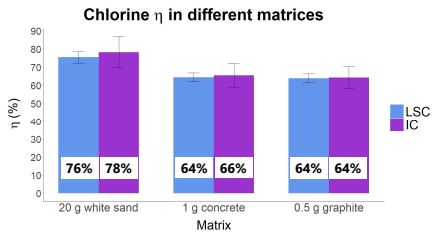
5 mL elution medium measured

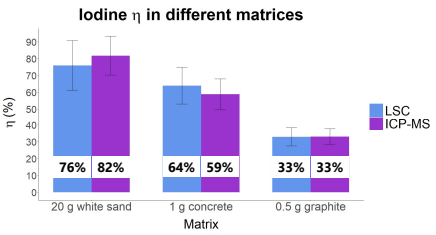
$$A\left(\frac{Bq}{g}\right) = \frac{CPS_{net}}{C_{eff} * m_{sample} * \eta}$$

0,1 mL subsample from elution medium

3. Method optimization and robustness testing

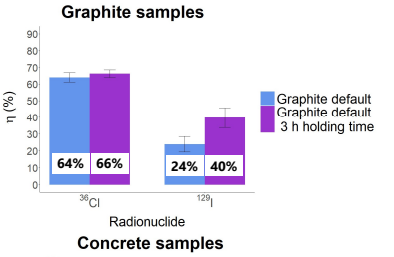
Testing different matrices and techniques

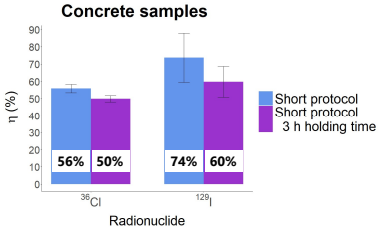




- Optimized procedure can be applied to different matrices
- Iodine low recovery in graphite samples can be due to physical adsorption and chemisorption
- LSC can be used for calculating the chemical recovery during the optimization step

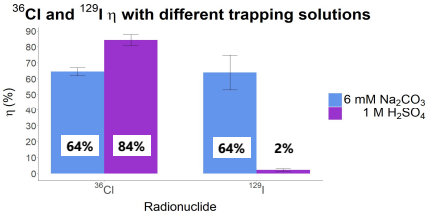
Testing different temperature protocols





- Shorter protocol can be used for inorganic matrices such as concrete
- Extra **holding time at 900 °C** reduce the η in concrete samples due to calcination
- Extra **holding time at 900 °C** enhance ^{129}I η due to the stimulation of I release at ↑ temperatures

Testing different trapping solutions



- 1 M H_2SO_4 better approach for single ^{36}Cl determination
- 6 mM Na_2CO_3 have to be used for simultaneous determination of ^{36}Cl and ^{129}I

Detection limit (DL)

- Based on measurements of several blanks
- According to ISO 11929

	^{36}Cl	^{129}I
Count rate blank	3.6 CPM	3.6 CPM
Counting time	100 min	100 min
Mass sample	1 g	1 g
η	64%	65%
Counting efficiency	98%	92%
DL (kBq/kg)	2E-2	2E-2
Release level (kBq/kg)	1	1E-2

4. Conclusions and further work

- Optimized procedure can be applied for analyses of real samples
- Blanks are required after running an active sample (memory effect evaluation)
- Optimized procedure can reach the required limits by the legislation for ^{36}Cl, however, some improvements are needed for ^{129}I determination
- → Application of Plastic Scintillating Microspheres (PSm) for ^{36}Cl determination

5. Scientific output

- Invited talk in international LSC 2020 Conference, China, 18-20th October 2021
- Oral Presentation in international vCARM 2021 Conference, UK, 24th November 2021
- Paper *"Method for the determination of ^{36}Cl and ^{129}I in solid materials coming from decommissioning activities"* to be submitted 30th November 2021

References

[1] WNA, 'Methodology to Manage Material and Waste from Nuclear Decommissioning', *Rep. No. 2019/001*, p. 112, 2019
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[5] A. Zulauf, 'Characterization of extraction chromatographic resins and method development for the separation and determination of Pd / Pt and ^{36}Cl / 129 I', 2010

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