CARBON-14 RELEASE FROM IRRADIATED STAINLESS STEEL

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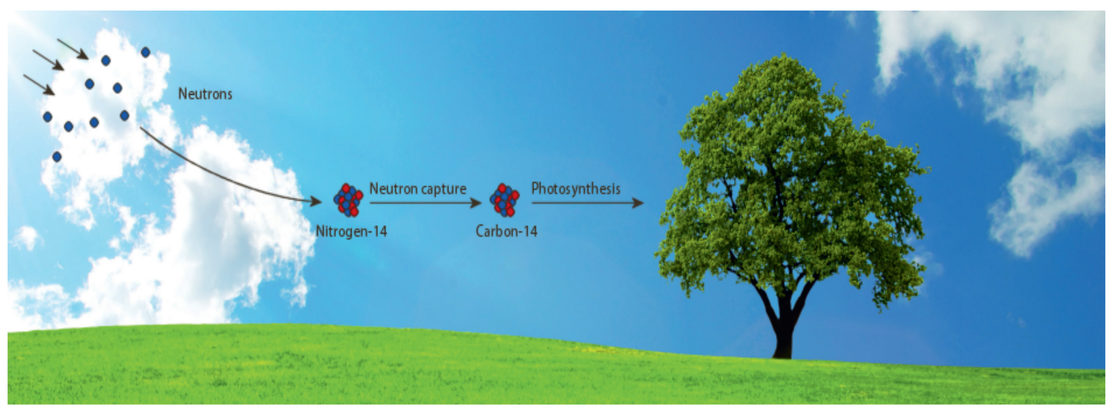
INTRODUCTION



CAST project http://www.projectcast.eu/

The CAST project (CArbon-14 Source Term) aims to develop understanding of the potential release mechanisms of carbon-14 from radioactive waste materials under conditions relevant to waste packaging and disposal to underground geological disposal facilities. The expected increase in understanding should decrease uncertainties in the long-term safety assessment and increase confidence in the safety case.
Carbon-14 may be released from irradiated steel wastes as they slowly corrode after closure of geological disposal facility. However, there is little information on the rate of carbon-14 release and its form. This study is investigating the form and rate of release of carbon-14 from an irradiated stainless steel under alkaline conditions.

EXPERIMENTAL

LEACHING EXPERIMENTS

- Leaching in 0.1M NaOH (pH 13) under nitrogen at the ambient temperature in the hot cell
- Duplicate experiments on irradiated steel samples
  - 3 CT specimens as obtained
- Identical experiment on un-irradiated steel sample (same batch)
- Gas and liquid phase periodic sampling
  - 1 week, 3 weeks, 6 weeks, 3 months, 6 months and 1 year
  - Gas phase purged and passed through RCD sampler system
  - 2 liquid samples for γ-spec (Co-60) and C-14 analysis (by NRG)
- Blank tests to measure C-14 background
- On termination, the container will be acid leached to recover any solid residues for γ-spec analysis

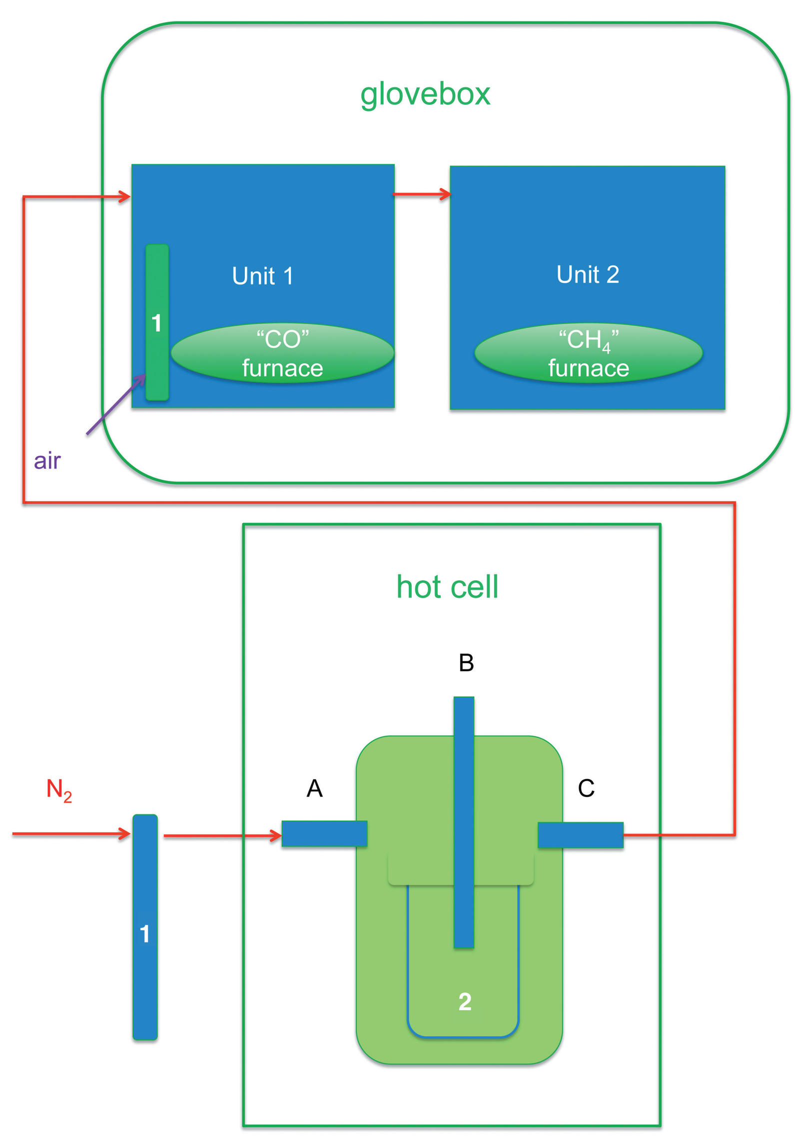
SAMPLES

- 316L(N) austenitic stainless steel from single sheet
- 6 compact tension (CT) specimens irradiated at HFR, Petten – SIWAS 07 experiment (2dpa, 80°C, 5 28-day cycles) in 1996/97
- C-14 and Co-60 inventory assessed by ORIGEN calculations
- Unirradiated steel from same sheet
- 3 experiments each with 3 CT specimens

Container	1	2	3
Mass (g)	228	221	222
Geo.SA. (cm^2)	104.4	114.4	114.4
C-14 (Bq)	0.1	4.9E+07	4.9E+07
Co-60 (Bq)	0	1.6E+10	1.6E+10

CT specimen
30x28.8x12 mm^3

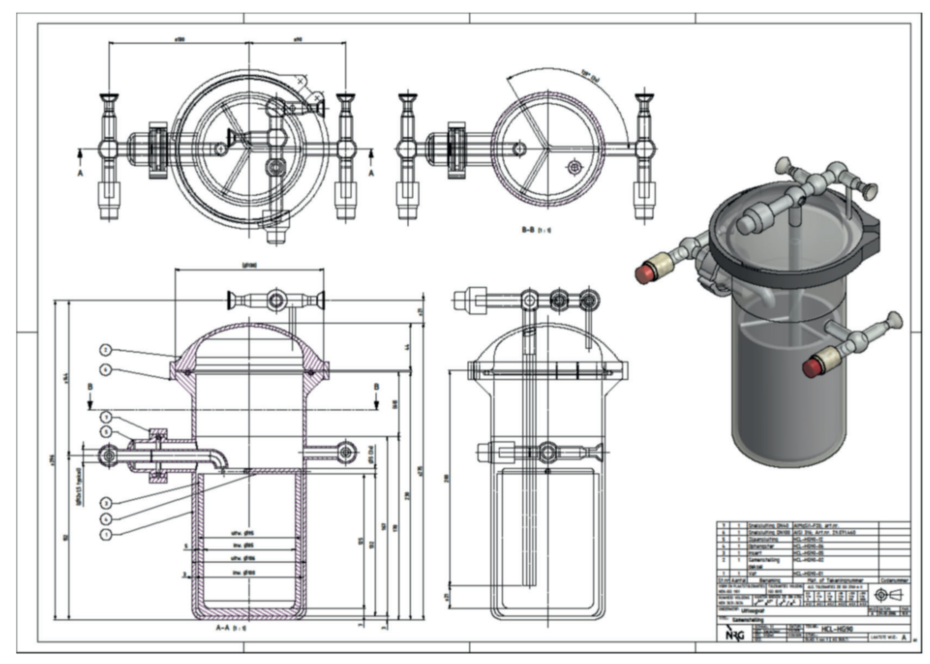
LEACHING SETUP



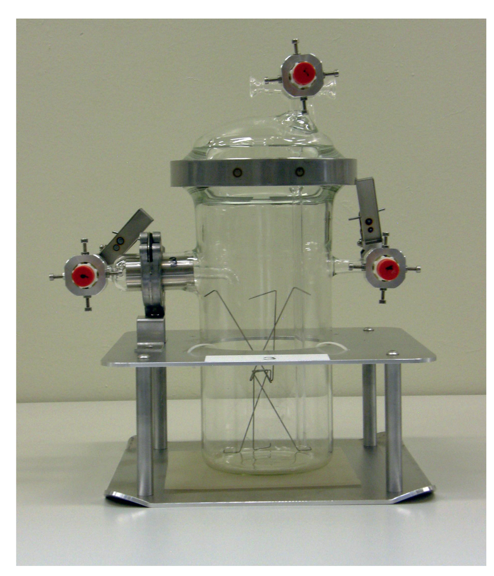
Setup for leaching tests: 1 is soda lime column for removing C-14 from nitrogen feed, 2 is leaching container where A is N2 inlet, B is dip leg and C is N2 outlet; this part is placed in the hot cell
Unit 1 and *Unit 2* are the parts of RCD rig for gaseous C-14 capture; placed in the glovebox

LEACHING CONTAINER DESIGN

- Borosilicate glass container with zirconia insert and dip- leg for sampling
- Adjusted for handling in the hot cellls



Technical drawing of the leaching container

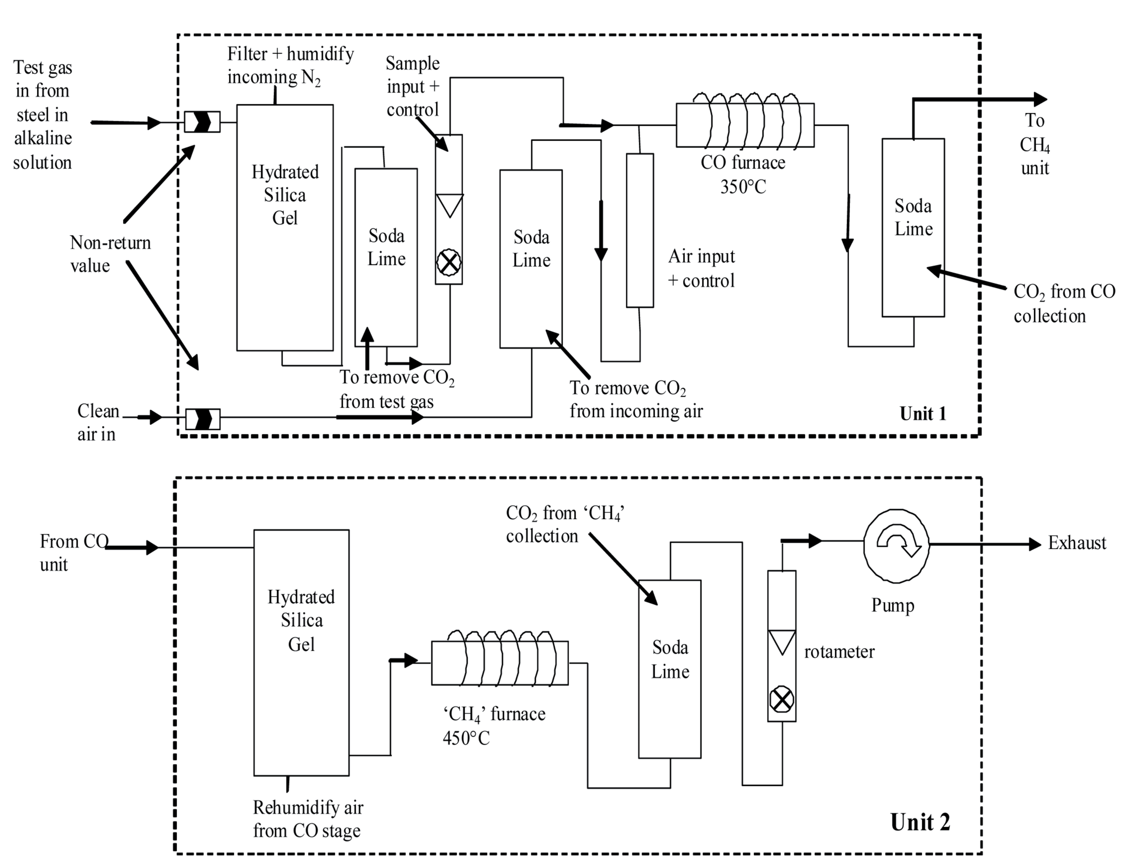


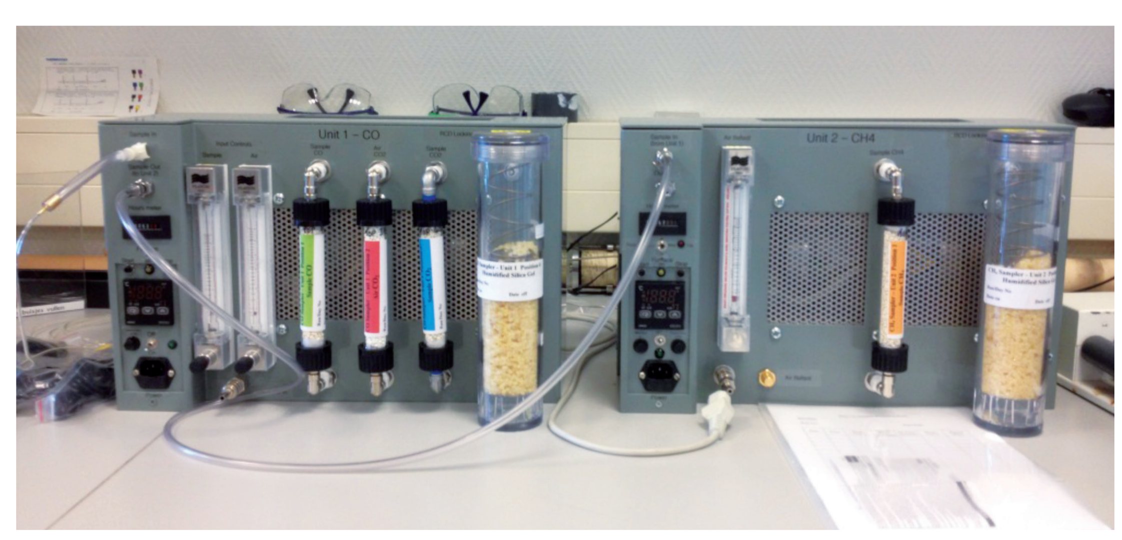
Picture of the leaching container (left) with a detail of fixing the CT specimens (below)

ANALYTICAL METHODS – GAS PHASE

RCD SAMPLER FOR THE SEPARATION AND QUANTIFICATION OF:

- carbon-14 released as CO_2;
- carbon-14 released as **CO** (any volatile oxygen-containing organic species e.g. alcohols, aldehydes and ketones that escape from solution into the gas phase would also be collected in this fraction); and
- carbon-14 released as **volatile hydrocarbons**, principally CH_4, (any other volatile carbon-containing species that have passed through the "CO" collection column would also be collected in this fraction).
- The three fractions are collected on separate soda lime columns (pre-loaded with fossil CO_2 as a bulk carrier).
- The CO_2 is recovered from each soda lime column by acidification, and is converted to benzene via reaction with lithium to produce lithium carbide, its hydrolysis to acetylene and catalytic trimerisation of the acetylene.
- Measured by liquid scintillation counting (Wallac Quantalus).
- Counting times of up to 2000 minutes have allowed limits of detection down to about 0.04 Bq carbon-14 per sample.





A schematic design (left) and a picture (right) of RCD rigs for collection of gas phase carbon-14

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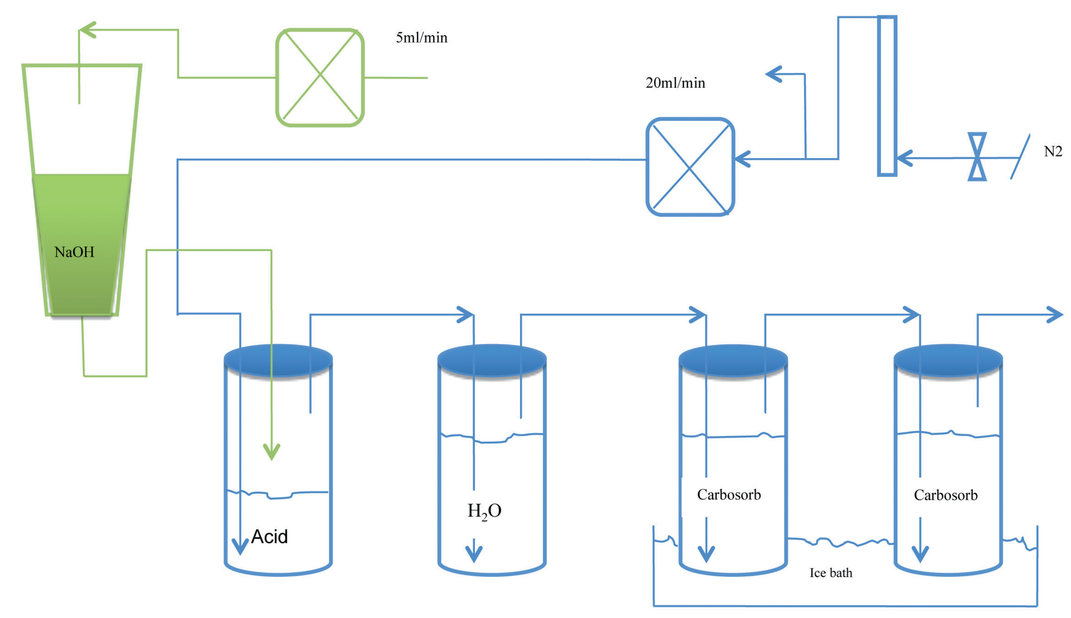
ANALYTICAL METHODS – LIQUID PHASE

C-14 ANALYSIS

- Separation of C-14 from the solution by acidification and capture in Carbosorb
- Measurement by Liquid Scintillation Counting - Packard TriCarb 3180 TR/SL
  - Minimum detectable activity is 0.1 Bq in sample



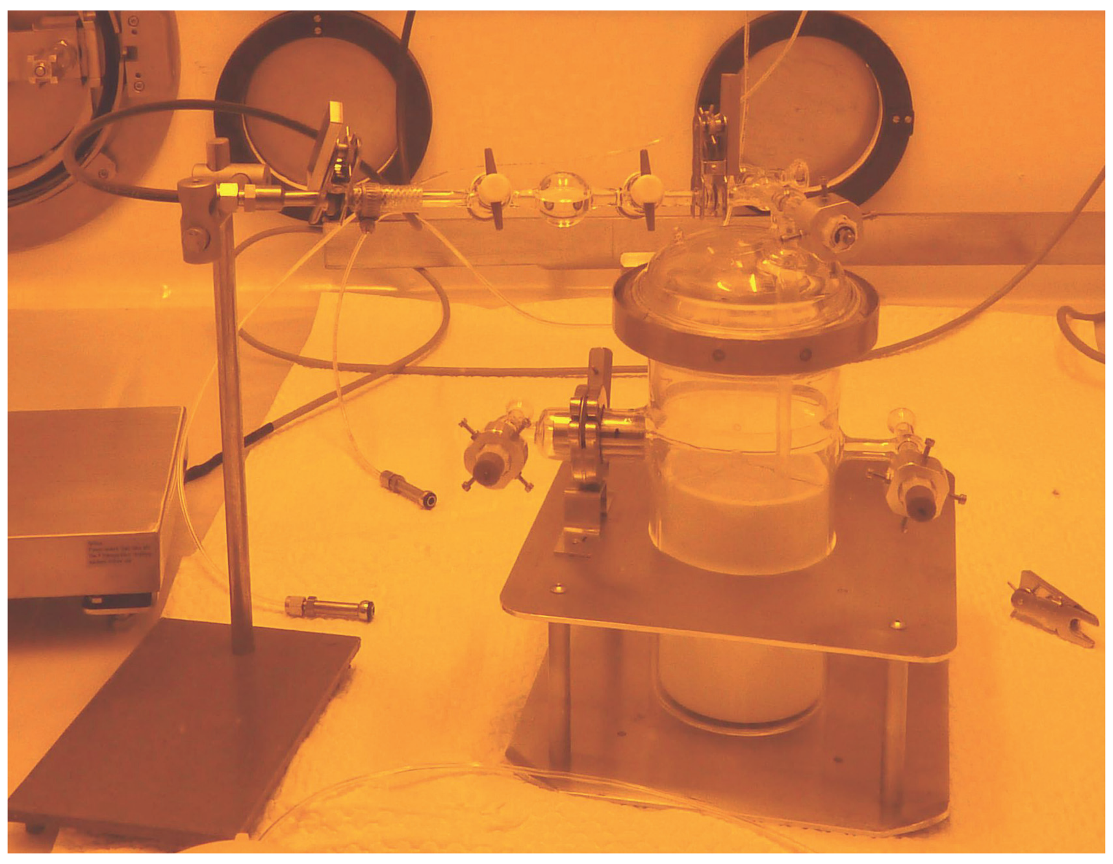
Setup for separation of C-14 from solution

CO-60 ANALYSIS

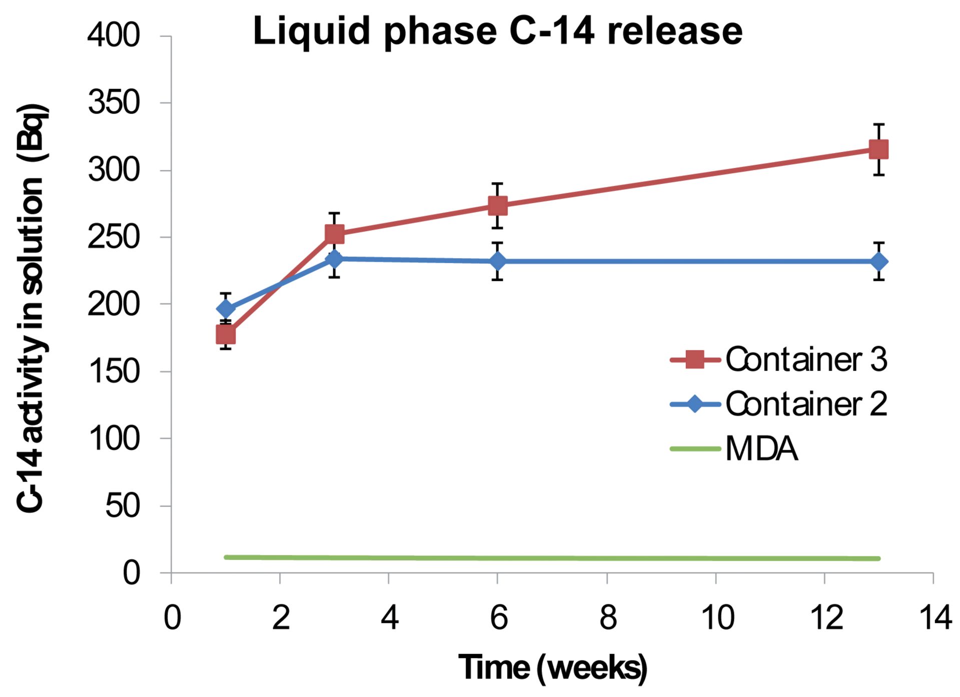
- Sub-samples transferred to 400 µl polythene container
- Gamma spectrometry – Canberra High purity Ge detector
  - Sample to detector - 10 cm
  - Measurement - 48 hours
  - Minimum detectable activity - 1.0 Bq

Gamma spec setup

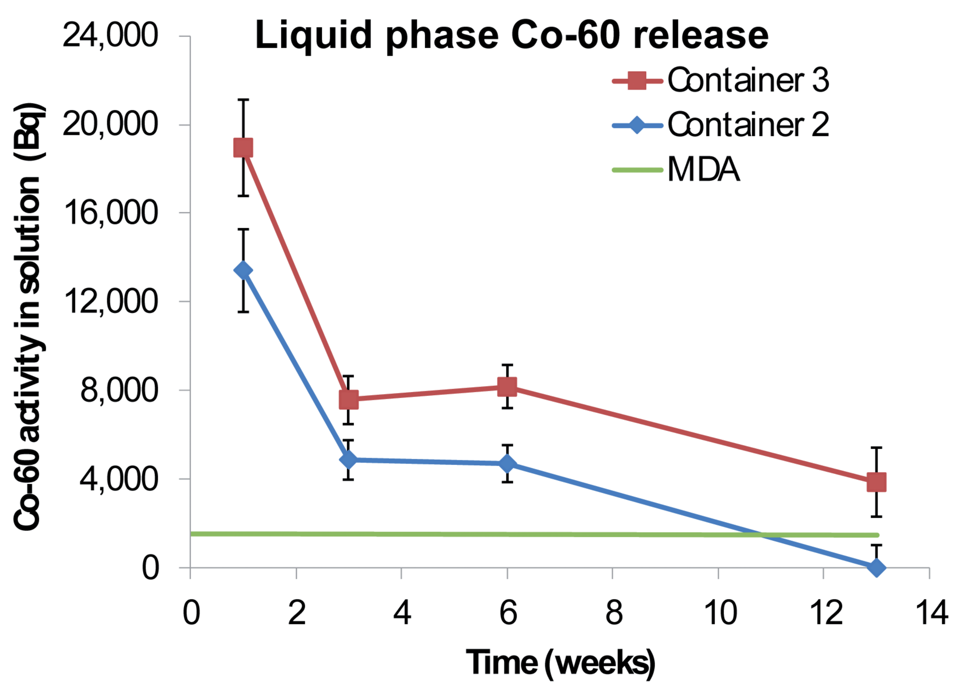
RESULTS



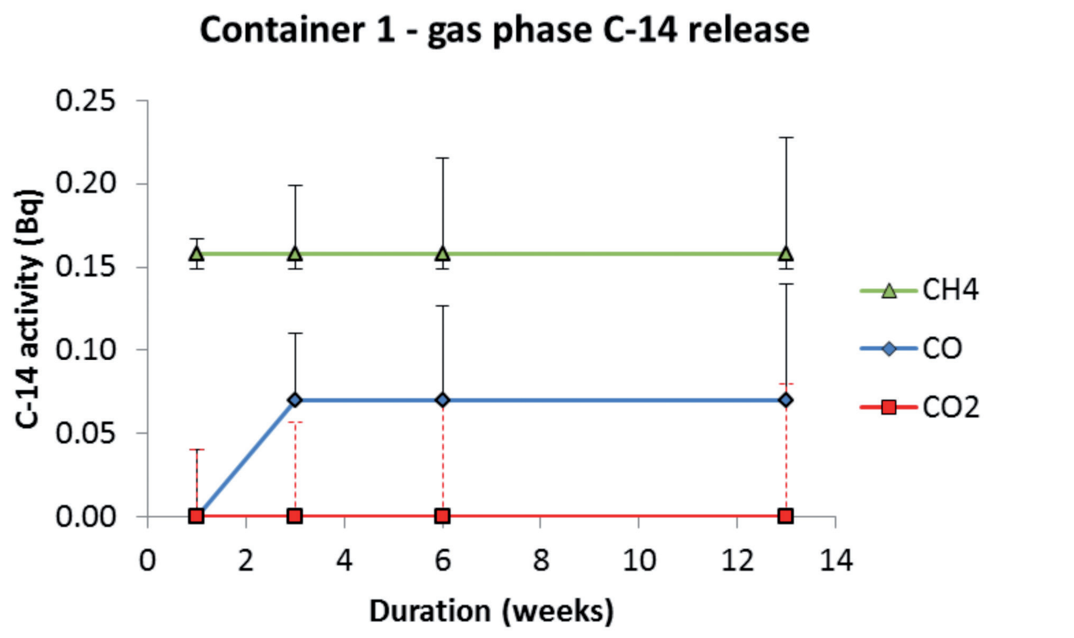
Leaching setup placed in the hot cell



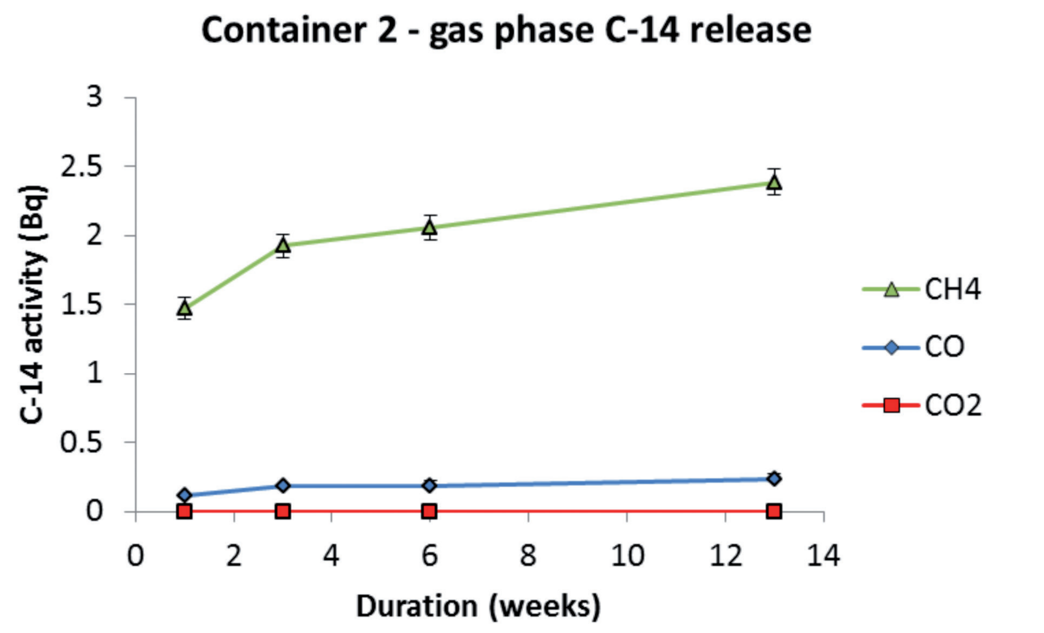
- Fast initial C-14 release, then rate decreases
  - Container 1 – no C-14 measurable
  - Container 2 – C-14 activity remains constant beyond 3 weeks
  - Container 3 – C-14 activity still increasing at constant rate



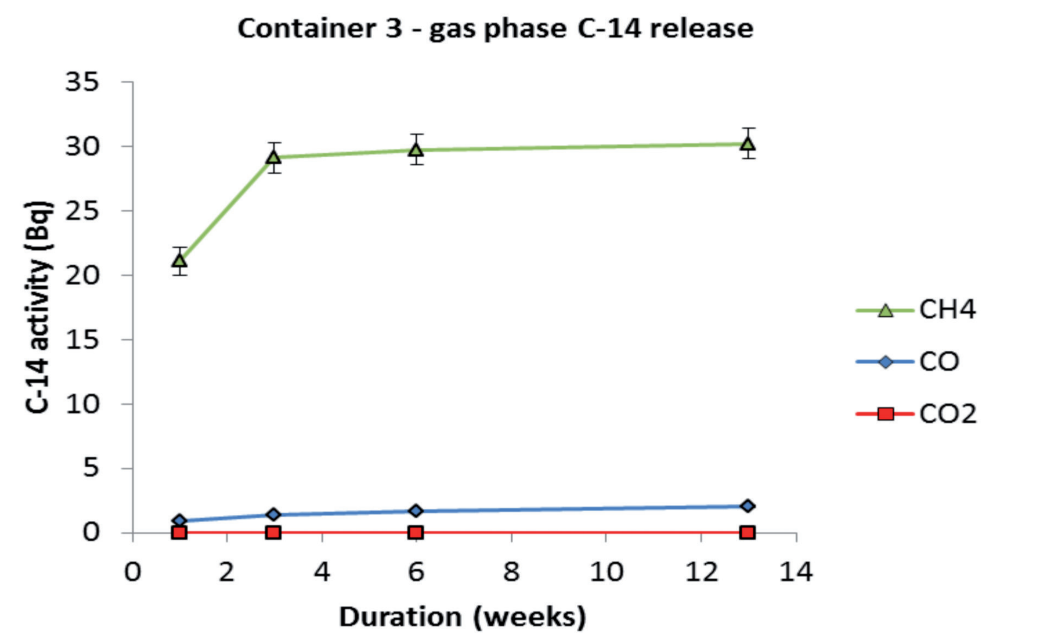
- High Co-60 activity in leachates after 1 week
  - 1 part in 106 of Co-60 inventory
- Then solution activity decreases
  - possible solubility limitation and/or sorption to vessel walls



- Unirradiated samples
- LoD <0.04 Bq
- 2 analyses give positive C-14 measurement above LoD



- **Majority C-14 release to gas phase as hydrocarbons**
- ~10% CO or volatile oxygenated organic compounds
- No measurable gas phase CO_2



- **Majority C-14 release to gas phase as hydrocarbons**
- ~6% CO and/or volatile oxygenated organic compounds
- No measurable gas phase CO_2

INTERIM CONCLUSIONS

- There is a relatively fast initial release of accessible C-14 species from the surface of the steel on immersion in water
  - Predominantly to solution phase but also to gas phase
  - Rate of release declines to near-zero over first 6 weeks in container 2
  - Continues at measurable rate in container 3, which also has higher gas phase release
  - Uncertainty over whether dissolved release is carbonate and/or soluble organic carbon-14 (e.g. carboxylic acids)
- The experiments are still running, further sampling of the experiments will provide data on the rate and speciation of carbon-14 release from the irradiated steels due to corrosion in the longer term. The experiments are planned to run for a minimum period of one year; once terminated, the leaching vessels will be emptied and the walls acid washed to recover deposits for cobalt-60 analysis by γ-spectroscopy.

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