EC 428 ECCN=N

CARBON-14 RELEASE FROM IRRADIATED STAINLESS STEEL

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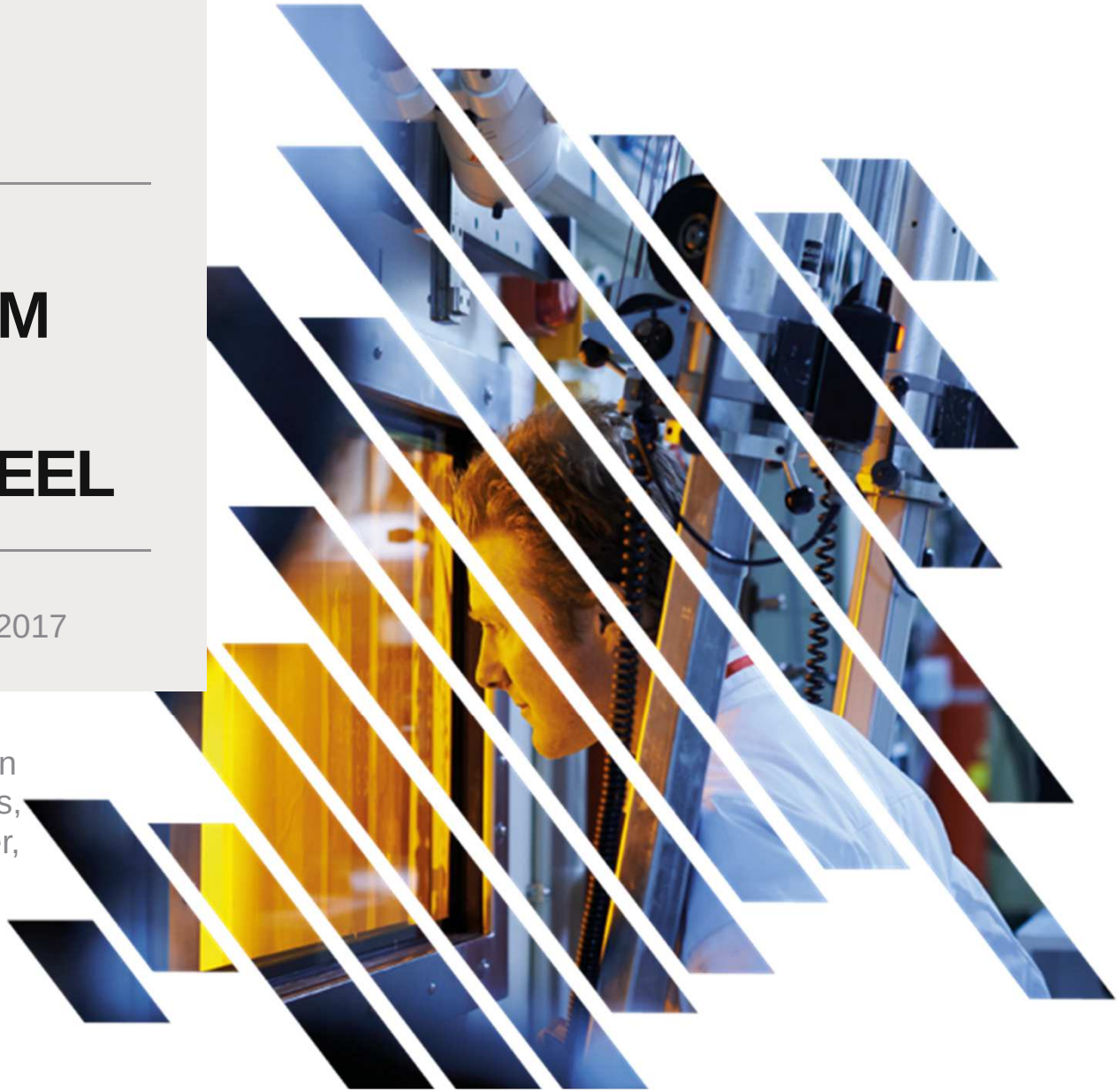


INTRODUCTION

- 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.

STEEL

- 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.

LEACHING EXPERIMENTS

- Leaching in 0.1M NaOH (pH 13) under nitrogen at the ambient temperature in the hot cell
  - slightly above room temperature
- Duplicate experiments on irradiated steel samples
  - 3 compact tension (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, 5 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)
- 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 CT specimens irradiated at HFR, Petten – SIWAS O7 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

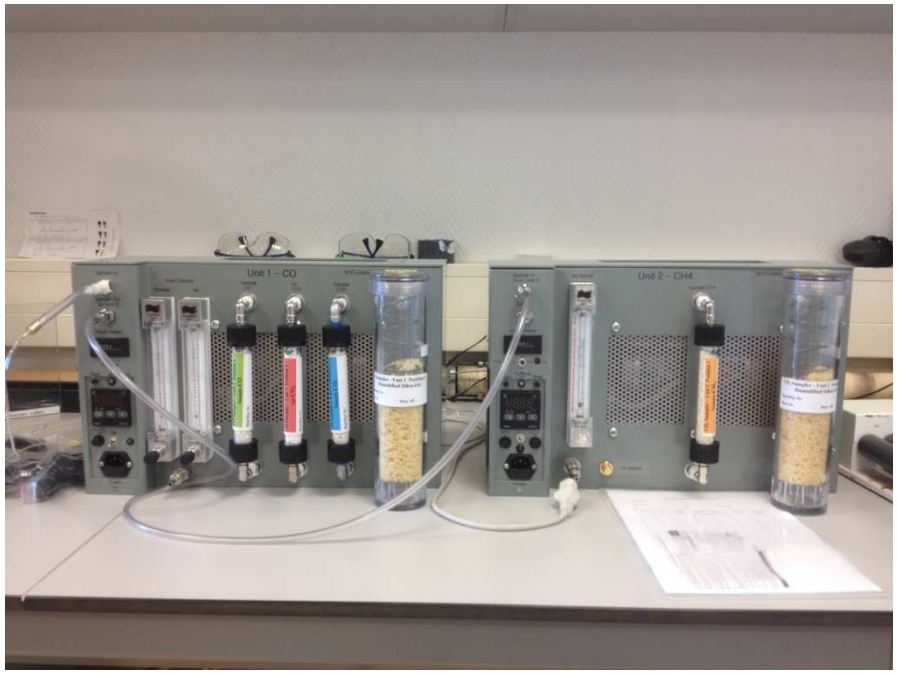
Container	**1**	**2**	**3**
Mass (g)	228	221	222
Geo.S.A. (cm^2)	104.4	114.4	114.4
C-14 (Bq)	0.1	4.9E+07	4.9E+07
Co-60 (Bq)	0	1.6E+10	1.6E+10

CT specimen
30x28.8x12 mm^3

GAS PHASE ANALYSIS

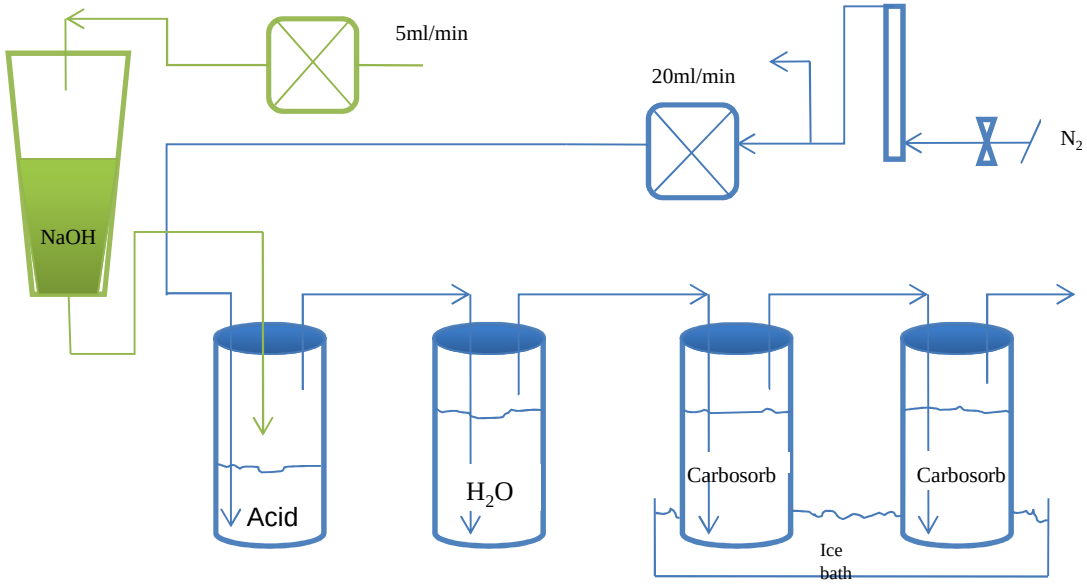
Designed and made at RCD in UK allows the separation and quantification of:

- ❑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).

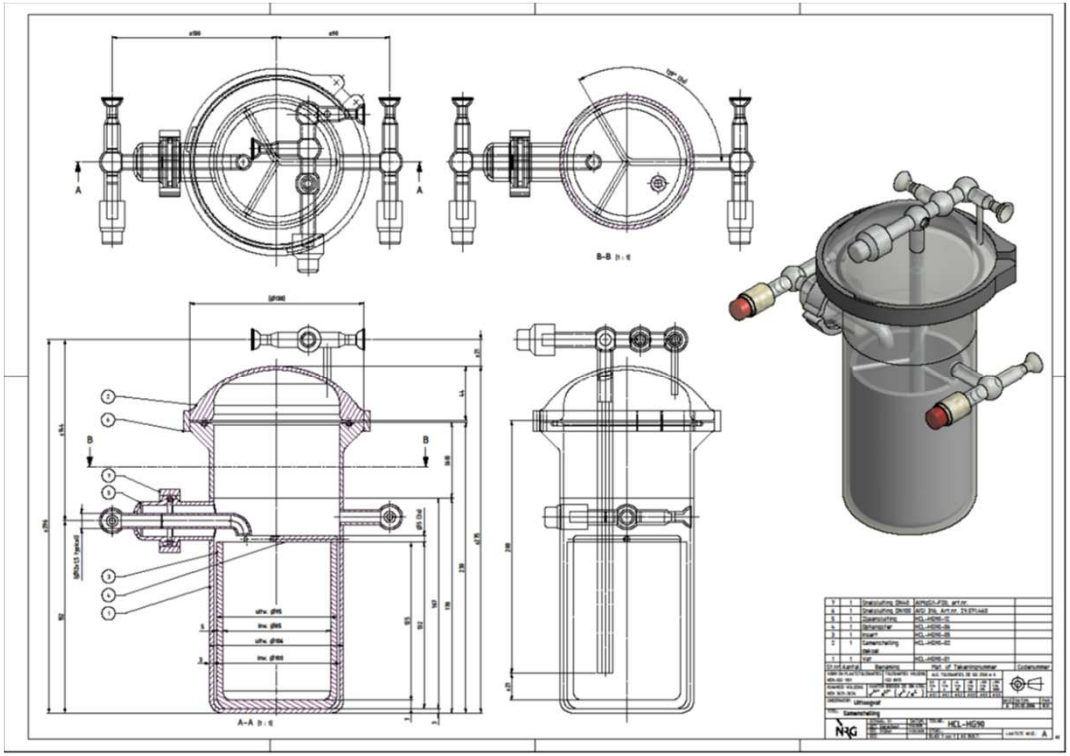


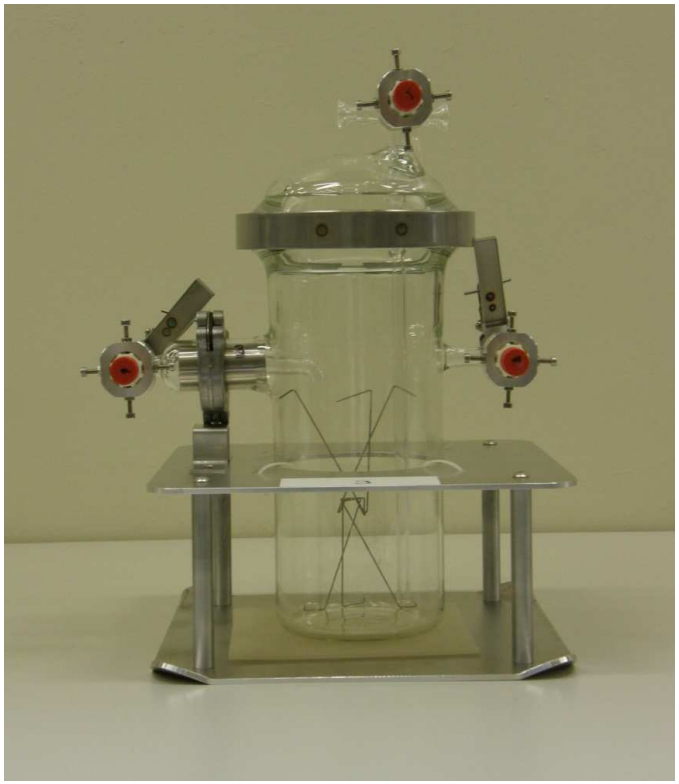
LIQUID PHASE ANALYSIS

- The rate of release of Co-60 into solution shall be measured by means of gamma spectrometry, in order to provide additional information on the corrosion of the sample.
- C-14 measurement will be done by LSC (Packard TriCarb 3180 TR/SL)

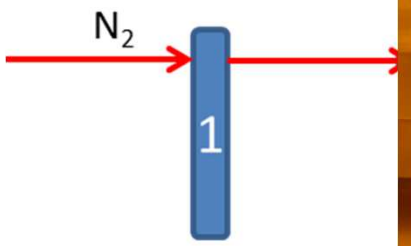


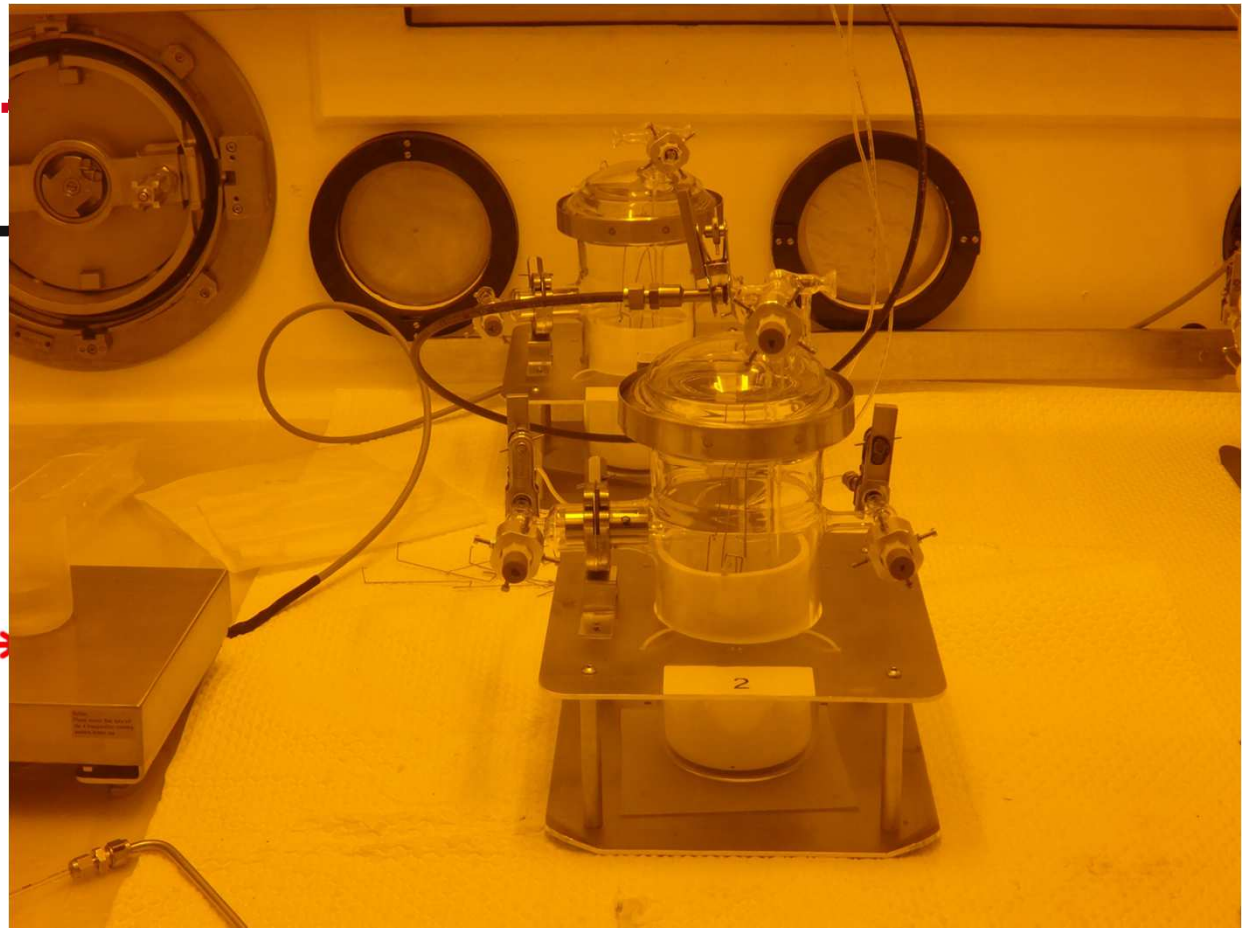
CONTAINER DESIGN





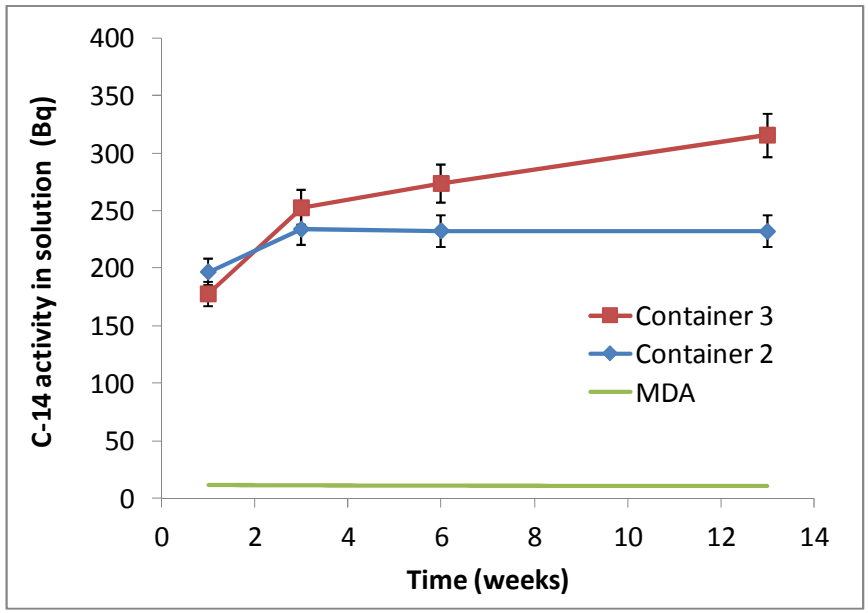
EXPERIMENT





Setup for blank tests: *1* is soda lime column for removing C-14 from nitrogen feed, *2* is leaching container where *A* is N_2 inlet, *B* is dip leg and *C* is N_2 outlet, *Unit 1* and *Unit 2* are the parts of RCD rig

RESULTS – C-14 SOLUTION PHASE



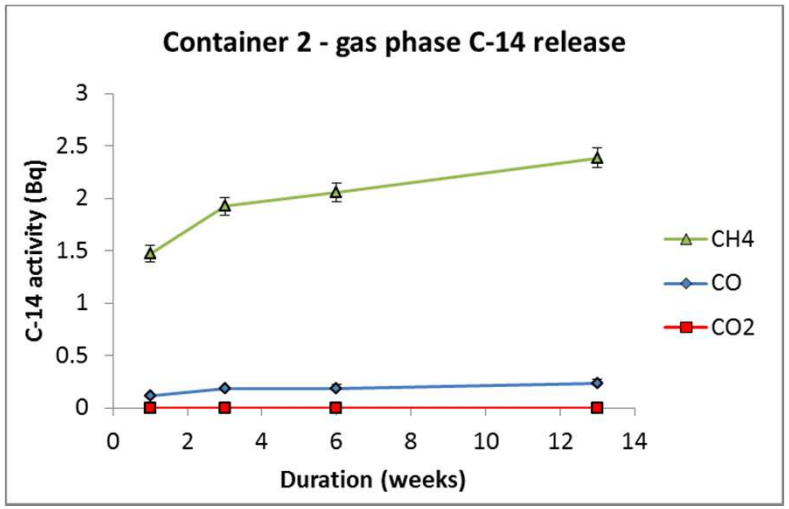
Containers 2, 3
49 MBq C-14

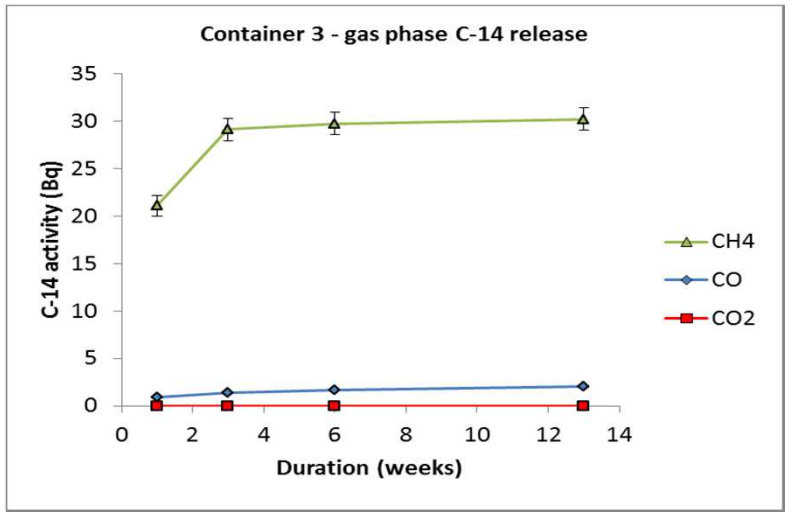
C-14 separated by addition of HNO_3, CO_2 released captured in Carbosorb E, analysed by LSC

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

RESULTS – C-14 GAS PHASE



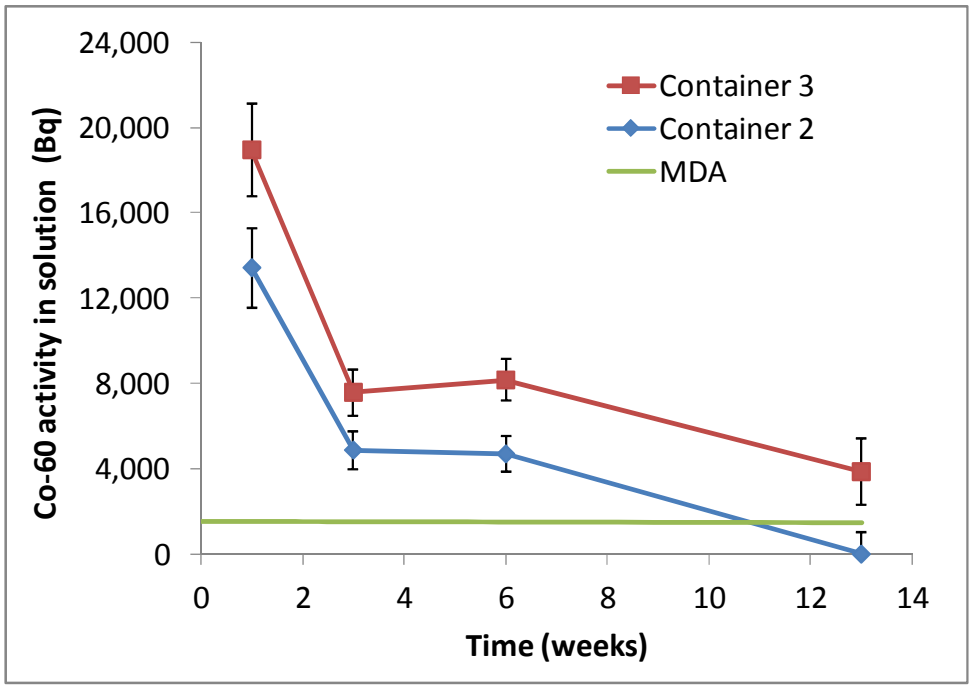


Majority C-14 release to gas phase as hydrocarbons

~6-10% release to gas phase as CO and/or volatile oxygenated organic compounds

No measurable gas phase CO_2

RESULTS – CO-60 SOLUTION PHASE



Container 2,3
16 GBq Co-60

High Co-60 activity in leachates after 1 week

- 1 part in 10^6 of Co-60 inventory

Then solution activity decreases

- possible solubility limitation and/or sorption to vessel walls

INTERIM CONCLUSIONS

- Initial releases of Co-60 and C-14 on immersion in solution much larger than expected (calculated)

Possibilities

- Higher C-14 and Co-60 concentrations on surface of steel?
- Higher initial corrosion rates than expected?
  - Some residual oxygen contributing to faster initial (aerobic) corrosion
  - Effective surface area larger than expected
    - surface finish, effects of prior corrosion, radiation damage?
  - Surface more reactive than unirradiated surface

PROJECT TEAM

Amec Foster Wheeler

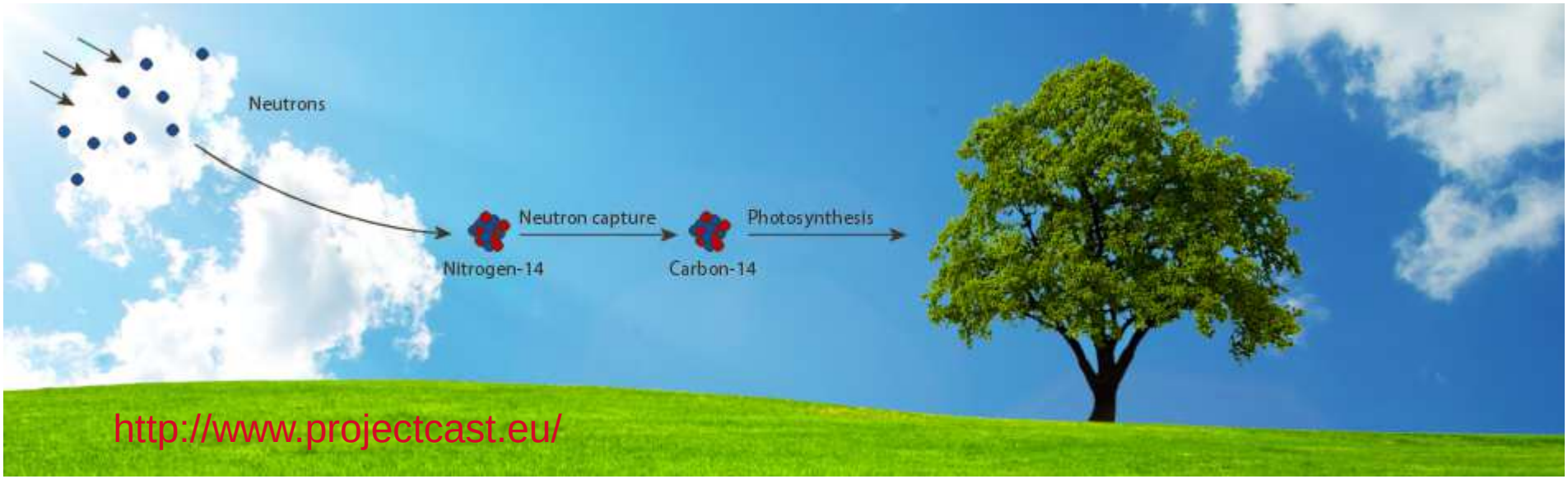
Nuclear Research and consultancy Group (NRG)

RadioCarbon Dating (RCD)

Radioactive Waste Management

Contact: devisser@nrg.eu

CARBON-14 SOURCE TERM (CAST) PROJECT



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