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Carbon-14 Source Term

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¹⁴CAST

Advisory Group Review of Year 3 WP Annual Reports and Minutes of Fourth CAST GAM (D1.9)

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CAST – Project Overview

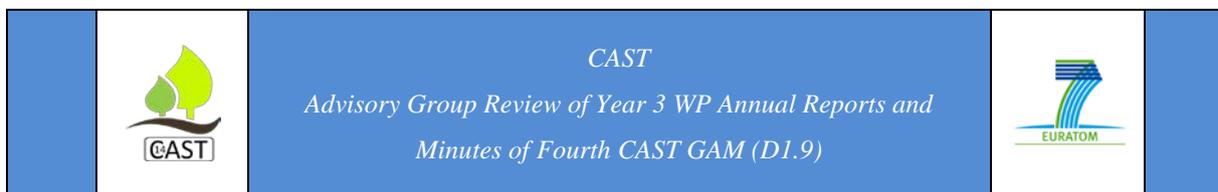
The CAST project (CARbon-14 Source Term) aims to develop understanding of the potential release mechanisms of carbon-14 from radioactive waste materials under conditions relevant to waste packaging and disposal to underground geological disposal facilities. The project focuses on the release of carbon-14 from irradiated metals (steels, Zircalloys), irradiated graphite and from ion-exchange materials as dissolved and gaseous species.

The CAST consortium brings together 33 partners with a range of skills and competencies in the management of radioactive wastes containing carbon-14, geological disposal research, safety case development and experimental work on gas generation. The consortium consists of national waste management organisations, research institutes, universities and commercial organisations.

The objectives of the CAST project are to gain new scientific understanding of the rate of release of carbon-14 from the corrosion of irradiated steels and Zircalloys and from the leaching of ion-exchange resins and irradiated graphites under geological disposal conditions, its speciation and how these relate to carbon-14 inventory and aqueous conditions. These results will be evaluated in the context of national safety assessments and disseminated to interested stakeholders. The new understanding should be of relevance to national safety assessment stakeholders and will also provide an opportunity for training for early career researchers.

For more information, please visit the CAST website at:

<http://www.projectcast.eu>



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Advisory Group Review of Year 3 WP Annual Reports and Minutes of Fourth CAST GAM (D1.9)

Executive Summary

One of the tasks of the CAST Advisory Group is to review progress towards meeting the various project goals. This report represents the fourth formal review performed by the Advisory Group and considers material published in the minutes of the fourth CAST General Assembly Meeting and the Year 3 annual progress reports from Work Packages 2, 3, 4, and 5. Section 1 contains the results of the review by Irka Hajdas and Section 2 describes the review of Fraser King.

Dr. Hajdas observed that during the third year of the CAST (Carbon-14 Source Term) Project activities were dedicated to the development of analytical methods leading to measurements of C-14 activities in various irradiated materials (steel, Zircalloys, ion-exchange resins, graphite) as well as in the carbon released from these disposal materials. Experiments simulating processes of release (corrosion and leaching) were performed. The anticipated research requires C-14 analysis that is mainly performed using counting techniques (LSC). However, some fractions can be of low carbon content and will require use of accelerator mass spectrometry AMS. During 2016 additional AMS facilities were contacted (Saclay, France; Helsinki, Finland; Seville, Spain) and analyses performed at RATEN ICN, Romania are reported.

Dr. King concluded that whilst the first two years of the CAST project involved establishing experimental protocols and obtaining irradiated samples, 2016 was focussed on obtaining new data and information. Progress continues to be made in all Work Package areas.

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1 Review of Dr. Irka Hajdas

1.1 WP2 (Steels) -- YEAR 3

12 organizations participate in WP2 (steels):

Amec Foster Wheeler (UK), Armines (FR), CIEMAT, ENRESA (ES), JRC (EU), KIT INE (DE), Nagra (CH; WP leadership), NRG (NL), PSI (CH), RWMC (JP), SCK·CEN (BE) and VTT (FI).

The reporting year (October 2015 to September 2016).

Technical Meetings: 1 shared with WP3 was held during the General Assembly Meeting on 13 October 2015 in Bucharest. A further Technical Meeting for WP2 and WP3 was organized by CIEMAT on 01 – 02 June 2016 in Madrid

WP2: group is working on following tasks:

Task 2.1 literature review on corrosion mechanisms, corrosion rates, release of C-14 and its speciation (completed)

Task 2.2 Development of analytical methods to measure C-14 in various species separated from liquid and gaseous phases at very low concentration.

Task 2.3 Testing corrosion of different materials at different conditions. Analysis of C-14 resulting from those experiments

Task 2.4 Synthesis and interpretation with focus on the inventory.

Activities reported for the 3rd year concentrated on Task 2.2. or 2.3

1.1.1 Reports by WP2 research units

Progress in Year 3 reported by research units is summarised:

1. Amec Foster Wheeler through RadioCarbon Dating Limited (RCD)-gas phase C-14, equipment tested, background analysis

2. SUBATECH/ARMINES: reports on the development in measurements of C-14 species formed during the corrosion of irradiated steels and Zircalloys for cement-based repositories in aqueous solution.
 - a. real leaching experiments
 - b. reduction of total activity using the ion exchange resins beta emitters for LSC and reduction of activity for AMS labs protocols
 - c. Removal of Cs137 using resins
 - d. Quantification of C-14 in liquid and gas phases: activity of carboxylic acids below detection limit of LSC (0.02 Bq/mL)

3. CIEMAT
 - a. Leaching experiments performed on samples of irradiated stainless steel from reactor upper of José Cabrera NPP. Gas chromatographic analysis performed for species of C (carboxylic acids)
 - b. LSC analysis of C-14 activities of leachates to be reported

4. ENRESA contribution: monitoring C-14 release from activated stainless steel under standard leaching test performed at El Cabril repository. Planned C-14 analysis using AMS technique available at Seville. The gaseous carbon products of leaching experiments can be trapped onto lime soda and then acidified to release CO₂ [Molnar *et al.*, 2013] or liquids can be purged with He stream to strip the CO₂ to be trapped and graphitized for subsequent AMS analysis.

5. JRC-ITU
 - a. Cladding Analyses of irradiated stainless steel cladding (15Ni-15Cr-Ti) from a fast reactor fuel provide estimates of C, O & N content: low for O and N content (on the order of 40 and 30 ppm, respectively) but high for C (C: 789 +/-55ppm).

- b. Planned leaching experiments (Dec2017): autoclave and glove box (molecular sieve to collect C for C-14 analysis. An issue of contamination between the hot cell and glove box.
6. KIT INE Similar to the experimental work in WP3 (Zircaloy cladding) dissolution of irradiated stainless steel was complete in autoclave. Liquid and gaseous products were sampled, diluted and C-14 was analysed. A total C-14 inventory of 2.7×10^5 Bq/g is higher by factor of 4 from the calculated values. The unknown nitrogen content of the steel might be a reason for the offset between experimental and modelled values [Schumann *et al.*, 2014].
7. NRG contribution: Testing background of the autoclave-RCD rig – LSC system. No additional contamination detected. Experiment performed on irradiated and irradiated samples. Activities measured in gaseous samples.
8. PSI:
 - a. Installation of thermal conductivity detection system for monitoring oxidation of alkanes and alkenes and separation of compound specific AMS C-14 analysis.
 - b. Time-dependent corrosion experiments under anoxic alkaline conditions show that hydrocarbons (reduced: alkanes, alkenes as well as oxidized: carboxylic acids, alcohols and aldehydes) are produced during anoxic corrosion of iron powders are produced at high rate in the first day of corrosion. Long term production is slower.
 - c. A reactor for anoxic NaOH leaching experiments was developed, background C-14 measurements show high level of $>0.1F14C$. The source of contamination needs to be established.
 - d. Separation of carboxylic acids using IC with high recovery obtained.

- e. C-14 activity of single compounds in leach solution after contact with activated steel: activity of collected fractions below limit of detection by LSC technique.
- f. C-14 content 38 ± 15 Bq/L of the leach fraction prior to chromatographic separation reproduced by the sum of the activities measured for fractions 21 ± 5 Bq/L.
- g. The AMS analyses are performed on CO₂. An oven required to oxidize gaseous organic compounds after GC separation is being developed

9. RWMC

- a. corrosion rates by H₂—after 2 years stable corrosion rates

10. SCK•CEN: accelerated corrosion tests in a designed glass chamber with Ca(OH)₂ as an electrolyte and electrodes allowing to apply potential, C-14 activity of liquid (leachate) is planned to be analysed by LSC

11. VTT

- a. 2 simulated groundwater solutions, CA85 and CA125, used for leaching experiments on un-irradiated Fe₃C and AISI316Ti steel powders.
- b. GC Analysed volatile organic alcohols from the leaching solutions such as methanol, ethanol and 1-propanol. For all of them except propanol, the quantification limit was too high.
- c. 2 samples from Loviisa nuclear reactor taken for future leaching experiments; based on N content the expected C-14 inventory after exposure is to be 2×10^3 Bq/g.

1.2 WP3 (Zircalloys)-Year 3

This WP3 aims at a better understanding of C-14 behaviour in disposed waste Zr fuel claddings: C-14 inventory (and origins), release from waste packages and speciation of released C-14 are under investigation. In order to achieve these objectives, the following tasks are performed:

Task 3.1&2 Review of analytical development

Task 3.3 C-14 inventory and C-14 release from leaching experiments and corrosion experiments in alkaline conditions close to future disposal conditions

During the third year of the CAST project, WP3 participants have essentially worked on Task 3.2, which is devoted to developing analytical methods for the characterisation of C-14 organic and inorganic molecules, techniques to measure C-14 inventory: LSC and potentially use of AMS for C-14 in inorganic/organic molecules.

To perform C-14 molecular speciation: GC, Infra-red, ESI-MS were applied

1.2.1 Reports by WP3 research units

8 research units are involved in this WP. Some tasks are overlapping with WP2:

1. ANDRA activities included
 - a. leaching experiments on irradiated Zircalloys
 - b. evaluation of analytical methods for AMS
 - c. estimates of the corrosion rate (CR) of Zr
 - d. The first results obtained on C-14 measurements (inventory + speciation)
 - e. The role of zirconia in C-14 release (IRF, dissolution rate)

2. Armines/Subatech

- a. IC for characterization of carboxylic acids combined with LSC (DL 0.04 Bq/mL). Lower activities with AMS at CEA Saclay.
- b. Ion exchange resins IER treatment to remove water soluble radionuclides: reduced activity, reduced beta emitters (for LSC)

3. CEA: The leaching test of Zy-4-type hulls started early in September 2016.

- a. $5 \text{ mgC/L}_{\text{solution}}$ measured in the organic form in the leachate sample $\Rightarrow 153 \text{ mgC/kg}_{\text{hull}}$.
- b. TOC is in the form of oxalic acid.
- c. Organic/inorganic C-14: 100% recovery of organic and pyrolysed C-14. Partition in leachates: total C-14 minus organic C-14.

4. ITU / JRC

- a. C, O, N were determined to estimate the potential for C-14 production in cladding
- b. Re-measured values C ($91 \pm 3 \text{ ppm}$) and N ($31 \pm 1 \text{ ppm}$) were used for modelling of the C-14 inventory. Leaching experiments and C-14 analysis undergoing

5. KIT INE: C-14 separation, quantification and speciation: acidic dissolution of irradiated Zircaloy-4 cladding a fuel rod segment irradiated in the Swiss Gösigen PWR

- a. Monte Carlo activation calculations to determine the inventory.
- b. Year 3: separation of C-14 and analysis.
- c. Improved removal of tritium prior to LSC.

- d. The measured C-14 inventory is reproduced 4×10^4 Bq/g Zy-4. Higher by a factor of 4 from the calculated values. Cs-137 is off by factor 117.
- e. Partition between organic/inorganic: (trapped after acid/oxidized CH₄). Most is inorganic.
- f. Most C-14 in liquid form (88%).

6. RATEN ICN

- a. Correlation between Co-60 and concentration of C-14—for gamma scanning
- b. Modelling of C-14 in CANDU Zy-4 tube: 1.776×10^4 Bq/g (0.481 μ Ci/g) of Zy-4.

7. RWMC

- a. Distribution of C-14 in cladding
- b. Measured 4 types of rings: with and without oxide & oxide itself. The average values on the order of 1.45×10^4 Bq/g, and 4.04×10^4 Bq/g for oxide layer.
- c. Oxygen as a target/source of C-14 in irradiation of cladding.
- d. But C-14 release after 6.5 years of leaching is on the order of 0.01%

8. SCK CEN

- a. Characterisation of material un-irradiated Zr-4
- b. Set up for corrosion experiments and C-14 speciation
- c. Corrosion tests planned as in WP2.

1.3 WP4 (SIERs)-Year 3

This WP focuses on speciation, source term, leaching, and gaseous release of C-14 from spent ion exchange resins

1.3.1 Reports by WP4 research units

1. CEA-EDF: 2 methods were used to measure C-14 in organic and mineral forms.
 - a. The combustion method returns the total of the C-14
 - b. acidic dissolution applied to one of the samples (Table 7 in relevant report) showed a problem in recovery of the total C-14.
 - c. Desorption experiments on sample EDF-B show a clear removal of C-14, however the estimated total activity obtained through desorption experiments (around 1500 Bq/g) and total C-14 activity measured after the combustion (2125 Bq/g see Table 8), evidence a loss of C-14 during desorption experiments.
2. ENEA
 - a. In order to estimate inventory of C-14 in SIERs samples of resins were oxidized: wet oxidation (addition of HNO₃ 65% + H₂O₂ 35% and digestion in microwave at 200°C). Problem of not full digestions can be a problem for C-14 estimates.
 - b. For speciation, stepped pyrolysis and/or complete oxidation is possible in a double chamber combustion cell
3. FZJ
 - a. Estimate of the total, organic and inorganic C-14 in the SIERs from BWR,
 - b. Release under different conditions, e.g. pH, radiolysis, degradation, rehydration, on C-14

- c. Speciation of C-14 in the gas phase using R-GC/GC-MS. (and AMS?)
 - d. HPLC used for liquid phase (counting LSC)
 - e. Efficiency for conversion of CO and methane to CO₂ is nearly 100%
 - f. Most of the C-14 in inorganic, only few % of the total C-14 was in organic form.
4. SKB: uptake of C-14 on the SIERs in operating reactor. Processing and storage having impact on the C-14 in disposed waste.
5. UJV:
- a. activity of wet SIERs at 40Bq/g lower then dry 2000Bq/g
 - b. release of C-14 from the resins –same amount of C-14 released independent of the resin activity
 - c. resins fixed in cement show no measurable release of C-14

1.4 WP5 (i-graphite)-Year 3

1.4.1 Reports by WP5 research

This WP focuses on the C-14 inventory and distribution in irradiated graphite (i-graphite).

Tasks involve:

- Measurements of the rate and speciation of C-14 release to solution and gas, and estimates of how efficient waste treatment approaches can be on C-14 releases from stored i-graphite.

1. IPNL

- a. Behaviour of implanted C-13 (simulating C-14) in graphite: effects of irradiation and temperature Irradiation of graphite and distribution of implanted C13

b. Ballistic versus electronic implantation

- i. Independent of the regime implanted C-13 is always stabilized into graphite
- ii. Initial graphite structure important for modification effect
- iii. Highly heterogeneous structure, but C-14 should be stabilized into sp² or sp³ structures as C-13

2. LEI

Completed task 5.1. Ongoing Task 5.2: estimates of C-14 inventories in graphite blocks and rings/sleeves of the Ignalina NPP.

3. RATEN ICN

- a. Irradiated sample of graphite from TRIGA blocks. long term leaching of this sample and estimates of C-14 release to liquid phase and organic/inorganic partition.
- b. C-14 activity of irradiated graphite (LSC): 451±16 Bq/g
- c. Undergoing experiment in anaerobic conditions, results were expected at the end of 2016.
- a. Present activities: inventory of C-14 in i-graphite, form and behavior as well as treatments prior to deposition.

4. ANDRA/ EDF

- a. Review of CARBOWASTE completed
- b. inventory of C-14 in i-graphite, form and behaviour as well as treatments prior to deposition

- i. semi-dynamic leaching to increase activities
 - ii. graphite in form of powder –increase surface (large samples)
5. ENEA: Experiments of the exfoliation-like process on the graphite from moderators of Latina NPP. Organic supported by mild sonication allows a stable and homogeneous dispersion of the graphite in a workable media. Effectiveness of removal of C-14 exfoliation-like process increases with time for all solvents (at least 10 hrs for RE% at ca. 15%).
 6. FZJ
 - a. Pre-selection of samples for leaching experiments: scanning for hot spots of activation products such Co-60.
 - b. Experiment in progress on porosity of graphite. This is relevant to the distribution of C-14 in i-graphite as well as to the release.
 7. CIEMAT
 - a. leaching experiments on irradiated powder graphite and on IGM graphite. i.e. a long term stable impermeable graphite matrix, possibly safer for final disposal.
 - b. Following preliminary tests experiments on powdered graphite were discarded and cylindrical graphite samples were chosen. 4 leaching periods on graphite cylinders and IGM have been completed using deionized water and granite-bentonite water.
 - c. Thermal treatment of graphite to release some of i-graphite's C-14 activity was tested at 3 temperatures and in O₂ flow. 4 experiments with irradiated graphite (1.4 10⁴ Bq/g) were designed to test effect of temperatures and gas flow on recovery. Higher T and oxygen flow have potential for reduced C-14 content in treated graphite.

- d. WP6 Safety i-graphite samples were collected from the inner and the outer surface of the graphite sleeve of the rod's edge

8. IFIN-HH

- a. AMS analysis at IFIN-HH (Romania) using 9MV (high activity) and 1MV facility for lower than 1F14C. Or dilution by a factor 100 with ultra-pure graphite (0 F14C).
- b. All samples but #6 (problematic analysis on 1MV AMS facility) show lower activity on the outside.
- c. The activities of C-14 and H-3 for each graphite samples were calculated based on LCS activity measurements, sample weight and retention yield (98% for H-3 and 80% for C-14).
- d. Comparison AMS (1MV) and LSC: factor of 10+ (See Table 2.8.4 and 2.8.2 in CAST Deliverable D5.9, repeated below)



Sample code	Distance from reactor core (cm)	Activity C-14 (Bq/g)	Standard deviation	Activity T (Bq/g)	Standard deviation	T/C activity ratio
D6.1.1	2	82518	6178.6	336124	31451.1	4.07
D6.1.2	20	59075	4284.3	183126	19502.9	3.10
D6.3	45	21532	1856.1	86342	8945.1	4.01
D6.1.2	80	9670	811.6	41330	4645.5	4.27
D6.2.2	98	3645	269.1	20132	1886.4	5.52
D5.1	102	3862	307.4	12687	1176.1	3.28
D5.2	138	1468	127.1	7868	862.3	5.35
D4.1	142	864	78.8	2867	279.0	3.31
D4.2	178	234	20.9	968	112.1	4.13
D3.1	182	117	10.3	521	62.5	4.43
D3.2	218	43	4.6	187	23.1	4.29
D2.1	222	21	2.4	112	13.4	5.22
D2.2	258	11.3	1.1	69	9.9	6.09
D1.2	300	6.9	1.3	39	6.7	5.74

Table 2.8.4 Specific activity of ¹⁴C and T in the samples collected from graphite column

Sample	Outer side		Inner side		Weighted Average ⁵
D1.2	0.64	0.04	6.85	0.45	0.69 ± 0.04
D2.2	2.39	0.24	2.71	0.08	2.68 ± 0.08
D2.1	3.73	0.3	8.48	0.43	5.29 ± 0.25
D3.2	16.04	0.27	28.74	0.72	17.56 ± 0.25
D3.1	20.28	0.95	29.13	1.61	22.55 ± 0.82
D4.2	29.92	0.37	29.33	6.42	29.92 ± 0.37
D4.1	14.88	0.22	27.95	0.13	24.67 ± 0.11
D5.2	19.07	0.59	40.16	0.31	35.69 ± 0.27
D5.1	Not	measured			
D6.2-2	51.38	5.37	48.43	1.06	48.54 ± 1.04
D6.2-1	64.76	0.02	48.62	0.72	64.74 ± 0.02
D6.1-2	Not	able	to	measured	
D6.1-1	Not	able	to	measured	

¹⁴C concentration results for samples collected from disc 5 and 6, even if they are presented in the table, are not trusted.

Table 2.8.2: ¹⁴C concentration (Bq/g) in graphite samples

- e. Corrected values obtained using 9MV AMS [CAST D5.14] show an agreement with values obtained by LSC technique (see Table 1, from CAST Deliverable D5.14; this report was delivered subsequently to D5.9 and includes any necessary updates).

Sample	14C (Bq/g)	Standard deviation
4.2	212.67	48.43
4.1	653.30	99.53
5.2	1381.52	241.84
5.1	3402.13	431.02
6.2.2	2649.83	721.84
6.2.1	7121.24	1830.65
6.1.2	51919.81	2405.52
6.1.1.	74850.73	2673.14

Table 1: ¹⁴C measured values by AMS in the thermal column.

The AMS system at the 9 MV tandem accelerator has the capacity of performing the depth profiling of the elemental concentration, in this case 14C in graphite. For this reason, samples were introduced in the AMS analyze as small square slices, 3 mm thick. Such a depth profiling is shown in Figure A6.

1.5 Minutes of GAM 2016

GAM meeting took place on Oct 26th and 27th 2016 in Luzern, Switzerland. The activities reported in WP 2, 3, 4&5 were presented as above.

1.5.1 WP6 Safety

Reported by Manuel Capouet from Ondraf/Niras:

WP6 technical meeting in March 2016 in the Nagra offices in Switzerland. Planned evaluation of data from WP2-5 and their impact on planned disposal conditions. Here sensitivity/uncertainty was an issue for an impact on the CAST outcome.

1.5.2 WP7 Dissemination

WP7 updated presented by Erika Neeft from Covra:

Web –well visited in 2016 (nearly 14000 hits, 8800 visitors)

Newsletters: 3 were planned, one published

Workshops targeting early stage researchers. 2 planned: early 2017 and one during the final conference Jan 2018.

1.6 Summary

Counting method of LSC has been successfully applied to determine inventories of C-14 in all the irradiated materials. The limit of detection of 0.04 Bq/ml is reached when product of corrosion or leaching, and specifically separated organic/inorganic molecules are analysed.

Technical development was reported by PSI (WP2). This research is dedicated to development of a direct AMS analysis (at University of Bern).

During 2016 additional AMS facilities (Saclay (France), Helsinki (Finland), Seville (Spain)) were contacted by the research units of CAST project for potential AMS analysis of low C-14 content samples. An interesting approach of a direct analysis on irradiated graphite was applied at the IFIN-HH (Romania) allowing for depth profiling of the reactor thermal rod.

In general, all experiments point to very low C-14 concentrations released from studied materials.

1.7 References

CAST Report on the release of C-14 from irradiated VVR-S graphite to solution and gas phase (D5.14)

Molnar, M., Hajdas, I., Janovics, R., Rinyu, L., Synal, H., Veres, M., and Wacker, L. 2013. C-14 analysis of groundwater down to the millilitre level. *Nuclear Instruments & Methods in Physics Research Section B-Beam Interactions With Materials and Atoms*, Vol. 294, 573-576. In English.

Schumann, D., Stowasser, T., Volmert, B., Gunther-Leopold, I., Linder, H., and Wieland, E. 2014. Determination of the (1)(4)C content in activated steel components from a neutron spallation source and a nuclear power plant. *Anal Chem*, Vol. 86, 5448-5454.

2 Review of Dr. Fraser King

2.1 Introduction

One of the tasks of the CAST Advisory Group is to review progress towards meeting the various project goals. This report represents the fourth formal review performed by the Advisory Group and considers material published in the minutes of the fourth CAST General Assembly Meeting (Scourse 2016) and the Year 3 annual progress reports from Work Packages (WP) 2, 3, 4, and 5 (Mibus et al. 2017, Necib et al. 2016, Comte et al. 2017, and Toulhoat et al. 2016, respectively), as well as a specific report on the progress of corrosion tests with Zr alloys (Druyts et al. 2016).

As previously, progress in each of the four work packages is presented in terms of the contributions to three key areas in the development of a ^{14}C source term, namely:

- Inventory - where, how much, and in what form is the ^{14}C present in the waste form?
- Release - what is the rate and mechanism of ^{14}C release, including the speciation?

Consideration is also given to the transport and reaction of ^{14}C once released, as this is important information for the safety assessment. However, participants have defined this area to be out-of-scope of the CAST project and it is included here only for completeness.

Information is required in each of these areas for the four different waste forms (steels, Zircalloys, spent ion-exchange resins (SIER), and irradiated graphite) in order to develop a robust source-term expression.

2.2 Review of 2016 GAM Minutes and Year 3 Work Package Annual Reports

2.2.1 Minutes of CAST General Assembly Meeting 4

The fourth CAST General Assembly Meeting was held in Luzern on 26-27 October 2016 (Scourse 2016) and was preceded by a day of meetings for the different work packages. The meeting minutes (Scourse 2016) represent an accurate description of the presentations and discussion that took place during the GAM.

Highlights from the GAM (as described in the minutes and from personal observation) include:

- WP2: a number of corrosion experiments on activated and non-activated samples have begun and more are planned. A total of eight partner organizations have either started experiments or have well-advanced plans.
- WP2: to assist in defining the inventory of ^{14}C , activation calculations are planned with confirmation by acid digestion and direct analysis. The major source of ^{14}C is considered to be activation of ^{14}N , although there will be a contribution from ^{17}O in the primary circuit water.
- WP2: initial results from leaching experiments suggest a rapid early release followed by a much slower long-term release, possibly below the sensitivity of the measurements. The question is whether this initial release should be interpreted as an IRF, or is it an artifact due to surface contamination? There is continuing uncertainty about whether the long-term release is congruent with the corrosion rate.
- WP3: there is an excellent database of long-term active and inactive corrosion rates for Zr alloys from measurements by RWMC for exposure time up to 6.5 years.
- WP3: RWMC were able to quantify the ^{14}C inventory in the oxide by selectively removing the oxide layer from the hulls. There is a higher amount in the oxide (on a volume or mass basis), but it only amounts to 7.5% of the total inventory compared with an earlier estimate of 20%. The oxide inventory is commonly assigned as the IRF in safety assessments on the assumption that it would (or could) be rapidly released. However, long-term leaching tests indicate that <0.01% of the total

inventory has been released after 6.5 years in alkaline media calling into question whether the oxide inventory should be treated as an IRF.

- WP3: as with the study of steels in WP2, there is still uncertainty over whether the long-term release is congruent.
- WP4: there is considerable variability in the ^{14}C content in SIERS from different locations within a plant and from plant-to-plant. However, analyses indicate that ^{14}C is present primarily in the inorganic form.
- WP4: the resins may age over time as a result of self-irradiation causing the individual particles to fracture and break, thus increasing the effective surface area and possibly resulting in loss of ^{14}C during storage and/or impacting the post-disposal leaching behaviour.
- WP4: of crucial importance is the fact that immobilizing SIERS in a cement matrix results in immeasurable release in relatively short term (a few months) laboratory experiments. Thus, the common safety assessment assumption of instantaneous or rapid release from SIERS could be highly conservative.
- WP5: release rates are being determined on powdered graphite and the relationship between the measured rates and those that can be expected from massive graphite structures is uncertain.
- WP6: plans are under way for a series of sensitivity analyses by various national programmes.
- WP6: the safety assessment participants have laboured under the disadvantage of not being able to influence the experimental WPs to focus on safety-relevant issues.
- WP7: a wide range of dissemination activities are planned or underway, including an impressive number of reports on the project website, the publically accessible website, newsletters, training courses, and workshops.

2.2.2 WP 2 Steels - Annual Report

The WP2 annual report describes progress made in 2016 on two tasks (Mibus et al. 2017):

- Task 2.2: Development of analytical methods for measuring ^{14}C speciation

- Task 2.3: Corrosion experiments and measurements of released ^{14}C

The areas in which the 2016 activities have contributed to an improved understanding of the overall ^{14}C source term for steels are highlighted in Figure 1 and are described in more detail below. A total of 11 organizations are participating in WP2 and the progress by each participant can be summarized as follows:

- Amec Foster Wheeler (AmecFW) are working with NRG (see below) on the development of sampling protocols for leaching tests on irradiated stainless steel specimens, plus one blank unirradiated test sample. Experiments are under way but exposure periods during the course of the CAST project will be limited to 1 week to 3 months duration.
- Armines/Subatech have devoted a lot of effort towards the decontamination of leachates in order to be able to reliably measure ^{14}C using liquid scintillation counting (LSC). However, it was then found that LSC is insufficiently sensitive to determine the small amounts of ^{14}C released during the corrosion of steels or Zr alloys in alkaline conditions and, instead, attention will be focussed on the more-sensitive accelerator mass spectrometry (AMS) method.
- CIEMAT have developed chromatography-based separation methods for inorganic and organics gases and for dissolved carboxylic acids. Active leaching experiments are under way using irradiated stainless steel in both alkaline and acidic solutions under aerobic conditions.. Exposures are planned to last up to 360 days, with gas and leachate samples taken (but not yet analysed) for exposure periods up to 90 days.
- ENRESA have developed a protocol for leaching studies and have set up a dummy test. The more-sensitive AMS technique will eventually be used for analyses.
- JRC-ITU are still in the development stage, but are planning irradiated and parallel inactive exposures of a Ti-containing stainless steel. However, it would seem likely that a maximum exposure period of only 90 days will be possible within the CAST project itself. Unirradiated material is available and analyses are under way to determine the chemistry.



- KIT-INE are focussed on calculating the ^{14}C inventory and comparing it with the inventory measured following acid digestion of irradiated samples. Based on results to date, the measured inventory for a sample of stainless steel taken from the plenum spring is within a factor of three of the calculated value. The measured value is higher, suggesting that either: (i) the N content assumed for the calculations was too low (by a factor of 3), (ii) the neutron fluence assumed was too low, (iii) there is another significant source of ^{14}C , or (iv) the sample was contaminated in some way.
- NRG are working in collaboration with AmecFW and are conducting two active and one blank (inactive) leaching test on stainless steel samples in anoxic alkaline solution at ambient hot cell temperature. Experiments are under way with a planned maximum exposure period of 3 months. Gas samples have been taken after periods of 1, 3, and 6 weeks but, as of the date of the annual report, no results were yet available. Both ^{14}C and ^{60}Co will be determined, with the latter used as a measure of the rate of corrosion. However, it is not clear why ^{60}Co should be released to solution congruently with the corrosion of the steel so if such a measure is to be used as a surrogate for corrosion then this relationship will need to be justified.
- PSI have continued both analytical development work and corrosion leaching-type experiments. A gas chromatography-mass spectrometry (GC-MS) method has been developed for separating and subsequently quantifying gaseous ^{14}C species by AMS. Equipment for conducting active leaching experiments has been commissioned. Inactive experiments have been conducted using iron powder, with the focus on whether the nature of the alkaline solution (NaOH or $\text{Ca}(\text{OH})_2$) makes any difference to the speciation of released carbon. In earlier work using artificial cement pore water solution ($\text{Ca}(\text{OH})_2$), both oxidised and reduced forms of organic carbon were observed, with a large release during the first day of leaching followed by much slower (immeasurable) leaching at longer times. They observed higher (30-fold) amounts of alkanes in NaOH solution and suggested that this indicated a higher corrosion rate in this solution compared with $\text{Ca}(\text{OH})_2$ despite a similar pH in both tests. The amount of organic acids released was comparable and are thought to be an artifact of the experiment rather than a product of the corrosion of the Fe

powder. As for $\text{Ca}(\text{OH})_2$ solution, a large initial release of both the hydrocarbons and organic acids was observed on the first day of exposure followed by an almost immeasurably slow long-term release. It is tempting to interpret this initial rapid release as an IRF but it could also be an experimental artifact of the preparation or configuration of the sample. In addition, it is not clear why exposure to a NaOH solution of the same pH should result in a corrosion rate 30 times higher than in $\text{Ca}(\text{OH})_2$ solution.

- RWMC have continued long-term corrosion experiments of inactive stainless steel. The rate decreases with time in both neutral and alkaline solution.
- SCK-CEN have been unable to demonstrate that ^{60}Co is congruently released due to problems with precipitation in solution and sampling. However, attempts to measure ^{14}C in solution using LSC were successful.
- VTT have focussed on the leaching of iron carbide and stainless steel powder samples in simulated groundwater solutions at neutral and alkaline pH. In inactive experiments, more organic than inorganic carbon is found in all cases, although the total amount of carbon released is higher for the iron carbide powder than for the stainless steel. There is no effect of the leachate pH on the amount of dissolved organic carbon, but more inorganic carbon is released to solution at the higher pH.

After the previous years preparations, most of the participants are now at the stage of conducting corrosion and/or leaching experiments. Both active and inactive samples are being used. A number of challenges remain, however, including:

- Distinguishing experimental artifacts (for example, due to surface contamination) from instant release. In addition, the long-term release of ^{14}C (or inactive carbon) seems to be so slow that it will be difficult to derive a release rate.
- There are relatively few measurements of the corrosion rate either planned or under way and it is not clear that the release of ^{60}Co can be used as a marker for corrosion. Therefore, it will be challenging to demonstrate whether ^{14}C is released congruently or not.

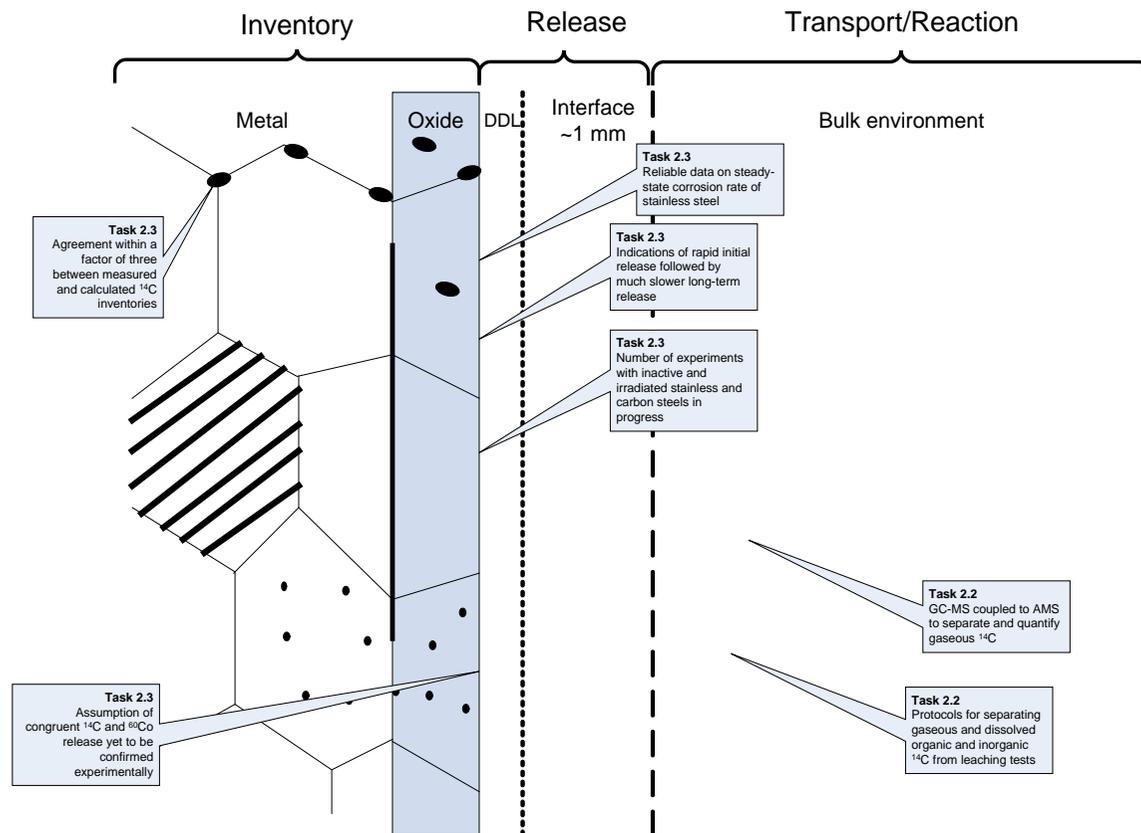


Figure 1: Summary of Progress in Developing a ¹⁴C Source Term for Steels in 2016.

2.2.3 WP 3 Zircaloy – Annual Report

The WP3 annual report describes progress made in 2016 on two tasks (Necib et al. 2016):

- Task 3.2: Development of analytical methods for measuring ^{14}C speciation
- Task 3.3: Characterization of ^{14}C released from irradiated zirconium fuel clad wastes

As previously, the annual report contains a number of useful tables that summarise the contributions of each participant to the various tasks. These tables summarize the irradiated and unirradiated samples being tested, the analytical techniques to be used or under development, the leaching conditions for both irradiated and unirradiated samples, and the techniques and existing data on corrosion rates. In addition to the annual report, deliverable D3.12 describing progress on various corrosion tests was also reviewed (Druyts et al. 2016).

The areas in which the 2016 activities have contributed to an improved understanding of the overall ^{14}C source term for Zr are highlighted in Figure 2 and described in more detail below.

A total of seven organizations are conducting experimental studies in WP2, with Andra acting as the co-ordinator. Highlights from the annual report and the summary of Druyts et al. (2016) include:

- Armines/Subatech worked in collaboration with various CEA laboratories to develop techniques for decontaminating and characterizing liquid leachate samples. Leachates from active experiments exhibit too high a radiation field to be acceptable for measurement of ^{14}C in AMS facilities, primarily due to the presence of ^{137}Cs , ^{125}Sb , $^{106}\text{Rh}/^{106}\text{Ru}$, as well as minor amounts of other radionuclides. Various commercial reagents were used for the decontamination process. Ion chromatography was used to separate inorganic and organic ionic species from an active decontaminated leachate but the resulting amounts were below the LSC detection limit.

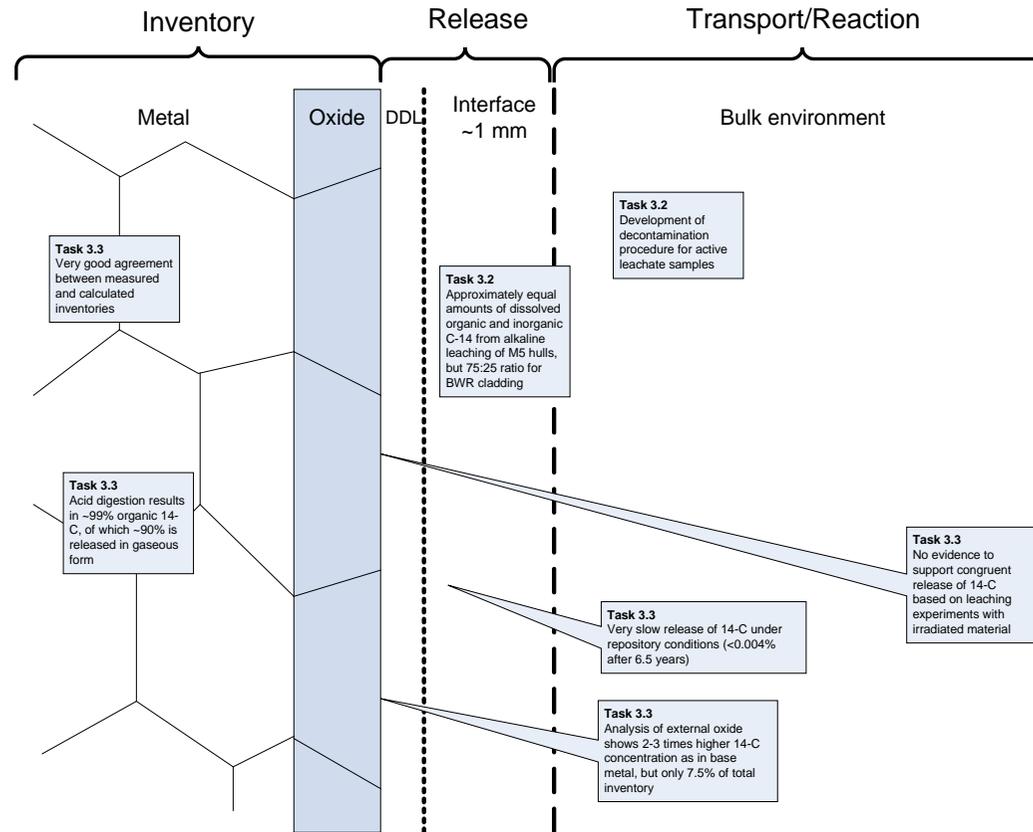


Figure 2: Summary of Progress in Developing a ¹⁴C Source Term for Zirconium Alloys in 2016.



- CEA are participating in both Tasks 3.2 and 3.3 and reported progress on both during 2016. Leaching tests were started using both M5 and Zy-4 hull samples supplied by Areva. Although repeated acid rinsings had been applied at La Hague and, to reduce the activity, upon receipt at CEA, Raman analyses suggested that the oxide structure was similar to that formed during in-reactor surface, from which it was concluded that the pre-treatment had not significantly altered the samples. Leaching tests in anoxic pH 12 NaOH solutions were started and samples from a 14-day exposure of the M5 hulls were analysed. Analysis of the leachate indicated that there was slightly more dissolved inorganic ^{14}C (54%) than organic (46%), although a smaller fraction compared to the distribution of total carbon (69% total inorganic carbon vs. 31% total organic carbon). The ^{14}C fractions represents approximately 0.0002-0.0003% of the total C.
- ITU/JR are commissioning equipment for an active leaching experiment. In addition, commercially available, unirradiated Zy-4 samples (not archive material) have been analysed to determine the concentrations of precursor C and N species in the material (91 wppm and 31 wppm, respectively).
- KIT INE successfully measured the inventory and speciation of ^{14}C released from acid digestion of irradiated Zy-4 cladding. The total ^{14}C inventory was found to be reproducible between samples and was determined to be $3.7 \pm 0.4 \times 10^4$ Bq/g based on results from six specimens. Of the ^{14}C released during acid digestion, more than 99% was organic, approximately 90% of which came off as gas with only ~10% remaining in the leachate. Because the organic was in the form of hydrocarbon, it was suggested that there should be no dependency on speciation on pH and that, therefore, the speciation resulting from acid digestion should be appropriate for repository conditions. Furthermore, activation calculations based on the ^{14}N content and irradiation history resulted in a theoretical inventory of 3.2×10^4 Bq/g, in good agreement with the measured value.
- RATEN/ICN started leaching experiments with irradiated CANDU Zy-4 cladding samples and with similar unirradiated material. A feature of certain types of



CANDU fuel is the use of a colloidal graphite layer known by the commercial name CANLUB on the inner surface of the cladding. Activation calculations, however, suggested that this layer is not a significant source of ^{14}C compared to the N content of the alloy itself.

- RWMC measured the ^{14}C content of both the external oxide and base metal of BWR cladding, the oxide having been removed mechanically. Samples were dissolved in acid and the released ^{14}C oxidized to CO_2 , trapped and then analysed by LSC. Although the ^{14}C concentration in the oxide is 2-3 times higher than in the base metal, the relative thicknesses mean that only 7.5% of the total inventory is in the oxide. This is important as the ^{14}C content of the oxide is commonly assumed to be released instantaneously in safety assessments and an IRF of 20% has been previously assumed. These new measurements allow a relaxation of the previous conservatism. PWR cladding samples exhibit thicker oxides and, as a consequence, a higher fraction of the total inventory. Because of the higher ^{14}C content of the oxide, it was concluded that ^{17}O activation is a significant source of ^{14}C based on the assumption that the N contents of the metal and oxide are similar. However, it is not clear whether the assumption of similar N contents in the alloy and oxide is based on measurement. Since the oxide grows during service, surely an alternative explanation for the higher ^{14}C content in the oxide is some type of partitioning mechanism rather than production by activation of ^{17}O . Leaching tests in anoxic pH 12.5 NaOH solution with different irradiated BWR and PWR cladding samples have now been completed up to times of 6.5 years. A BWR sample with only external oxide leached for 6.5 years had released only 0.004% of the total ^{14}C inventory, with 14% released in the gas phase and the remainder in the aqueous phase. Of the dissolved ^{14}C , approximately 75% was organic and the remainder inorganic. Based on the assumption of congruent ^{14}C release due to corrosion of the Zy substrate, it was concluded that the large majority (>96%) of the ^{14}C was released by the oxide rather than by corrosion of the matrix. The small fractional release of 0.004% after 6.5 years can be compared with the oxide inventory of 7.5%, from

which it is apparent that it may not be appropriate to assume that the ^{14}C in the oxide is released “instantly”.

- SCK-CEN have performed metallography on unirradiated Zy-4. Because of the low impurity content, it is difficult to identify the distribution of the precursor N species (and even more difficult to determine the chemical form), although a phase consistent with ZrN (but also consistent with ZrC, ZrH₂, as well as other phases) was identified. Static leaching and accelerated electrochemical dissolution tests were commissioned, the purpose of the latter simply being to release a measurable amount of ^{14}C under alkaline conditions representative of the repository environment (as an alternative to the acid digestion technique).

Significant progress has been made in the previous year and common trends have been observed between different partners. However, a number of challenges remain. Highlights of the past years work include:

- Based on the preliminary data from CEA, it remains to be seen whether the speciation of ^{14}C is the same or different from that of the total carbon. Clearly, it would be advantageous if the speciation was the same as it would be easier to separate and quantify the total C as opposed to the much smaller ^{14}C component.
- There is some consistency in the speciation of dissolved ^{14}C from alkaline leachates from work at CEA and RWMC, although the predominantly gaseous release of almost entirely organic ^{14}C in acid-digestion tests at KIT INE calls into question whether this approach can be used to determine the speciation under repository conditions.
- The RWMC measurements on the distribution of ^{14}C between oxide and base metal provide a firmer basis for estimating the IRF, although leaching tests up to 6.5-year duration show only minimal release. Thus, the ^{14}C contained in the oxide is not “instantly” released and some thought could be given to reducing the IRF even further.
- Currently, there is no evidence to support the assumption of congruent release.

2.2.4 WP 4 Ion-exchange Resins – Annual Report

Of the four tasks in WP4, those for which progress was reported in the annual report (Comte et al. 2017) were:

- Task 4.2: ^{14}C inventory and speciation in SIERS
- Task 4.3: ^{14}C release from SIERS and its speciation

A total of six partners contributed to WP4 in the reporting period. The areas in which the 2016 activities have contributed to an improved understanding of the ^{14}C source term for SIERS are highlighted in Figure 3.

Highlights from the contributions from the individual partners include:

- CEA-EDF studied the desorption of ^{14}C by sequentially leaching resins in aliquots of 10^{-3} mol/L LiOH, 1 mol/L LiOH, and distilled H_2O . Interestingly, little organic or inorganic ^{14}C was released after two leachings in dilute LiOH solution, but leaching in concentrated LiOH solution a total of six times resulted in complete desorption. This apparent pH (or ionic strength) effect could have implications for the behaviour in the repository.
- ENEA have focussed on characterising the inventory in SIERS and ageing effects that could impact the ability of the resins to retain ^{14}C during storage. Different types of degradation behaviour were observed on aged resins, including fractured resin beads and surface corrosion. Anionic resins were found to degrade more than cationic resin.
- FZJ are participating in all aspects of WP4 and reported preliminary results on a number of topics. Largely inorganic ^{14}C has been reported.
- RATEN ICN are determining the organic/inorganic ^{14}C fractions in spent CANDU resins and the rate of desorption under alkaline conditions. Much of the effort during the reporting period was dedicated to technique development, including decontamination procedures and commissioning of a catalytic furnace.

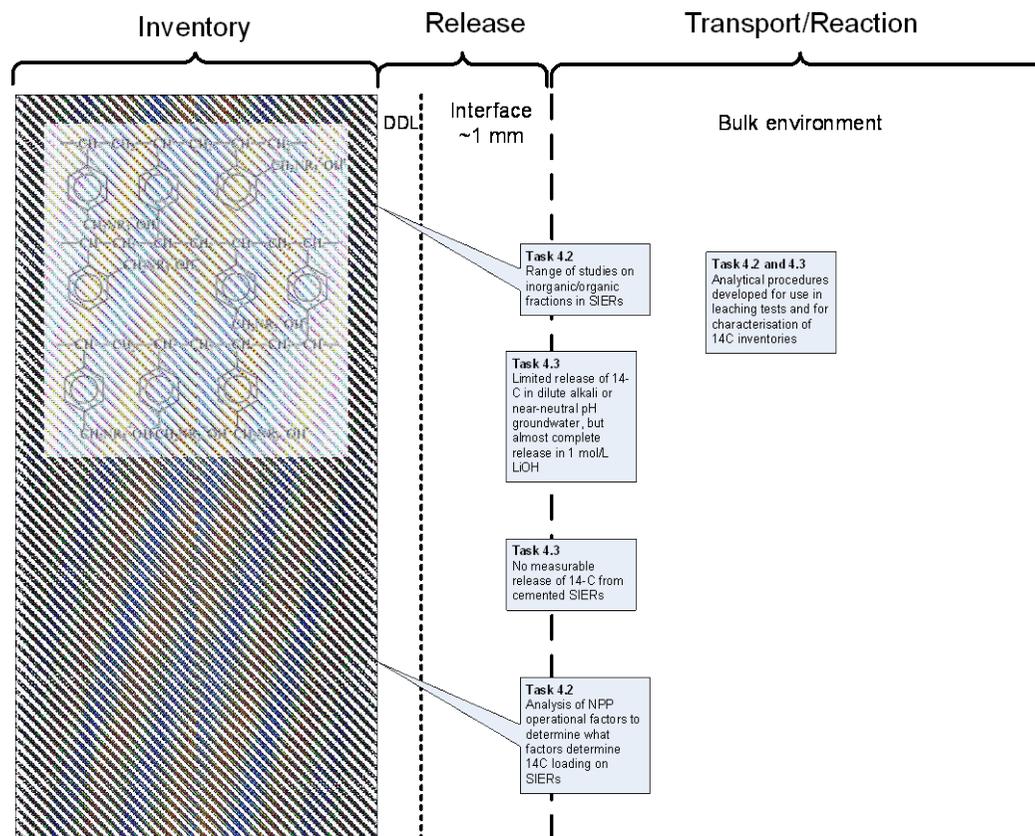


Figure 3: Summary of Progress in Developing a ¹⁴C Source Term for Spent Ion-exchange Resins in 2016.



- SKB briefly reported on ongoing efforts to model historical and future ^{14}C production. Interestingly, only a small fraction of the ^{14}C that is generated is subsequently captured by the IERs (and, therefore, require storage and disposal), with much of it being dispersed from the plant during operation.
- ÚJV focussed on the desorption/leaching of ^{14}C from resins under alkaline conditions and when immobilised in a cement matrix. Only a few % of the inventory was desorbed over a period of approximately 30 days when the resins were contacted by either pH 12.7 NaOH solution or by a near-neutral dilute synthetic groundwater solution. Perhaps more importantly, no release at all was detected over a 90-day period when the resins were immobilised in a cement matrix. This latter result could have important implications for the treatment of the ^{14}C source-term in safety assessments.

Significant findings from the work in WP3 in the reporting period include:

- Confirmation of the variation in speciation and loading of SIERs depending upon the reactor type and location of the exchange bed within the plant.
- Evidence for the degradation (ageing) of the resins during storage, due in part to gamma irradiation, which could impact the release of ^{14}C during storage prior to disposal.
- ^{14}C is not readily released from SIERs when contacted by dilute alkali solution (<0.01 mol/L) or near-neutral pH solution, but is released by contact with 1 mol/L OH^- .
- Immobilisation of the SIERs in a cement matrix substantially reduces the release of ^{14}C from the waste form which could have important implications for safety assessment.

2.2.5 WP 5 Graphite – Annual Report

Of the five tasks in WP 5, those for which progress was reported in the annual report (Toulhoat et al. 2016) were:

- Task 5.2: Characterisation of the ^{14}C inventory in i-graphite
- Task 5.3: Measurement of release of ^{14}C inventory from i-graphite
- Task 5.4: New waste forms and ^{14}C decontamination techniques for i-graphite

A total of eight participants reported progress during the year. The areas in which the 2016 activities have contributed to an improved understanding of the ^{14}C source term for i-graphite are highlighted in Figure 4.

Significant highlights and achievements from the various partners in 2016 include:

- CNRS/IPNL reported the results of detailed studies of the effects of irradiation and temperature on the stability, distribution, and retention of ^{14}C in i-graphite. Unlike the metallic wasteforms considered in WP2 and WP3, the primary source of ^{14}C in i-graphite is the activation of ^{13}C not of ^{14}N . The stability of the ^{14}C within graphite structure depends not only on the irradiation and thermal history but also on the original degree of ordering in the matrix. Regardless, except for near-surface species, the results suggest that ^{14}C should be stabilised within the graphite structure as a result of in-reactor conditions.
- LEI are applying the outcomes of the earlier CARBOWASTE project to the inventory of ^{14}C for the RBMK reactor design.
- RATEN ICN are focussing on the development of a source term for the i-graphite from the TRIGA reactor thermal column. Leaching experiments were designed during the reporting period.
- Andra/EDF issued a deliverable on the review of the speciation of ^{14}C released from French i-graphites conducted in Year 2 of the CAST project.

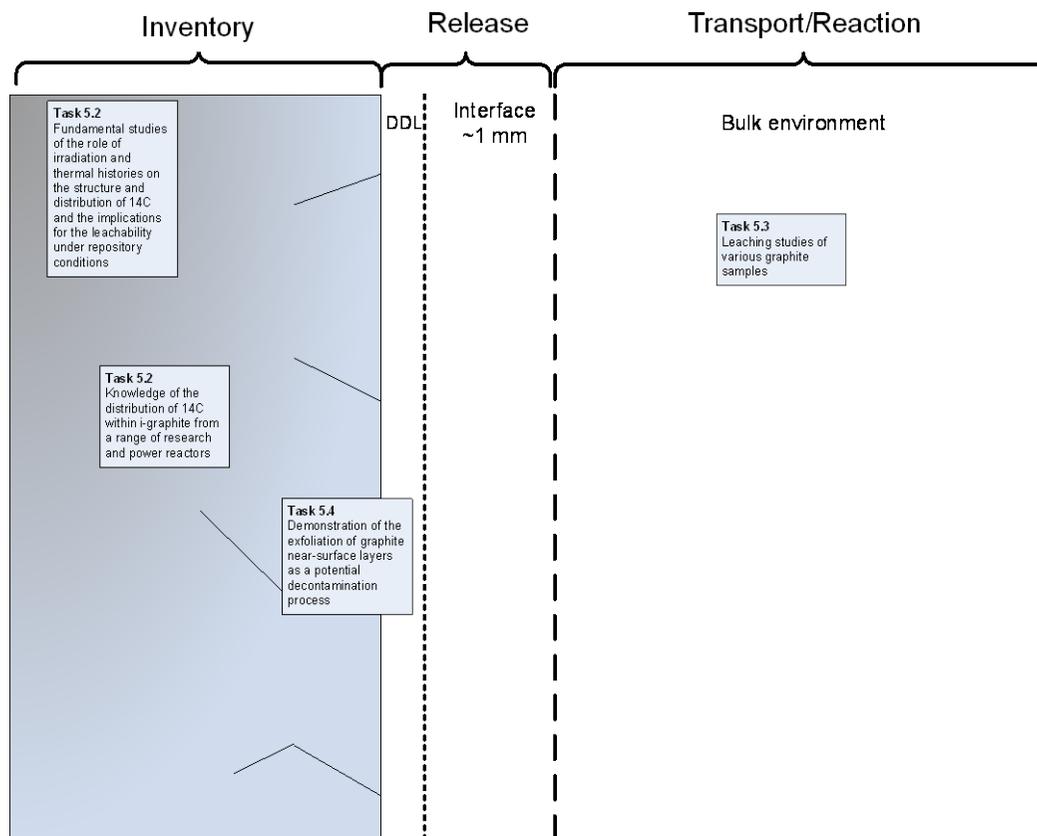


Figure 4: Summary of Progress in Developing a ^{14}C Source Term for Irradiated Graphite in 2016.

- ENEA are developing a method for pre-treating i-graphite by sonification in organic solvents to exfoliate the graphite, thus exposing closed pores and other occluded regions within the waste and aiding in the recovery of ^{14}C . During the reporting period, the impact of different solvents and sonification conditions on the ^{14}C recovery efficiency were investigated. At this point, the recovery efficiency is modest (up to 15%) but work continues to optimise the process.
- FZJ plan to conduct leaching experiments on i-graphite samples from a research reactor. Prior to leaching, the samples have been characterised by gamma radiography which revealed hotspots thought to be due to accumulations of ^{14}C . Since the precursor ^{13}C would be expected to be uniformly distributed, this suggests that much of the ^{14}C is produced by activation of ^{14}N impurity located in pores and defects within the graphite structure. Therefore, knowledge of the graphite pore distribution is important.
- CIEMAT changed course slightly during the reporting period and moved away from leaching powdered graphite to leaching monolithic graphite samples instead as being more representative of actual waste forms. Leaching studies up to 90 days have been completed and analyses are in progress.
- IFIN-HH are examining the distribution of ^{14}C in a thermal column from a decommissioned Hungarian research reactor. The distribution of ^{14}C and ^3H as a function of distance from the reactor core were determined.

There are different types of work being conducted in WP5, ranging from fundamental studies of the effects of irradiation and thermal history on the location and stability of ^{14}C in graphite structures, to the development of techniques for pre-treating i-graphite, through to the application of existing information from the CARBOWASTE project to various research reactor components and other types of power reactor. This WP is a good example of dissemination of knowledge among the various EU partners.

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