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REPORT ON THE RELEASE OF C-14 FROM IRRADIATED VVR-S GRAPHITE TO SOLUTION AND GAS PHASE (D5.14)

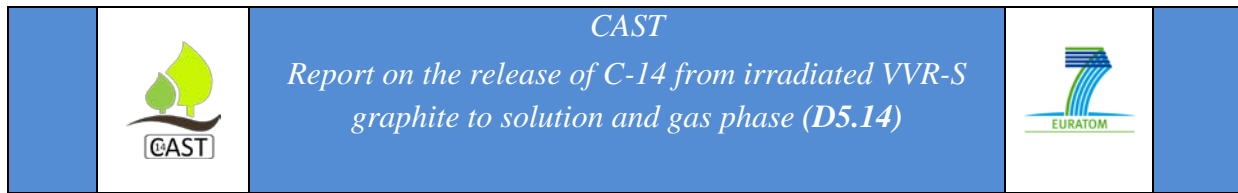
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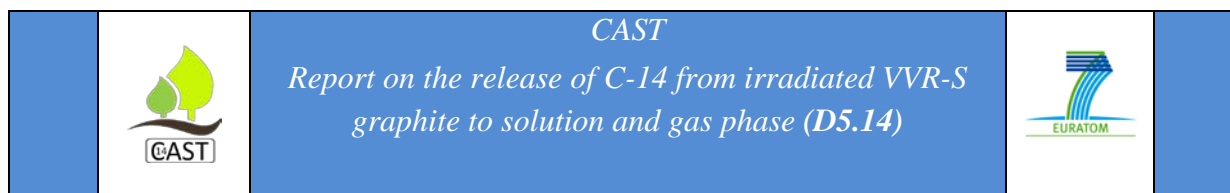
Document title
REPORT ON THE RELEASE OF C-14 FROM IRRADIATED VVR-S GRAPHITE TO SOLUTION AND GAS PHASE (D5.14)

Executive Summary

The main objective of the IFIN-HH in Task 5.3 of the Work Package 5 was to measure the release rate of C-14 (and tritium) into gas and solution phase from irradiated graphite (intact and crushed samples from VVR-S reactor will be used) in order to improve the understanding of the mechanism of release of C-14 from irradiated graphite in contact with aqueous solutions. An experimental apparatus has been designed and manufactured to measure the total release of C-14 (and H-3) to gas phase from irradiated graphite from the thermal column of the VVR-S Reactor.

Irradiated graphite, dismantled from the thermal column of VVR-S Reactor, contains different radioisotopes as activation products, including carbon-14 (C-14) and tritium (H-3). In repository-relevant conditions, this graphite could be a source of gases containing C-14 and H-3.

In the experiments performed by IFIN-HH we have investigated the release of C-14 and H-3 from two intact samples of irradiated graphite which have been cut from the thermal column disc located near the reactor core. The samples were collected after the reactor was shut down and the spent fuels assemblies were transferred to the final repository. The graphite samples were submerged in a pH 10 solution of sodium hydroxide (NaOH) during the experiment to replicate the alkaline conditions that are likely to appear in an underground repository once grouted wastes became saturated with water. The C-14 and H-3 contents in submerged solution were measured at different times over a long period of time (12 month). The study also investigated the long term release of C-14 and H-3 to the gas phase.



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1 Introduction

Generally, C-14 is produced in all types of reactors, mainly through neutron-induced reactions with isotopes of carbon, nitrogen and oxygen. They are present in the fuel, fuel cladding, coolant, moderator and structural materials of the reactor. The majority of C-14 in irradiated graphite arises through the irradiation of nitrogen-14 (n,p) reaction, while carbon-13 (n, γ) reaction is the second greatest contributor. Irradiated graphite contains a range of activation products but, from the point of view of geological disposal, another radionuclide which has to be taken into account is tritium (H-3 or T), produced through neutron induced reaction $\text{Li}(n,\alpha)\text{T}$.

C-14 and H-3 is located in the thermal column of the VVR-S Reactor. The mass of irradiated graphite of the thermal column is estimated to be 5.3 metric tons.

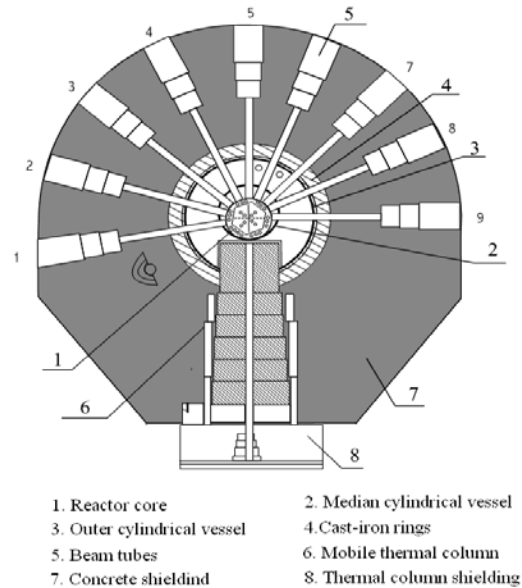
The key radioactive gases which are expected to be released from irradiated graphite waste in a cementitious matrix are compounds of C-14 and H-3 ($^{14}\text{CO}_2$, ^{14}CO , $^{14}\text{CH}_4$ and HT).

C-14 and H-3 are both beta emitters (half-lives 5730 and 12.3 years respectively). As a consequence of the relatively short half-life of H-3 the inventory of H-3 will be reduced to level over period of couple of hundreds of years and therefore the release H-3 containing gases is important for operational safety of the disposal facility, which is not the case for the release of C-14 containing gases from irradiated graphite.

Irradiated graphite itself is not reactive under geologic repository conditions. Consequently the C-14 atoms in the graphite lattice are expected to be released very slowly, if at all, but the C-14 atoms formed from nitrogen-14 impurities trapped within closed gaps or from nitrogen-14 atoms incorporated in pore surface are more reactive compared with the carbon atoms in the graphite lattice. The mechanism of the release of C-14 and tritium from irradiated graphite under repository conditions is the subject of ongoing international research; it is generally accepted that the majority of C-14 in irradiated graphite is immobile under conditions relevant to geological disposal.

The goal of these experiments was to measure the leaching of C-14 and tritium into solution and gas phase from irradiated graphite of the thermal column of VVR-S Reactor.

A horizontal cross section of the VVR-S Reactor core is presented in Figure 1.



VVR-S REACTOR HORIZONTAL CROSS-SECTION

Figure 1: Horizontal cut-view through the VVR-S Reactor

The VVR-S Reactor from IFIN-HH was a research reactor with a maximum thermal power of 2 MW moderately cooled and reflected with distilled water, and fueled with enriched uranium (10% in the beginning and 36% subsequently). It was commissioned in 1957 and dedicated to nuclear physics research and radioisotopes production. Until 1984 the reactor was operated with nuclear fuel type EK-10 (10% enrichment) and from 1984, this fuel was replaced by S-36 (36% enrichment). The reactor was operational until 1997 when the reactor was definitively shut-down and now is under decommissioning. During 40 years of operation, the VVR-S Reactor produced 9.59 GWd. The maximum flow of the thermal neutrons was $2E13 \text{ n/cm}^2\text{s}$.

2 Materials and Methods

2.1 VVR-S Reactor thermal column

The thermal column of the VVR-S Reactor (Figure 2) is made out of 6 graphite discs placed on a mobile truck. Graphite discs are installed into a 20 mm wall thickness aluminum cylinder. Initially, the thermal column was provided with a cooling system that axially penetrated the graphite plate connected to the water-cooling system. On the basis of subsequent operational experience, it was concluded that this system was no longer necessary; it was not used any further. Therefore, the horizontal tubes of the cooling system were filled with nuclear grade graphite rods of the same type as the discs of the thermal column. Samples have been collected from these graphite rods and the surrounding graphite sleeve of the central graphite rod (Figure3)



Figure 2. Thermal column



Figure 3. Thermal column disc no 6

2.2 Irradiated graphite samples

The irradiated graphite investigated at IFIN-HH has been taken from nuclear grade graphite rods which were used to fill the horizontal tubes of the cooling system of the thermal column of the VVR-S Reactor

For the leaching test the graphite sampling was performed from the small graphite rod (diameter=4.4 cm, length=90 cm) located around the central graphite rod of the disc no 6, which is near the reactor core. One piece was cut from rod's edge near the reactor vessel (**Sample 6.1**) and the other piece from the opposite part of the rod (**Sample 6.2**). From each piece collected from the graphite rod, two separate samples have been taken (e.g. Sample 6.1-1, Sample 6.2-1)

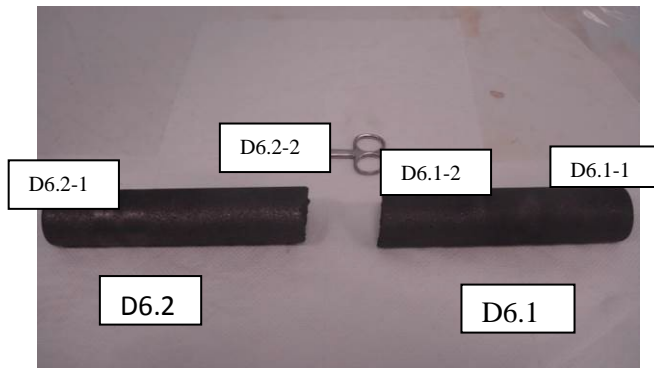


Figure 4: Irradiated graphite samples collected from the small rod of disc no 6

The mass and dimensions of the two irradiated graphite pieces which have been used for the leaching test are presented in Table 1. In the table, based on our previous results, we also present the estimation of C-14 and H-3 total activity for each sample. (WP5 2nd Annual Progress Report – Year 2 (D5.9) (2015)) [1].

Table 1 Main characteristic of the i-graphite samples

Sample code	Dimension: diameter x length (mm)	Mass (g)	C-14 mean specific activity (Bq/g)	T –mean specific activity (Bq/g)	C-14 inventory (kBq)	T inventory (kBq)
6.1.1	44x45	83.37	74266	261512	6191.5	21802.2
6.2.1	44x45	79.88	4237	25342	338.4	2024.3

2.3 *Leaching experimental methodology in anaerobic conditions*

The graphite samples were placed in two 1 litre flasks with thread screw caps with 4 outlets/inlets and were submerged in solution of 0.1 M of sodium hydroxide. All outlets and inlets of the reaction vessel were valved to allow sealing. The 700 ml of solution added to each flask ensured that the graphite samples were completely submerged. (Figure 5)



Figure 5: Graphite sample submerged in sodium hydroxide solution placed on a stirrer

To evaluate C-14 and H-3 long-term release in solution, under conditions relevant to a cement-based geological disposal facility, the leaching test was performed under nitrogen atmosphere in closed vessels. The flask was placed on a magnetic stirrer and adapted to allow the replacement of the air with nitrogen in order to ensure anoxic conditions.

The sampling was performed at different time periods: the 1st, 7th, 14th, 21st, 28th, 42nd days, 3rd, 6th, 9th and 12th months. After each sampling, the quantity of the leachant removed was replaced with a fresh one.

The amount of C-14 and H-3 released in the liquid phase was measured by liquid scintillation counting using a TRICARB 3110 TR counter, allowing measurements in low levels.

2.4 Release of C-14 and H-3 from i-graphite to gas phase. Experimental methodology

Intact samples from VVR-S irradiated graphite have been cut from the small graphite rod (diameter=4.4 cm, length=90 cm) located around the central graphite rod of disc no 6, which is near the reactor core. The mass of the graphite samples and the calculations of the total activity of C-14 and H-3, according to our previous results, are presented in Table 2:

Table 2 Irradiated graphite samples characteristics

Graphite sample	No of discs	Mass (g)	C-14 inventory (Bq)	H-3 inventory (Bq)
6.1.2	2	2.548	179861	593671
6.2.1	2	1.982		
6.2.2	2	2.109		

The irradiated graphite samples were sealed in a glass vessel provided with 4 outlets and inlets (figure no 6).



Figure 6: Graphite samples sealed in a glass vessel

Forty days after the samples were sealed, the vessel was connected to a catalytic oxidizer furnace that contains a CuO catalyzer inside. The C-14 and H-3 compounds released in the gas phase were oxidized in a low oxygen flow rate atmosphere and trapped separately in 5 vials (Figures 7 and 8). The measurement continued for one hour after the connection with the furnace was opened. At the end of the measurement the vessel containing the graphite samples was resealed and the experiment was repeated after 3 months.

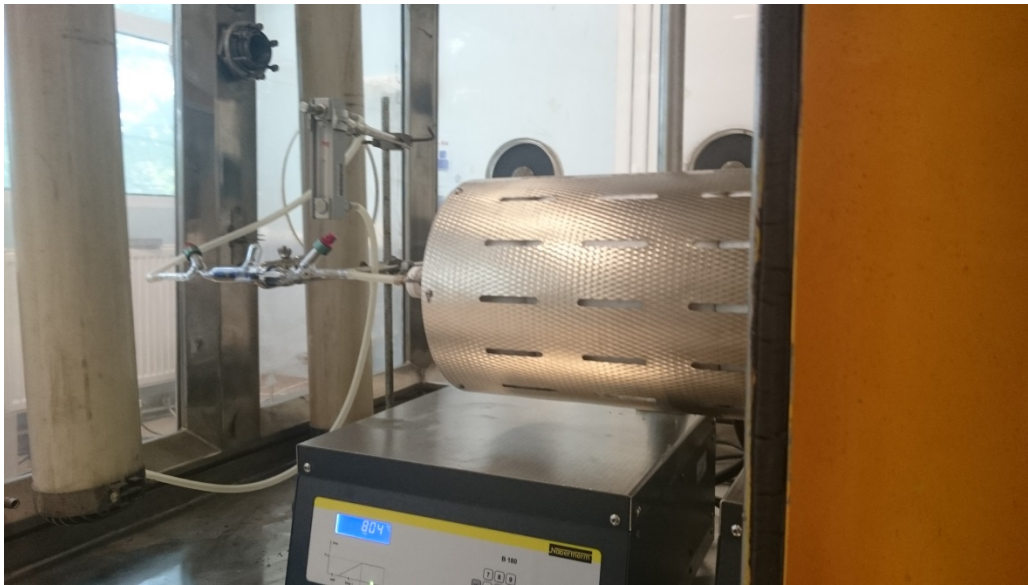


Figure 7: Apparatus for C-14 and H-3 determination release in gas phase

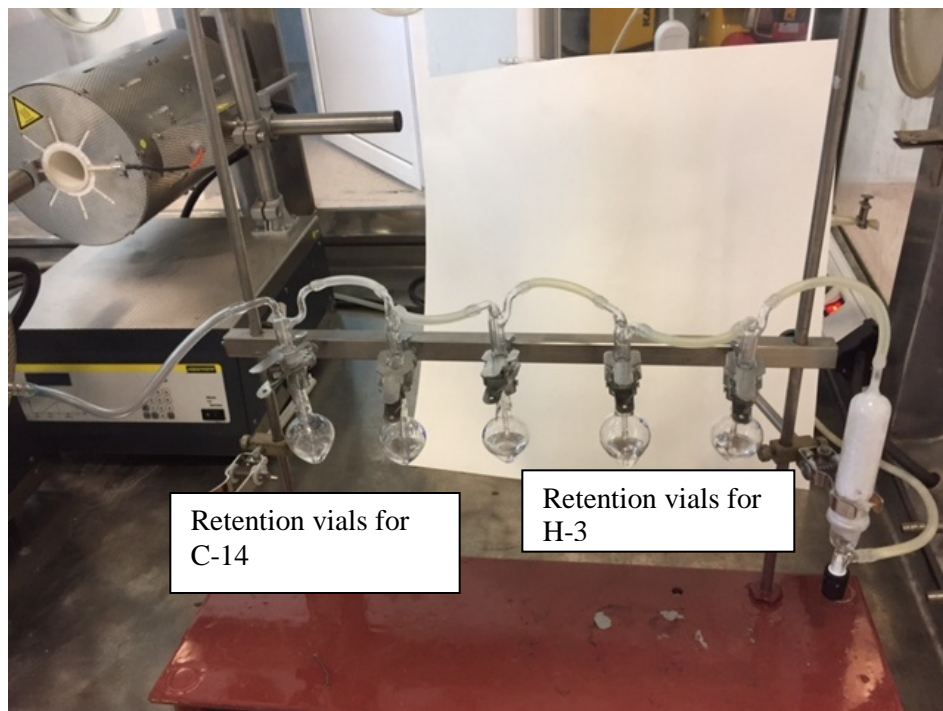


Figure 8: Vials for C-14 and H-3 retention

3 Experimental Results

3.1 *C-14 and H-3 release from VVR-S graphite to liquid phase. (Long term leaching experiment)*

The leaching experiment began in September 2016 and was performed at room temperature. Two graphite samples collected from reactor thermal column were involved in the experiment.

After each sampling, in which a volume of 5 ml of leachate was sampled, the same volume of fresh leachant was added. C-14 and H-3 activity was measured in the immersion liquid by liquid scintillation counting (Carbon-Tritium measurement protocol) and the total C-14 (as CO₂) and H-3 (as HTO) activity released were calculated (WP5 2nd Annual Progress Report – Year 2 (2015)[1].

In order to determine if the radioisotope Cl-36 was present or not in the leachant, the following experimental methodology was used:

- 1 ml of leachant solution was mixed with 1 ml NaCl 0.1 N
- 1 ml Ag NO₃ 0.1 N was added to precipitate chlorine followed by an ultrafiltration step
- the filtrate was analyzed by liquid scintillation counting and C-14 content was determined.

The difference between C-14 activity before and after co-precipitation of chlorine was in experimental errors range. At the conclusion of this experiment, the presence of Cl-36 in the immersion solution was not detected.

In Tables 3 and 4, the C-14 and H-3 total activities released to the liquid phase are presented, for Sample 6.1.1 and 6.1.2.

Table 3 C-14 activities in the liquid phase

Graphite sample 6.1.1	
Sampling period	Total C-14 activity (Bq)
Day	Total C-14 activity (Bq)
1	21763.2
7	68057.8
14	93162.3
21	109302.9
28	127151.3
42	131457.8
56	133648.5
77	136320.0
180	138810.2
270	140612.5
365	141600.1

Graphite sample 6.1.2	
Sampling period	Total c-14 activity (Bq)
Day	Total c-14 activity (Bq)
1	782.6
7	1669.8
14	2205.1
21	2961.3
28	3346.3
42	3434.8
56	3486.3
77	3554.8
180	3710.3
270	3825.4
365	3860.4

The starting inventory of C-14 in graphite samples was 6191.5 kBq for sample 6.1.1 and 338.4 kBq for sample 6.1.2 respectively; the inventory of H-3 was 21802.2 kBq for sample 6.1.1 and 2024.3 kBq for sample 6.1.2.

At the end of the leaching experiment (September 2017) the radionuclide inventory of the immersion solution will be determined by gamma spectrometry using an Ortec Gamma HPGe detector type GMX.

Table 4 Tritium activities in the liquid phase

Sampling period	Graphite sample 6.1.1	Sampling period	Graphite sample 6.1.2
Days	Total H-3 activity (Bq)	Days	Total H-3 activity (Bq)
1	2556.1	1	62.3
7	5772.0	7	78.2
14	7672.3	14	143.8
21	9265.3	21	151.1
28	11046.8	28	197.1
42	12228.7	42	207.9
56	13677.3	56	218.6
77	14789.4	77	236.0
180	15590.1	180	281.2
270	15905.4	270	315.5
365	15971.2	365	320.3

The activities of C-14 and H-3 presented in the tables were calculated based on our previous results taken into account the dilution factor, recovery yield and background. (WP5 3rd Annual Progress Report – Year 3 (2016)) [2].

3.2 C-14 and H-3 leaching rate to liquid phase.

The C-14 average leaching rate (Bq/day and per kg) and H-3 average leaching rate (Bq/day and per kg) were calculated by dividing the activity measured to the sampling period and the sample weight, see Table 5.

Table 5 Carbon-14 and Tritium average release rate into liquid phase

Sampling period	Graphite sample 6.1.1		Graphite sample 6.1.2	
	C-14 average release rate (Bq/day per g)	H-3 average release rate (Bq/day per g)	C-14 average release rate (Bq/day per g)	H-3 average release rate (Bq/day per g)
1	259.6	30.66	9.39	0.744
7	92.55	6.43	1.77	0.032
14	43.02	3.26	0.92	0.112
21	27.66	2.73	1.30	0.013
28	30.58	3.05	0.66	0.039
42	3.69	1.01	0.08	0.009
56	1.88	1.24	0.04	0.009
77	1.03	0.43	0.03	0.007
180	0.32	0.10	0.02	0.006
270	0.24	0.04	0.02	0.005
365	0.13	0.01	0.01	0.001

In Figures 9 and 10, a graphic representation of C-14 and H-3 activities in the immersion liquid during the leaching test are shown.

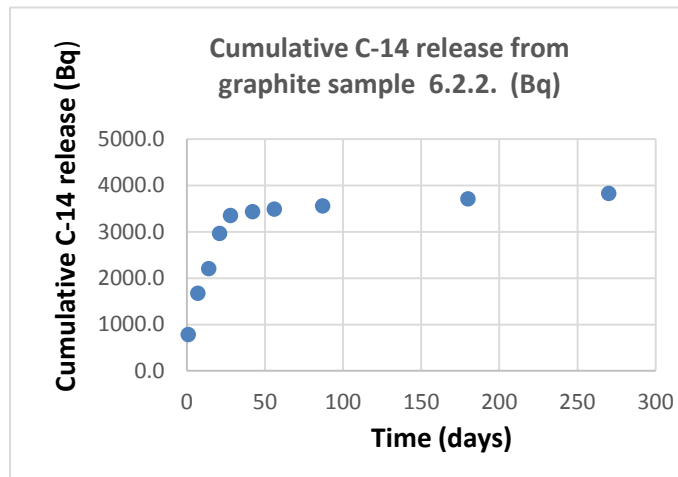
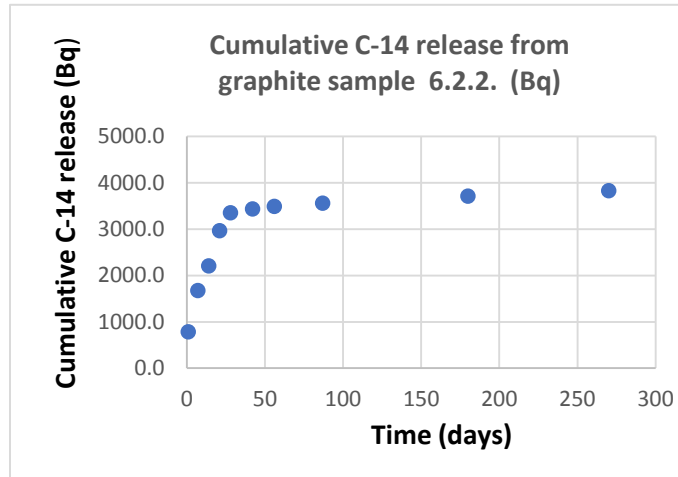


Figure 9. Cumulative C-14 release from i-graphite samples into liquid phase

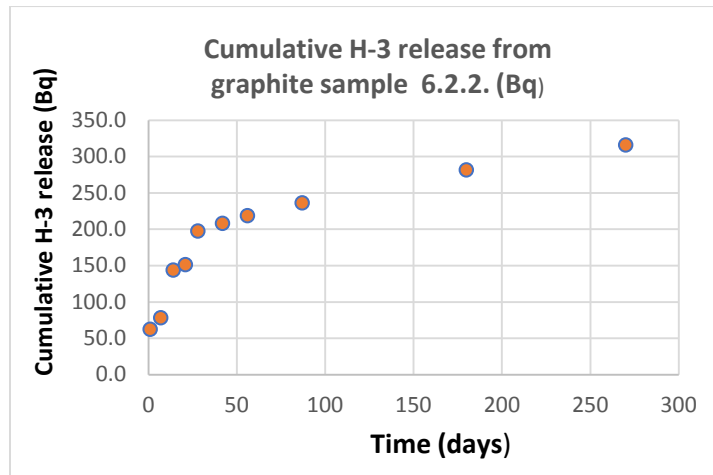
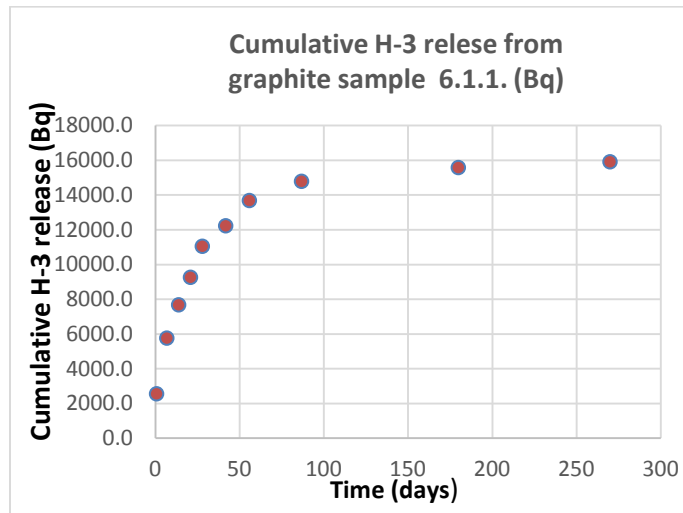


Figure 10. Cumulative H-3 release from i-graphite samples into liquid phase

The average release rate, expressed as percentage of the initial inventory C₁₄ and H-3 activities in the immersion solution for a sampling period, was calculated. The results are presented in Table 6.

Table 6 Carbon-14 and tritium average release rate into liquid phase

Sampling period	Graphite sample 6.1.1		Graphite sample 6.1.2	
	C-14 average release rate (%)	H-3 average release rate (%)	C-14 average release rate (%)	H-3 average release rate (%)
1	0.352	0.012	0.231	0.003
7	1.099	0.026	0.493	0.004
14	1.505	0.035	0.652	0.007
21	1.765	0.042	0.875	0.007
28	2.054	0.051	0.989	0.010
42	2.123	0.056	1.015	0.010
56	2.159	0.063	1.030	0.011
77	2.202	0.068	1.050	0.012
180	2.242	0.072	1.096	0.014
270	2.271	0.073	1.130	0.016
365	2.287	0.073	1.138	0.016

3.3 C-14 and H-3 release from VVR-S graphite to gas phase.

Fourteen days after the samples were sealed, the vessel was connected to a catalytic oxidizer furnace that contains a CuO catalyser inside. The C-14 and H-3 compounds released in gas phase were oxidized in a low oxygen flow rate and trapped separately in 5 vials (Figures 7 and 8). The oxidation process continued for one hour after the connection with the furnace was opened. The temperature of the furnace was kept constant at 800°C during the experiment. At the end of the test, the vessel containing the graphite samples was resealed and the experiment was repeated after 3 months.

The first two trapping vessels each contained 5 ml of sulfuric acid 0.1 N (for tritium trapping) and the following three vessels each contained 5 ml of Carbo-Sorb E (for C-14 trapping). The sulfuric acid solutions in the first two vessels were mixed together. An aliquot (2 ml of retention solution) was transferred to a 20 ml LSC vial, and 16 ml of ULTIMA GOLD cocktail was added. The same procedure was followed for the vessels containing Carbo-Sorb, but this time Permafluor scintillator was used. C-14 and H-3 activity was measured by a TRICARB TR 2800 liquid scintillation counter. Taking into account the dilution factor and the values of the activities measured, the total C-14 and H-3 activity released by the graphite samples into the gas phase was calculated. The results are presented in the next table.

Table 7 Carbon-14 activity and tritium activities in the gas phase

Sampling period (days)	C-14 total activity (Bq)	Background (Bq)	H-3 total activity (Bq)	Background (Bq)
14	0.9±12.5%*	0.5±0.3**	0.8±12.5%*	0.3±0.2**
60	12.15±12.5%*	0.5±0.3**	10.95±11.3%*	0.3±0.2**

* - overall uncertainties-estimated

**-2σ counting uncertainties

The next experiment will be performed after 6 months, in September 2017.

4 Conclusions

- 4.1** Although, the leaching experiment was primarily designed to measure C-14 release from irradiated VVR-S graphite to liquid and gas phase, the apparatus also allowed the measurement H-3; these experimental results have been reported.
- 4.2** The experiments to measure the release of C-14 and H-3 into liquid and gas phase were performed over a 12 months period. The irradiated graphite was collected from the thermal column of the VVR-S Reactor (at IFIN-HH). Two intact graphite samples were taken from the graphite rod located near the reactor core (graphite disc no 6) for use in the liquid leaching test. The mass of the samples were: 83.37 g and 79.88 g respectively. The inventory of C-14 and H-3 is shown in Table 1. The graphite samples involved in the gas phase release experiment were taken from the same graphite rod. The mass and the C-14 and H-3 inventory are presented in Table 2.
- 4.3** The experiments were primarily designed to measure the total release of C-14 and H-3 into liquid and gas phase. The liquid leaching test was performed under condition similar to those relevant to geological disposal (NaOH solution 0.1 M, pH=10, anoxic conditions). Over the 270 days that the leaching experiment was running for, 2.27% of the initial C-14 inventory of graphite in sample 6.1.1 was released to the immersion solution, and 1.13% of the initial C-14 inventory of graphite in sample 6.1.2 was released to the immersion solution. In the case of H-3, the percentages of the initial H-3 inventory released into the liquid phase were 0.073% and 0.016% respectively for these samples. The cumulative release of C-14 and H-3 from graphite samples 6.1.1 and 6.1.2 are presented in Figures 9 and 10.
- 4.4** The leaching rates for C-14 and H-3 were calculated by dividing the activity measured by the sampling period and are shown in Table 5. As it can be seen, these leaching rates were high in the first period of the leaching test (30 days) with a maximum in the first week and then decreasing for both radioisotopes. Generally, the leaching rate for H-3 decreased with time to a lower value compared with C-14 leaching rate.

- 4.5** The rate of release of C-14 into liquid phase decreased to 130 Bq /day per kg after 365 days for graphite sample 6.1.1 and to 10 Bq/day per kg for graphite sample 6.1.2.
- 4.6** The rate of release of H-3 into liquid phase decreased to 10 Bq /day per kg after 365 days for graphite sample 6.1.1 and to 1 Bq/day per kg for graphite sample 6.1.2.
- 4.7** The total C-14 released from graphite samples 6.1.1 and 6.1.2 and retained in solution represented approximately 2.271% of the initial estimated inventory and respectively 1.130%.
- 4.7** The total H-3 released from graphite samples 6.1.1 and 6.1.2 and retained in solution represented approximately 0.073% of the initial estimated inventory and respectively 0.016%.
- 4.8** The experimental results regarding the release of C-14 and H-3 from VVR-S irradiated graphite to liquid show that a major fraction of the total release occurs in the first months and a slower release on long time scale. However, these results should be applied cautiously for a long time prediction.
- 4.9** The total C-14 and H-3 released to gas phase represent approximately 0.00676% and respectively 0.00184% of the estimated inventory in the graphite samples.
- 4.10** The experiment on the release of C-14 and H-3 from VVR-S irradiated graphite to gas phase will be repeated in December 2017, in order to have a better statistic measurements.
- 4.11** The extension of these results over a long period of time should be applied with caution due to the fact that small cracks could appear in graphite block in geological repository conditions and as a result, the release rate could increase. However, on the basis of results presented herein, it is clear that the majority of C-14 and H-3 present in the samples of irradiated graphite tested is not released in leachate or as a gas over the timescale of the experiment; this is consistent with international work on other irradiated graphite, see e.g. [3].

References

1. CAST WP5 2nd Annual report 2015.
2. CAST WP5 3rd Annual report 2016.
3. Carbon-14 Project Phase 2 Irradiated Graphite Wastes, AMEC report to Radioactive Waste Management Limited, AMEC/200047/004 Issue 2, March 2016.

Appendix A

Determination of ^{14}C in graphite samples collected from graphite column of the VVR-S reactor using AMS 9 MV facility

1. Introduction

Carbon-14 has a half-life of 5700 ± 30 years and is mainly produced through neutron-induced reactions with the nuclides ^{14}N , ^{13}C and ^{17}O . They are present in the fuel, fuel cladding, coolant, moderator and structural materials of a nuclear reactor.

Since ^{14}C is decaying to the stable isotope ^{14}N by an electron with maximum energy of 156 keV (mean energy 49 keV) the first experimental option worldwide was to measure it by the beta-decay method.

In 1977, at the Lawrence Livermore National Laboratory, Richard A. Muller and his team recognized that modern linear tandem accelerators could accelerate the radioactive carbon particles to energies where magnetic and electric forces are sufficiently high to eliminate the background interferences. They constructed the first complete and compact Accelerator Mass Spectrometry (AMS) machine that was applied to precise carbon dating

The comparison of the radiocarbon measuring methods (AMS and Beta-Decay) has clearly shown that for the same analytical precision, AMS is about thousand times faster in time, and the sample size needed for the experiment is one thousand times smaller than in the beta decay method. Due to this great success most of AMS machines were engaged to carbon dating. Their main goal was to push the background level (sensitivity) towards 50000 years. Memory effect in the ion source had to be dramatically reduced and measurements were limited to concentrations below the modern carbon level of 10-12 for the $^{14}\text{C}/^{12}\text{C}$ ratio. In many laboratories AMS measurements of higher radiocarbon concentrations were prohibited. However, there is an important need of such measurements for medical application and for Nuclear Reactors, especially for decommissioning procedures.

In our AMS laboratory at Horia Hulubei National Institute for Physics and Nuclear Engineering (IFIN-HH) in Bucharest two machines exist. The first is a new and compact Cockcroft Walton type 1 MV HVEE AMS system that performs precise and low background measurements of ^3H , ^{14}C , ^{10}Be , ^{26}Al and ^{129}I . The second is an AMS facility we constructed 17 years ago, based on our multipurpose 9 MV tandem accelerator. It was intensively used in the frame of the Euratom Program for depth profiling of tritium high dose concentrations. With regard to this machine, the large dose restrictions in experiments are limited to the general radioactive protection standards and regulations. In this frame we were able to respond to some requirements of the decommissioning procedures at the VVR-S Reactor in IFIN-HH, and we measured the neutron-produced ^{14}C concentrations in thermal column of the reactor.

2. Sample description and sample preparation for AMS measurements

The goal of these experiments was to determine ^{14}C accumulation in the thermal column of VVR-S Reactor during a functioning time of about 50 years. Samples for the AMS were collected after the final shut down and fuel removal of the reactor core. Figure A1 presents the layout of the thermal column inside the reactor.

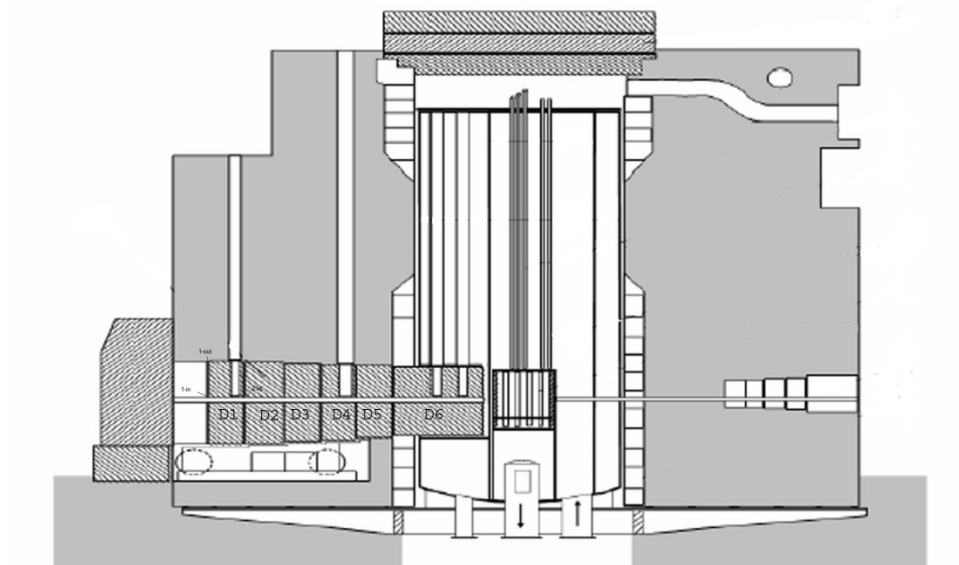


Figure A1: Vertical cut-view through the VVR-S reactor and the movable thermal column with its 6 Discs.

Each graphite disc of the thermal column was enclosed during the reactor functioning in a thick (3mm) aluminum cover. It is about 3m long with the function of moderating the neutron energies. A 50 cm round opening along the structure is forming the thermal column.

During fission operation the column was filled with a graphite rod, covered by a 1 cm thick cylindrical graphite coating that sealed the entire opening in the column. The sampling for the AMS analysis was performed from several positions inside and outside the graphite coating of the rod. Samples were finally cut in small squared slices 7 x 7 x 3 mm from the graphite contained in the rod at locations corresponding to the 6 discs of the thermal column.

The sample dimensions were required by the target holder of the AMS sputter ion source. Since samples are made of solid carbon, preserving their bulk structure a depth profiling of the accumulated isotope concentration is also possible by our analyzing method.

3. Experimental method and results.

Large or high dose ^{14}C AMS experiments were performed by our analyzing machine constructed on the multipurpose 9 MV FN tandem accelerator in Bucharest. A general layout is presented in Figure A2.

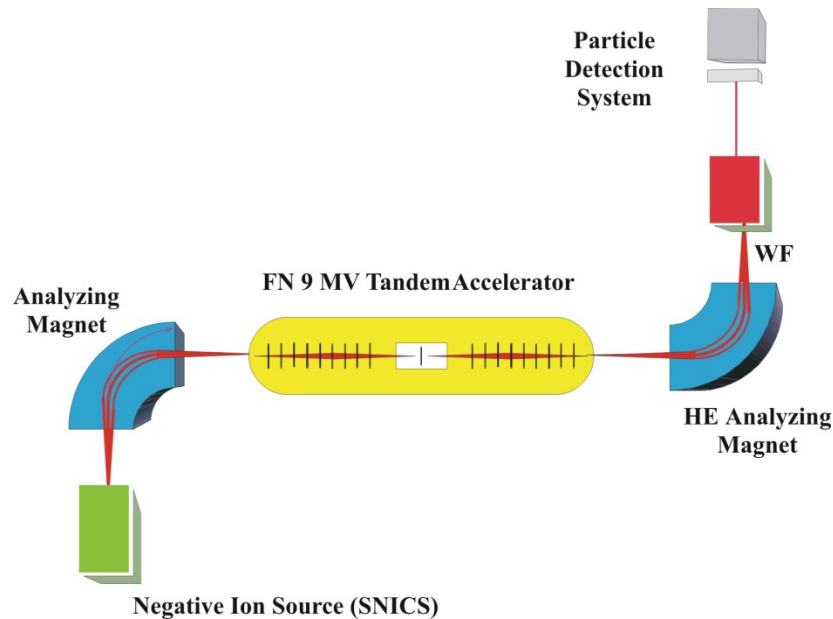


Figure A2: Schematic diagram of the AMS- facility based on the 9 MV FN tandem accelerator.

The main parts are the AMS dedicated ion injector with the ion sputter source and the powerful vacuum system acting on the ion source sputter chamber and a 900 double focusing analyzing magnet. Then, a pre-acceleration stage of 140keV is placed before the entrance into the accelerator for the improvement of beam emittance followed by the 9MV accelerator with a foil stripper in the terminal. At the accelerator exit a powerful analyzing magnet is located on the high energy side, and a Wien velocity filter is then located before the detection system, based on an array of surface barrier detectors Si(Au). For ^{14}C only the first detector was used since we tuned the terminal voltage at 2.7 MV to operate the accelerator at charge state $3+$ (total energy).

Figure 3(a) shows the registered spectrum before setting into operation the Wien velocity filter. The accelerated beam shows for ^{14}C a small peak positioned at the left (lower energy 10.8 MeV) of the large group of ions with $3+$ charge (^{13}C and C^{12}). The $^{13}\text{C}^{2+}$ shows itself as a pronounced, separated peak.

After the final analyses through the velocity filter, a clean signal of ^{14}C emerges. For high dose measurements exceeding the acceptance of our detection system (event frequency 2.5 kHz) we use a calibrated ion beam attenuator of 1000.

A new Preamplifier (ORTEC 142A) introduced recently in the last experiments contributed essentially to both the energetic resolution and increase of signal acquisition from the detector (Figure A3). Figure A4 presents the actual set up of the new preamplifier, the detector system and Velocity Filter.

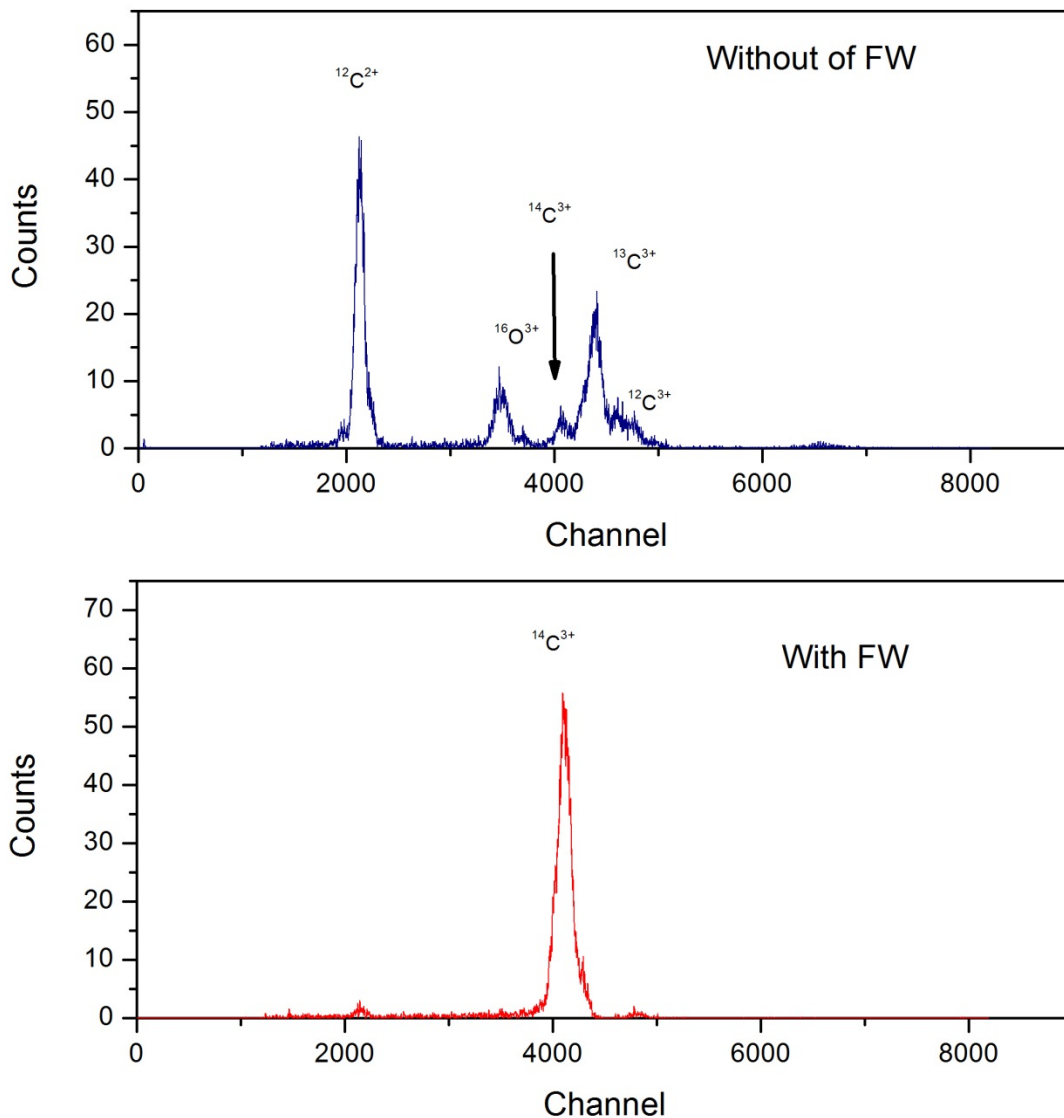


Figure A3: Detection of ^{14}C at an acceleration energy of 10.8 MeV . a) Spectra measured in the detector after acceleration through the tandem and analyses in the high energy magnet. b) Spectrum after the velocity filter is switched on. Both spectra were measured for 60 sec.



Figure A4: Preamplifier, Detector and Velocity filter set up.

Results of experiments

Figure A5 shows the results of measurements performed in this AMS experiment. Four sectors from the Disk 6 and one from Disk 5 were measured. Disk 6 is located close to the active region, as described in section 2. Radiocarbon concentrations are ranging from a maximum value of 22 kBq/g close to the reactor fission vessel down to about 0,5 kBq/g at the end of Disk 6. In the next Disk, first sector is even lower in the ^{14}C concentration. Exact values are given in Table 1.

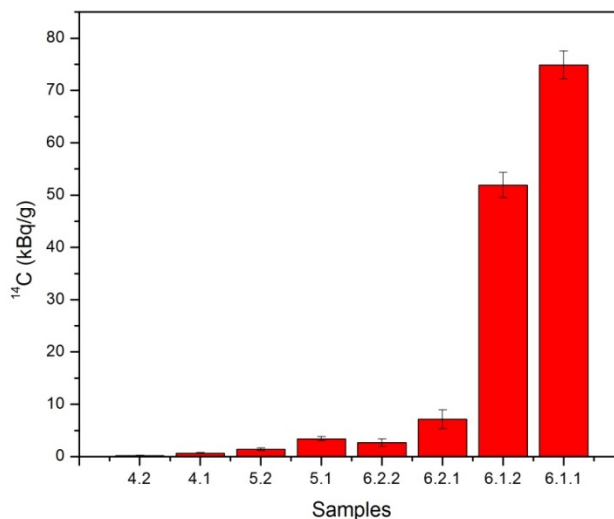


Figure A5: AMS measurement of ¹⁴C concentrations in Disk 6 and Disk 5 of the thermal column of the decommissioned VVR-S reactor in Bucharest

Sample	¹⁴ C (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.

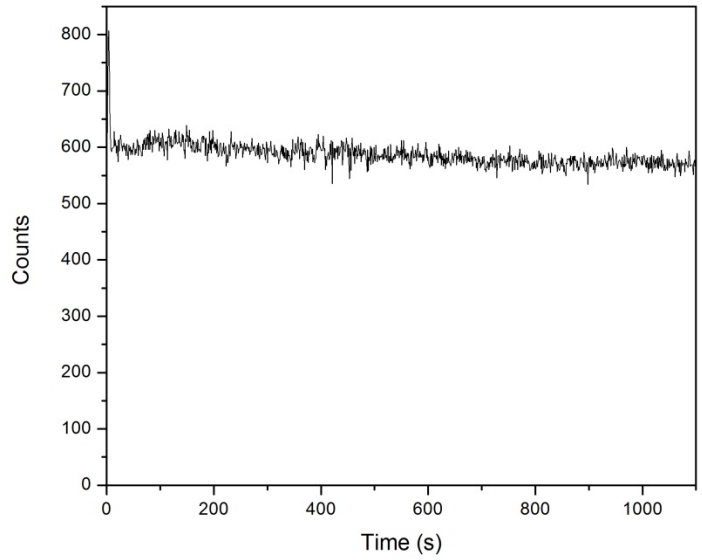


Figure A6: Depth profiling of 14C concentration in graphite of the thermal column.

Since the neutron fluxes were intense, the 14C depth profile concentrations were constant and monotone for each sample 3 mm thickness, only.