EUROPEAN COMMISSION

Community Research

SEVENTH FRAMEWORK PROGRAMME

CARBOWASTE

Treatment and Disposal of Irradiated Graphite and Other Carbonaceous Waste

Grant Agreement Number: FP7-211333

Deliverable (D-1.1.4)

WP 1 Review Report – France

Author(s): **L Petit**

Reporting period: 05/2008– 03/2009

Date of issue of this report: 04/2009

Project co-funded by the European Commission under the Seventh Framework Programme (2007 to 2011) of the European Atomic Energy Community (EURATOM) for nuclear research and training activities		
Dissemination Level		
PU	Public	
RE	Restricted to the partners of the CARBOWASTE project	X
CO	Confidential, only for specific distribution list defined on this document	

Start date of project : **01/04/2008** Duration : **48 Months**

Distribution list

Person and organisation name and/or group	Comments
CARBOWASTE WP 1 Partners	Draft for review
All CARBOWASTE participants	Issue 1 final version

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

CARBOWASTE		
Work package: 1 **Task: :** 1.1	**CARBOWASTE document no:** CARBOWASTE-0904-D-1.1.4	**Document type:** D=Deliverable /
Issued by: EDF (France) **Internal no.:**		**Document status:** **Final**

Document title
Integrated Waste Management Approach WP 1 Review Report - FRANCE
Executive summary

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

This chapter addresses irradiated graphite wastes in France, i-graphite represents about 23,000 tons originating mainly from UNGG reactors (Gas Cooled Reactors). Most of i-graphite lies in shutdown reactors except for small quantities of graphite sleeves temporarily stored in silos. EDF and CEA that were the operators of the nine UNGG reactors from the 60's to the 90's, are the main producers of i-graphite in France. For all these nine UNGG reactors, the AIEA level 2 decommissioning status has been achieved.

In 2001, EDF took the decision to achieve AIEA level 3 decommissioning status before 2025 for all the six EDF UNGG reactors, including three other shutdown reactors without i-graphite (Chooz A, the first French Pressurized Water Reactor - Brennilis, a heavy water moderated reactor - Superphenix, a fast breeder reactor). This new strategy aims :

- To show the feasibility of a total dismantling on an industrial scale ;
- To show the capability to manage all the generated material and wastes ;
- To take this opportunity to set up ad to organize and internal decommissioning body (EDF Ciden in Lyon) ;
- To be able to take in charge, when it occurs, the future decommissioning of the operated PWR, through the case of Chooz A.

This new strategy can also be considered as a response to the French Nuclear Safety Authority that pointed out the risk of loosing information about a shuttered plant over longer periods.

The UNGG Bugey 1 reactor is intended to be first UNGG reactor to be dismantled by 2013. The decision was taken by EDF that the dismantling works would be done underwater mainly because the shielding afforded by the water would enable a more "hands on" approach to be adopted rather than a fully remote operation. It is considered that it allows a greater flexibility to respond to unforeseen technical problems during dismantling.

French i-graphite waste from UNGG reactors are considered as Low-Level activity waste but because small amount of two long-lived radionuclides (C-14 and Cl-36) are present, their disposal at the existing Aube surface repository site for Low-Level and Short-Lived waste, is not acceptable. A new repository site for i-graphite is to be commissioned. According to the French Act of Parliament of June 2006, a research and investigation program has been established with a view to developing disposal options for graphite waste in order for a corresponding disposal facility to be commissioned in theory in 2013. According to the latest developments of this program, the commissioning date is intended to be more probably 2019. The design, licensing, construction and operation of this new disposal facility for i-graphite is under the responsibilities of ANDRA, the French national radioactive waste management agency.

The preliminary i-graphite radiological inventory has been assessed from the highest measured values for each radionuclide among the numerous samples that were analyzed. This method led to an overestimated radioactive inventory used for preliminary studies on disposal. A new method based on data assimilation and activation calculation is being developed by EDF. Its objective is to provide a more realistic inventory for safety studies on the disposal, able to explain the variation of radionuclide measurements that was observed depending on the sample localization.

Concerning studies on graphite waste disposal, a major focus have been made on long-lived radionuclides' behavior (C-14 and Cl-36) in i-graphite leaching experiments. It is shown that C-14 leaching rate is very low. These results are consistent with EDF activation calculation results showing that the reminding C-14 in i-graphite originates mainly from C-13 activation. More studies that aimed to be carried out in the WP6 are needed, in order to assess the long term C-14 behavior in disposal conditions.
The results obtained on Cl-36 leaching experiments are very variable from one sample to another. The temperature history of i-graphite during operation seems to be a key parameter for Cl-36 behavior: the lower the working temperature is, the higher is the Cl-36 release. More studies are needed in the WP6 to explain the observed variation on Cl-36 leaching rate in order to assess the impact of underwater retrieval on graphite stack radioactive inventory and consequently the Cl-36 behavior in long term disposal.

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

Revisions						
Rev.	Date	Short description	Author	Internal Review	Task Leader	WP Leader
01	23/12/08	Issue for WP review	*L Petit (EDF)*	L. Gosmain (CEA) F.Bérenger (EDF)	H Eccles (UK NNL)	H Eccles (UK NNL)
02	21/03/09	Final issue	*L Petit (EDF)*	L. Gosmain (CEA) F.Bérenger (EDF)	H Eccles (UK NNL)	H Eccles (UK NNL)

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

Chapter Contents

Deleted: 11
Deleted: 11
Deleted: 12
Deleted: 15
Deleted: 16
Deleted: 20
Deleted: 21
Deleted: 21
Deleted: 23
Deleted: 24
Deleted: 25
Deleted: 25
Deleted: 26
Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

Deleted: *30*
Deleted: 35
Deleted: *39*
Deleted: *39*
Deleted: *41*
Deleted: *41*
Deleted: 43
Deleted: *43*
Deleted: *48*
Deleted: *49*
Deleted: 49

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

1 Introduction

This chapter reviews the i-graphite waste management challenges for France. Consistent with other chapters the magnitude of the i-graphite inventory (about 23,000te) is discussed and the current disposition of the material, largely in off-line reactors. The design and configuration of the reactors and the challenges these pose to retrieval are addressed. The graphite characteristics and analysis undertaken on a comparative large number of samples are explained. The French decommissioning policy/strategy and how this could influence the selection of graphite retrieval options is also discussed. For four of the nine existing UNGG reactor, an underwater technique is adopted.

Deleted: Currently the French are adopting a underwater technique

2 The legislative and regulatory framework

Most of the graphite waste originating from natural-uranium gas-cooled reactors (acronym in French UNGG) are classified in the long-lived low-level (LL-LL) waste category. In France, this category includes the particular category of waste containing a significant quantity of radium and producing radon. This type of waste was notably produced in the past by the rare-earth industry.

2.1 National policy on graphite waste

Decommissioning operations produce radioactive waste which has to be managed with the principle of reliability, transparency and of a stringent management of the waste in order to ensure the protection of individuals, the preservation of the environment and the limitation of undue burdens imposed on future generations. In this respect two important acts were promulgated in 2006:

- the “Transparency and Security in the Nuclear field” Act (13 June 2006), sometimes called TSN Act [1] ;
- the Planning Act on the sustainable management of radioactive materials and waste (28 June 2006). The articles of this 2006 Planning Act are now part of the Code of Environment [2]. Concerning more especially i-graphite, this planning act of 28 June 2006 establishes a research and study program with a view to “developing disposal options for graphite and radium-bearing waste in order for a corresponding disposal facility to be commissioned in 2013”.

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

A National Plan for the management of radioactive materials and waste is considered as an important tool to improve radioactive waste management. The plan (PNGMDR) has been issued at the beginning of 2007 by the Nuclear Safety Authority (ASN) and the General Directorate for Energy and Raw Materials (DGEMP). It is notably based on the National Inventory of radioactive waste and recoverable materials [3], issued by the National Radioactive Waste Management Agency (Andra). Concerning i-graphite, the Decree of 16 April 2008 sets forth relevant requirements for this National Management Plan a follows:

- Graphite and radium-bearing waste shall be stored pending the availability of a disposal facility;
- The disposal facility shall be studied and design to accommodate graphite and radium-bearing waste. The possibility to take over other LL-LL waste shall also be investigated;
- On the basis of local investigations, Andra shall submit by the end of 2009 an analysis of suitable sites for the implementation of a disposal facility.

Andra's scientific board and the National Review Board (CNE - see below) have emphasized the extremely constraining nature of the commissioning date of 2013 for the graphite disposal facility as established in the planning act of 28 June 2006. At the request of the government, Andra drew the following realistic schedule for the project [4], that may be refined in 2009, during the update of the PNGMDR:

- Launching of the site-search phase in mid-2008;
- Site selection at the end of 2010;
- Development of disposal solutions in preparation for the implementation - license application in 2013;
- Expected commissioning in 2019.

2.2 Relevant institutions

2.2.1 Energy policy

The institutions in charge of energy policy are the French Parliament [5], the French Government [6] and the DGEMP [7] (General Directorate for Energy and Raw Materials) of the Ministry of Ecology and Sustainable Development.

2.2.2 ASN, the Nuclear Safety Authority

The Act of 13 June 2006 (or TSN Act) creates a nuclear safety authority as an independent administrative body. The acronym of the nuclear safety authority is ASN [8] (Autorité de

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

Sûreté Nucléaire). The missions of this new ASN were previously assigned to *the Direction générale de la sûreté nucléaire et de la radioprotection*" (DGSNR), and *the Divisions de sûreté nucléaire et de radioprotectio*n" (DSNR) set up within the Regional Directorates for Industry, Research and the Environment (DRIRE).

ASN takes its regulatory decisions after the assessment of the license applications by the Institute for Nuclear Safety and Radiation Protection (IRSN - technical support of the ASN - see below). In the context of application for Basic Nuclear Installations (INB) licenses, modification or final shutdown and decommissioning decrees, the Consultative Commission for Basic Nuclear Installations (CCINB) is consulted by the Minister for Ecology and Sustainable Development and the Minister of Finance, Economy and Employment.

2.2.3 Andra, the National Radioactive Waste Management Agency

A specific public agency, Andra, has the responsibility for the long-term management of radioactive waste. This agency operates waste repositories, defines the acceptance criteria for waste packages in these repositories and controls the quality of their production. The agency is also in charge of designing and constructing new disposal facilities. It keeps up to date the National inventory of radioactive waste and recoverable materials in France.

2.2.4 CEA, the Atomic Energy Commission

The CEA [9] was created by Ministerial Order 45-2563 of 18 October 1945 to carry out R&D for the implementation of civilian nuclear activities (energy, industry, research and health) and to provide the necessary support to the development of National Defense activities (Nuclear deterrent forces).

Since the 2006 Planning Act, CEA still carries out R&D as support to Andra program on graphite waste disposal on the basis of bilateral agreement.

2.2.5 CNE, the National Review Board

In addition an already existing regulator (ASN, see above), the December 1991 Waste Act has prescribed the creation of an expert review committee, the CNE (National Review Board) [10], to assess and review the various programs carried out for the management of high-level and long-lived intermediate-level radioactive waste.

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

Moreover, the 2006 Planning Act has extended the National Review Board (now called CNE 2 to distinguish it from the former CNE established by the December 1991 Waste Act) mission to the review of all radioactive waste management R&D programs mentioned in the PNGMDR, including the graphite waste disposal. Its reports are issued on a yearly basis for the government. The annual report shall be transmitted by the government to Parliament, which in turn forwards it to the Parliamentary Office for the Evaluation of Scientific and Technological Choices (OPECST), before being made public.

2.2.6 IRSN, the National Institute for Radiation Protection and Nuclear Safety

IRSN (*Institut de radioprotection et de sûreté nucléaire)* [11] is the public body in charge of the scientific assessment of nuclear and radiation risks. It is mandated for advising the public authorities and contributing to public policies, for developing the research activities necessary to support its scientific appraisal and for delivering services to industry and other organizations.

2.2.7 Operators and producers of graphite waste

In France, operators are responsible for financing the management of their waste and the decommissioning of their nuclear installations. Three operators are involved in graphite waste production:

- EDF [12]: French utility operated six UNGG reactors, now all shutdown. The amount of graphite waste that EDF will produce in dismantling these units is about 17,000 tons (including graphite sleeves temporarily stored in silos in Saint-Laurent);
- CEA [13]: French government-funded technological research organization that operated the three first UNGG reactors linked to the French nuclear deterrent force in Marcoule. CEA also operated or still operates experimental reactors using graphite. The amount of i-graphite in CEA is about 4,700 tons.
- AREVA [14]: French business corporation covering all industrial activities in the field of nuclear power. AREVA within its activity in used fuel treatment, have temporarily stored in silos graphite sleeves. The amount of i-graphite in AREVA is a little less than 1,000 tons.

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

3 Policy objectives of decommissioning

3.1 Regulations concerning decommissioning / dismantling

Decommissioning/dismantling operations are covered by the decree of 2 November 2007. This decree takes into account the provisions of the SD3-DEM-01 document published by ASN in 2003. It requires that the final shutdown and the decommissioning shall be licensed by a specific decree. A public inquiry takes place now systematically if the licensee has submitted its application after the 3 November 2007. The licensee shall submit a detailed decommissioning/dismantling plan one year before the final shutdown. After completion of the decommissioning/dismantling operations and the clean-up of the site, the former nuclear site may be still covered by some restrictions of use, after a public inquiry.

3.2 Decommissioning strategy

The dismantling scenario (immediate or deferred) is selected by the operator. Although the regulations do not stipulate dismantling as soon as reasonably feasible, the ASN is in favor of immediate dismantling for various reasons such as loss of familiarity with the design and operation of the installation, the minimal advantage gained from radioactive decay, or the risk of equipment obsolescence.

In January 2001, EDF chose to adopt a new dismantling strategy for all its nuclear facilities which have been finally shutdown (Brennilis, Bugey 1, Saint-Laurent A, Chinon A, Chooz A and Superphénix), based on level 3 dismantling of the reactors in advance without a waiting period. This new strategy aims:

- To show the feasibility of a total dismantling on an industrial scale;
- To show the capability to manage all the generated material and wastes;
- To take this opportunity to set up ad to organize and internal decommissioning body (EDF Ciden in Lyon);
- To be able to take in charge, when it occurs, the future decommissioning of the operated PWR, through the case of Chooz A.

The UNGG Bugey 1 (BU1) reactor is intended to be first UNGG reactor to be dismantled by 2013. The methodologies and techniques being developed for BU1 are intended to be applied across the other reactors where practicable to do so. The safety report describing the proposed

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

methodology and setting out the safety and environmental justifications for the methodology and equipment design has been submitted to the French safety organization. The license to dismantle BU1 is expected to be issued in the second half of 2008. The beginning of the dismantling operation (flooding of the main pressure vessel) is intended to start in 2013.

4 Current situation in France

The two operators involved in nuclear graphite reactors decommissioning in France are EDF (Electricité De France) and CEA (Commissariat à l'Energie Atomique).
Among the nine EDF nuclear reactors to be decommissioned in the next 20 years, six of them are gas-cooled moderated-graphite reactors (GCR). They were used to produce electricity in the seventies and the eighties, and shutdown in the beginning of the nineties mainly for economical reasons.

CEA reactors were in most cases small experimental reactors that were used to test and develop new concepts, or reactors linked to the French nuclear deterrent force. Except for ULYSSE (CEA teaching reactor shutdown in 2007 in Saclay) and PHENIX, a fast breeder CEA reactor using small quantities of graphite (neutronic shielding), still in operation (and that will be shutdown in 2009), the other CEA reactors were shutdown about 20 years ago.

The location of these nuclear reactors using graphite and of some interim storage (silos) is reported in Figure 1. A summary table of irradiated graphite that will be produced in the next years from UNGG dismantling in France is presented in Table 1. The total amount of irradiated graphite for these reactors is about 23,000 tons.

Deleted: Figure 1
Formatted: Font: (Default) Times New Roman, Not Bold
Deleted: Table 1

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

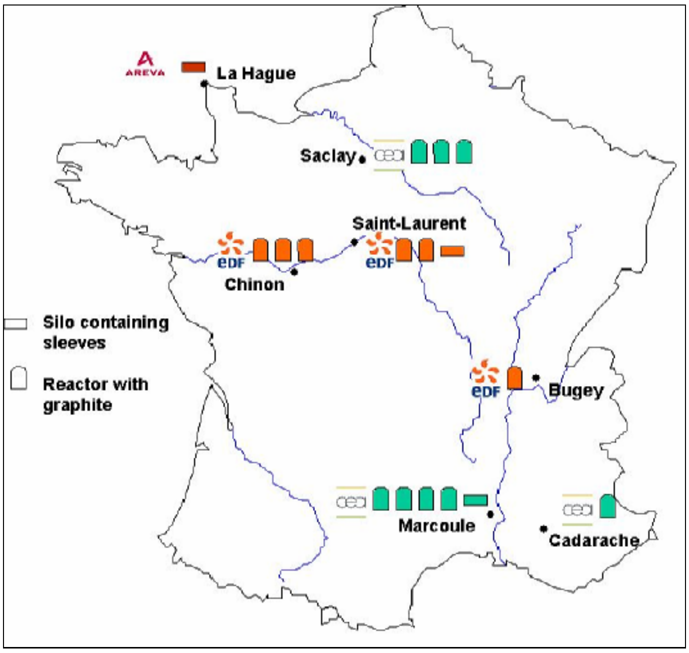

Figure 1 : Location and owners of irradiated graphite in France

Table 1 : Summary table of irradiated graphite for each producer in France.

Weight (tons)	CEA	EDF	AREVA NC	Total
Graphite from stacks	3,980	15,000	0	18,980
Graphite sleeves	730	2,030	970	3,730
Total	4,710	17,030	970	22,710

4.1 EDF UNGG reactors

EDF is in France the owner of six GCR[1] that will have to be totally dismantled in the next 20 years. For all these six reactors, fuel has been removed. All ancillary plant has been removed for CHA1, CHA2, SLA1 and SLA2. The AIEA level 2 decommissioning status has been achieved for CHA1.

[1] For practical reasons each reactor is designated in the text with its acronym : BU1 = Bugey 1, SLA1 & SLA2 = Saint-Laurent A1 and Saint-Laurent A2, CHA1 & CHA2 & CHA3 = Chinon A1 and Chinon A2 and Chinon A3.

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

These reactors are of so-called in French, UNGG reactor type (natural-uranium gas-cooled reactors). That means that they were graphite moderated, cooled by carbon dioxide and fuelled with natural uranium. The design of UNGG reactors (Figure 2) is, in its general principle, very close to that of the British Magnox reactors, that was developed independently in the same period. One of the main differences between the two concepts lies in the cladding made of magnesium-zirconium alloy in UNGG, instead of magnesium-aluminum alloy in Magnox reactors. The main characteristics of EDF UNGG reactors are summarized in Table 2.

Deleted: Figure 2

Deleted: Table 2

Although these six reactors are built following the same general principles, their design is very different on some aspects. These differences are mainly due to technological developments aiming at increasing their efficiency on an economical point of view and the power of each new unit that was built [15]. From CHA1, the first EDF UNGG reactor in operation in 1963 to BU1 the last one built in 1972, the UNGG reactors type moves from a specific power of 1.5 W / g (of fuel) in CHA1 to 5.7 W / g in BU1.

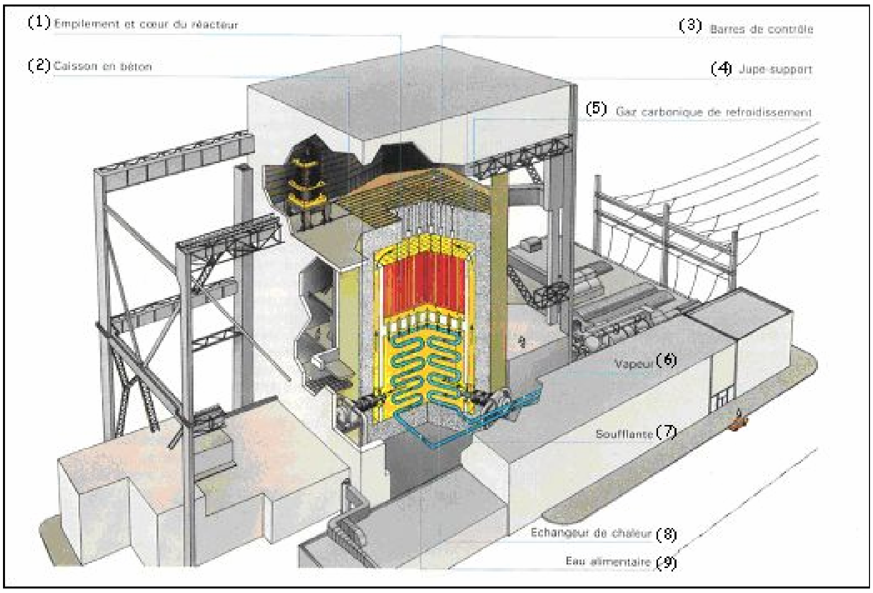

Figure 2 : Schematic view of an UNGG type reactor - (1) Stack and core of the reactor - (2) Pre-stressed concrete pressure vessel - (3) Control rod - (4) Support skirt - (5) Cooling gas - (6) Steam - (7) turbo-blower - (8) Heat exchanger - (9) Water feeding.

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

A more detailed description of the internal parts of the reactor building is presented for BU1 reactor in Figure 3. General elevations for CHA1, CHA2, CHA3 units are presented respectively in Figure 4, Figure 5and Figure 6.

Formatted: Font: (Default) Times New Roman, Not Bold
Deleted: Figure 3
Deleted: Figure 4
Deleted: Figure 5
Deleted: Figure 6

4.1.1 Graphite stack

The graphite stack is made of eight to twelve levels of graphite columns with different shape depending on the unit considered (Figure 7). The mechanical assembly of the graphite stack is assured via a simple "mortise and tenon" system-like and a metallic corset all around the stack. Its dimensions for BU1 are of about 10 meters high, 15 meters in diameter and weighing some 1,200 tons. Each graphite column has a hole through - the channel - in which fuel cartridges or control rods were loaded. Except for CHA1, the channels are in the vertical position (Figure 8) mainly in order to simplify fuel handling during operation. The reflector bricks of the external parts of the stack are solid (no hole).

Deleted: Figure 7
Formatted: Font: (Default) Times New Roman, Not Bold
Formatted: Font: (Default) Times New Roman, Not Bold
Deleted: Figure 8

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

Table 2 : Main characteristics of EDF UNGG reactors.

UNGG Reactor	Chinon A1 (CHA1)	Chinon A2 (CHA2)	Chinon A3 (CHA3)	Saint-Laurent A1 (SLA1)	Saint-Laurent A2 (SLA2)	Bugey 1 (BU1)
Criticality date	June 1963	February 1965	August 1966	March 1969	August 1971	April 1972
Shut down date	April 1973	June 1985	June 1990	April 1990	May 1992	May 1994
Projected decommissioning dates (main pressure vessel) [19]	2017-2023	2015-2023	2014-2023	2015-2022	2014-2021	2012-2020
Location	Avoine			Saint-Laurent		Bugey
Thermal power (MW)	300	850	1 560	1 650	1 700	1 920
Electrical power (MW)	70	210	365	480	515	540
Cooling (pressure MPa)	CO_2 (2.5)	CO_2 (2.6)	CO_2 (2.6)	CO_2 (2.7)	CO_2 (2.9)	CO_2 (4.3)
Mass of graphite (t)	1,120	1,650	2,350	2,570	2,440	2,039
Graphite temperature (°C) during operation	150-360	190-390	240-410	230-430	240-440	220-450

4.1.2 Fuel cartridge

Each fuel channel was loaded with 12 to 15 fuel cartridges (the fuel in its cladding). As fuel cartridges located in the lowest position were not able to support the mechanical stress generated by the weight of the other cartridges above, graphite sleeves (Figure 9) were used to allow the transmission of the vertical efforts (except for CHA1). The graphite sleeve is a tube made of graphite weighting about 10 kg, sliding in the channel and equipped inside with a graphite seat support so as to maintain the fuel cartridge in its center. The graphite seat support is attached to the sleeve via metallic wires called in French “fil de selle”.

In order to increase the thermal power exchange capacity, different designs of fuel cartridges were successively developed. Cooling fins with different shapes were added. The standard fuel

Deleted: Figure 9

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

cartridge used for CHA3, SLA1, SLA2 and BU1 is presented in Figure 10. This fuel cartridge was also used in the Vandellos UNGG reactor in Spain

Deleted: Figure 10

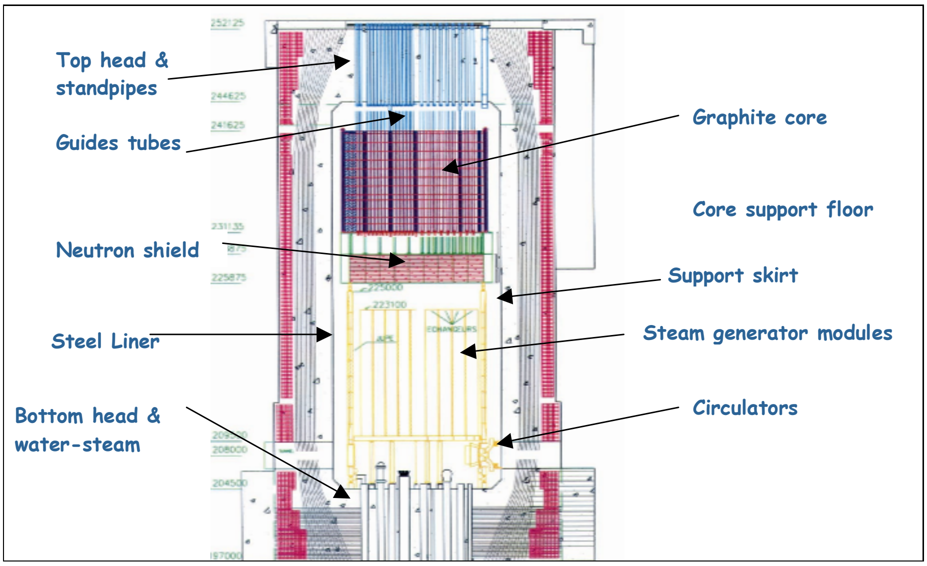

Figure 3 : BU1 reactor elevation [26]

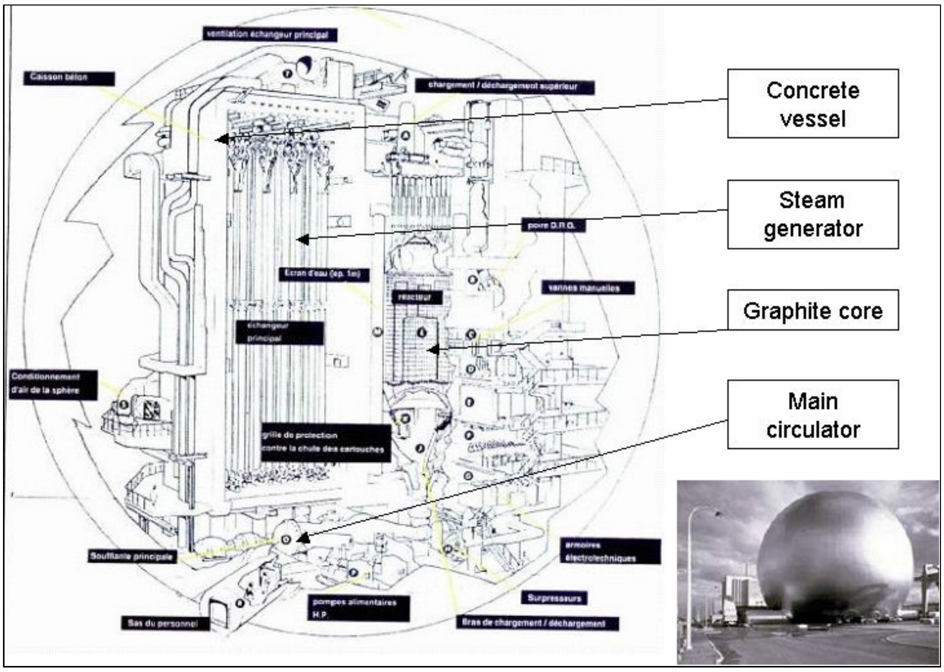

Figure 4 : CHA1 reactor elevation and external photography.

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

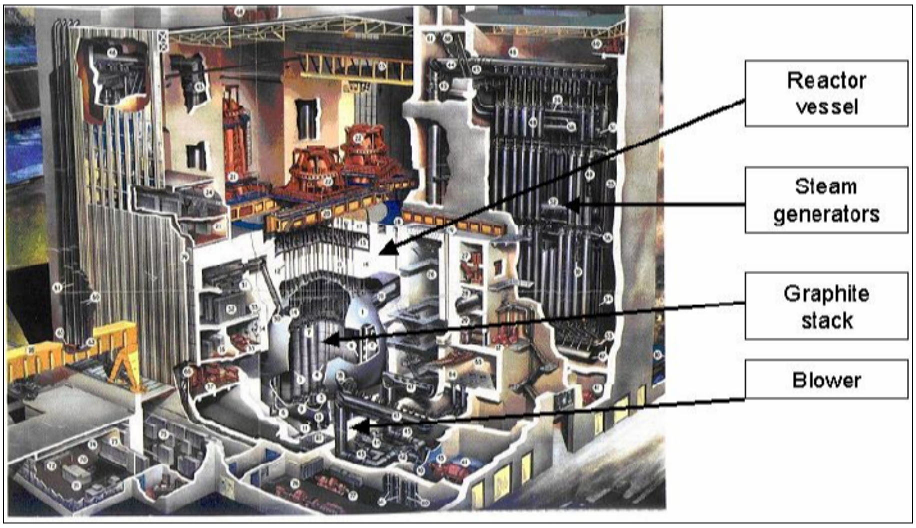

Figure 5 : CHA2 reactor elevation.

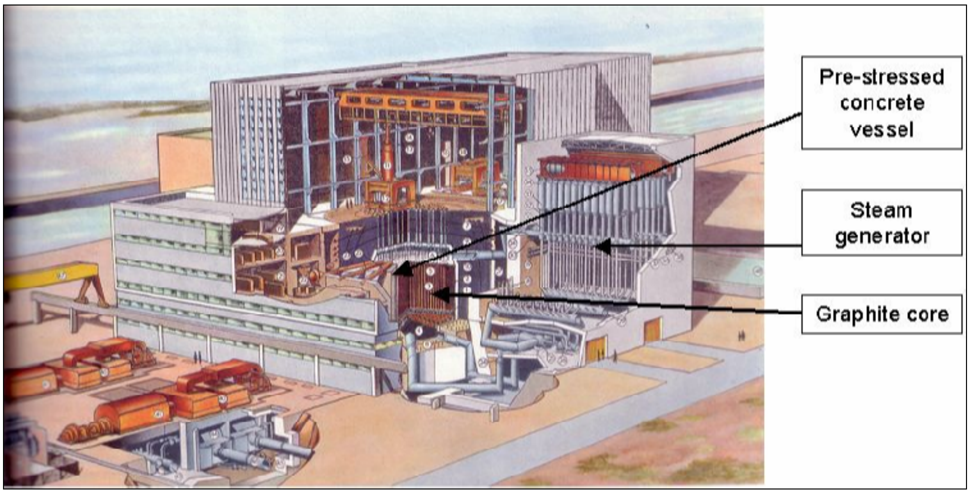

Figure 6 : CHA3 reactor elevation.

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

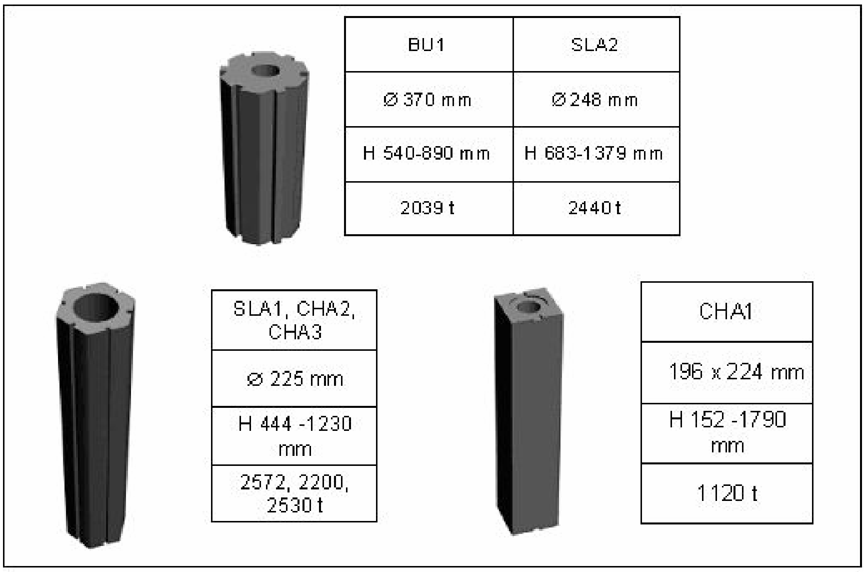

Figure 7 : Different shape of graphite bricks used for the stack of EDF UNGG reactors [18].

Figure 8 : Stack of graphite columns during Chinon A3 building in 1965

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

4.1.3 Reactor vessel

4.1.3.1 The reactor pressure vessel

For CHA1 and CHA2 (see Figure 4 and Figure 5), the pressure vessel is made of concrete. For CHA1, the concrete core is confined in a spherical steel envelope. For CHA3, SLA1, SLA2 and BU1, the reactor pressure vessel is made of pre-stressed concrete (5 to 5.5 meters thickness). In these later, a steel liner carries out the sealing with respect to CO_2 pressure.

Deleted: Figure 4

Deleted: Figure 5

4.1.3.2 The steam generation unit location

For the first units located in Chinon, the steam generator is located apart from the reactor vessel. The three last units in Saint-Laurent and in Bugey have integrated boilers under the core inside the reactor pressure vessel.

4.1.3.3 The thermal insulation of the concrete walls of the pressure vessel

Different thermal insulation devices were used to protect the concrete walls against the reactor temperature. For BU1, cooling water pipes were managed in the concrete wall under the steel liner (the steel liner was used as framework for the casting of the concrete walls during the reactor vessel building). In the case of Saint-Laurent reactors (SLA1, SLA2) and CHA3 reactor, a thermal insulation device mainly made of “béton ponce” (special lightweight concrete) and steel casing was managed inside the reactor to protect the steel liner and the pre-stressed concrete walls of the reactor vessel.

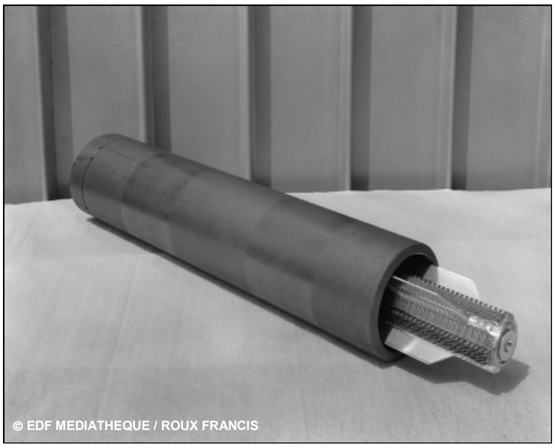

Figure 9 : Fuel cartridge with its graphite sleeve.

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

4.1.4 Operating conditions

4.1.4.1 Gas flow

For Chinon reactors (CHA1, CHA2 and CHA3), cooling gas flows from the bottom to the top of the reactor (the lower working temperature of graphite was at the bottom of the stack). For Saint-Laurent reactors (SLA1, SLA2) and BU1, cooling gas flows in the opposite direction (from top - lower temperature - to the bottom).

4.1.4.2 Cooling gas composition

In BU1 methane injections were used in order to limit radiolytic oxidation and weight loss of the graphite stack [24].

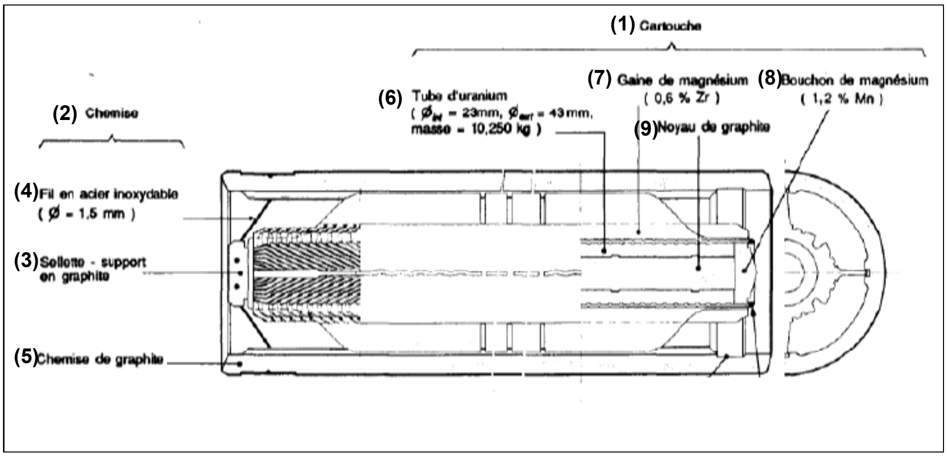

Figure 10 : General scheme of a standard fuel cartridge (CHA3 - SLA1 - SLA2 - BU1 - Vandellos) - (1) cartridge - (2) sleeve - (3) seat made of graphite - (4) stainless steel wire - (5) graphite sleeve - (6) uranium pipe - (7) magnesium cladding - (8) magnesium end plug - (9) graphite central rod [25].

4.1.5 CEA reactor using graphite

CEA is the owner of four main types of nuclear reactors using graphite:

- The first UNGG reactors developed in France: G1, G2 and G3. These reactors were linked to the French nuclear deterrent force. G1 was an air cooled reactor and G2 and G3 were carbon dioxide cooled reactor. Graphite is used as moderator for the stack and as reflector and shielding for the internal walls of the reactor. No sleeve was used for the fuel cartridges.

  Their working temperature was lower than that in EDF UNGG reactors, and the geometry of the stack is slightly different as shown in **Figure 11** and **Figure 12**. The channels of the stack in which fuel cartridge were loaded, are in horizontal position (as

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

in EDF CHA1 reactor). For all these three reactors the AIEA level 2 decommissioning status has been achieved.

- Fast breeder liquid sodium cooled reactors: Rapsodie in Cadarache and Phenix in Marcoule, the last one in operation until 2009. Phenix is an experimental reactor now used for long-lived isotopes transmutation experiments. Graphite is used as neutronic shield in some parts of the core.
- Piles EL2 and EL3 located in Saclay were experimental reactor moderated with heavy water. For these two reactors, the AIEA level 2 dismantling status has been reached.
- ULYSSE reactor, located in Saclay is a small experimental and teaching reactor (100 kW) moderated with graphite and cooled by light water. This reactor was shutdown in 2007.

The main characteristics of CEA reactors using graphite to be decommissioned are summarized in table 3.

Deleted: ¶
¶
¶
¶
¶
¶
¶
¶
Table 3 : **Main characteristics of CEA reactors to be decommissioned and containing irradiated graphite.**

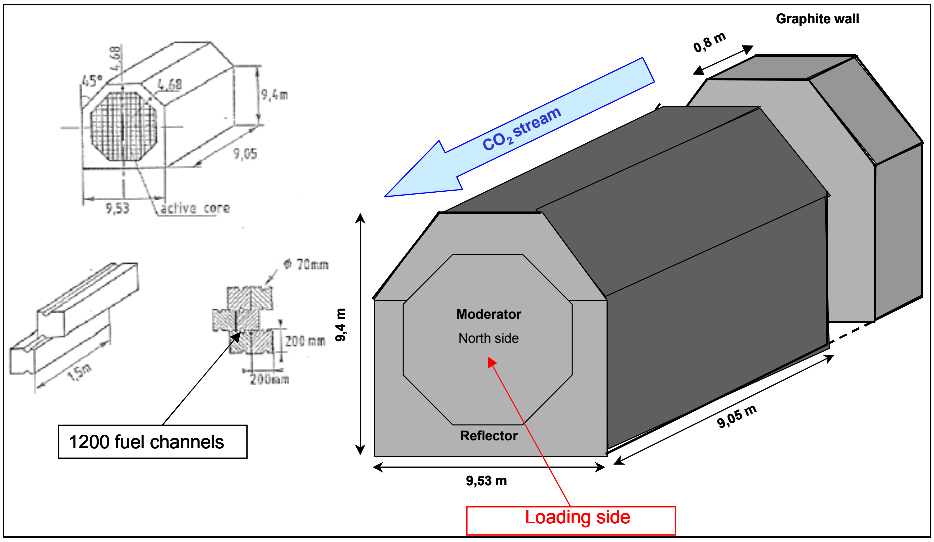

Figure 11 : General scheme of G2 reactor (G3 is identical) [21].

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

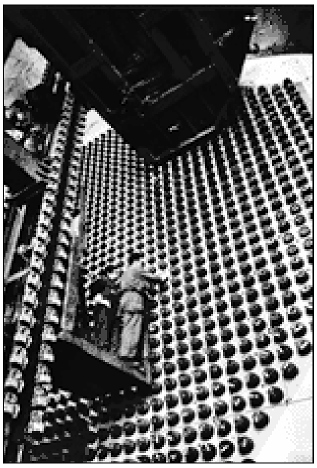

Figure 12 : Photography of G2 reactor in Marcoule - © Copyright CEA / Pierre Jahan [22]

4.1.6 Silos containing graphite sleeves

Graphite sleeves were used in CHA2, CHA3, SLA1, SLA2 and BU1 UNGG reactors. The operating time of these sleeves in reactor was of about 2 nominal power equivalent years. Except for sleeves from BU1, they have been temporarily stored in silos as reported in Table 4. According to a special agreement, each owner of the site where these sleeves are stored is considered as the producer. For this reason, a third actor, that did not operate any graphite reactor, appeared as an irradiated graphite producer in France: AREVA NC in La Hague (Figure 1).

Deleted: Figure 1

Formatted: Font: (Default) Times New Roman, Not Bold

The sleeves used in BU1 are already located in their definitive disposal: 85 % are in the Andra Manche waste facility disposal, and 15 % in the Andra Aube waste facility disposal. The Andra Manche waste facility disposal is now closed.

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

Table 3 : Main characteristics of CEA reactors to be decommissioned and containing irradiated graphite.

CEA Reactors	G1	G2	G3	Phenix	Rapsodie	EL2	EL3
Type	Plutonium production			Fast breeder		Piles	
Criticality date	January 1956	July 1958	June 1959	August 1973	1968	October 1952	July 1957
Shut down date	October 1958	February 1980	June 1984	2009	1983	Sept. 1965	March 1979
Projected decommissioning dates	-	-	-	-	-	-	-
Location	Marcoule				Cadarache	Saclay	
Thermal power (MW)	46	250	250	563	40	2.8	20
Electrical power (MW)	2 [23]	38 [23]	38 [23]	250 [20]	-	-	-
Cooling (pressure MPa)	Air (0.1)	CO_2 (1.5)	CO_2 (1.5)	Liquid sodium	Liquid sodium	-	-
Mass of graphite (t)	1,200	1,300	1,300	59	12	57	52
Graphite temperature (°C) during operation	30-230	140-400	140-400	-	-	-	-

Table 4 : Silos containing graphite sleeves.

Owner	EDF	CEA	AREVA NC
location	Saint-Laurent	Marcoule	La Hague
Mass of graphite (t)	2,030	730	970
Origin	Sleeves from SLA1 and SLA2 reactors	Sleeves from CHA2 and CHA3	Sleeves from CHA2 and CHA3

5 Graphite characteristics

This section dedicated to graphite characteristics deals mainly with graphite for UNGG reactors. Only one referenced paper [21] has been found on graphite from other reactors.

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

5.1 Virgin graphite

Nuclear graphite stack and sleeves were produced for EDF and CEA UNGG reactors by the SGL (former Pechiney) factory in Chedde (France). Some sleeves were also produced by a Savoie Acheson (now Graftech UCAR SNC) factory located in Notre-Dame de Briançon (France). Raw materials from different origins were used.

Graphite was single impregnated for the graphite stack and double impregnated for the graphite sleeves, in order to obtain the specified density. Fluoride compounds (NaF, MgF_2) were used to purify graphite to obtain a nuclear grade graphite. The improvement of the manufacturing process led to increasingly pure graphite with lower cross sections as shown in Figure 13.
For EDF graphite manufacturing, three petroleum cokes have been identified: Lockport L, Lockport M and Lima. Lockport L and M were produced by a Texaco company refinery (now closed) located in Lockport (Illinois - USA), and Lima coke was produced by a Sohio company refinery located in Lima (Ohio - USA). The pitch used as binder and for impregnations was from different origins and produced (purified) in European factories (Libercourt in France, UCC in United Kingdom) [15].

The main known physical characteristics of virgin graphite for the stack and for the sleeves for EDF UNGG reactors are reported in the Table 5.
In the case of CEA UNGG reactors, at least 2 to 3 different graphite were used in each reactor depending on their function (moderator, reflector, wall) [17] :

- a graphite made of special grade A coke was used mainly for the moderator in G2 and G3 ;
- a graphite made of special grade B coke was used for the moderator in G1 ;
- Lockport L graphite made was used as reflector or walls for both reactors.

The impurities contained in the virgin graphite were determined in some cases. Published data on chemical impurities in EDF graphite are presented in Table 6. One can notice that in most cases, there is no available data concerning chlorine and nitrogen content, both precursors of long-lived radionuclides (Cl-36, C-14) in i-graphite.

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

5.2 Irradiation history

The operating history for each GCR except for G1 is available in [28]. The cumulated produced energy is presented in Figure 14.

Deleted: Figure 14

Formatted: Font: Not Bold

These data are not directly related to irradiation history of graphite. Irradiation history of graphite stack depends on operating history of each reactor of course, but also on graphite location and on the neutron flux cartography in the stack (itself depending on operating parameters of the reactor : history of control rod position). Numeric calculations are needed to determine the irradiation history of graphite for each reactor. These data are not published.

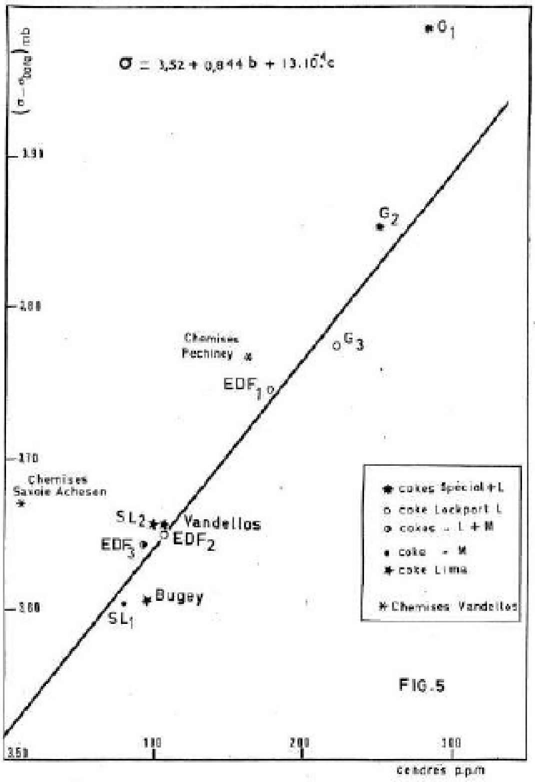

Figure 13 : Cross section versus ashes for French graphite - EDF1, EDF2 and EDF3 stands for CHA1, CHA2 and CHA3 - SL1, SL2 stands for SLA1 and SLA2.

5.2.1 I-graphite

The Wigner energy effects depend mainly on the irradiation temperature of graphite. Below 115 / 120°C, the accumulated Wigner energy is very high and spontaneous heat release can

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

occur. Above 350°C, no Wigner energy is accumulated. Wigner energy concerns only the low working temperature UNGG reactors, mainly UNGG CEA reactors (especially G1) and piles.

The weight loss of graphite stack induced by radiolytic corrosion has been monitored for BU1 [24]. This phenomenon was very important in BU1 because of its high specific energy and of the cooling gas pressure much higher than in other UNGG reactors. A monitoring of the graphite stack corrosion presented in Figure 15 shows an increase of the weight loss during operation that led to a 35 % weight loss in 1991 (3 years before the reactor shutdown). It leads to a dramatic decrease of its compressive strength that reached about only 30 % of its initial value (virgin graphite) in some parts of the stack. The loss of compressive strength of the BU1 stack was one on the reasons with economical one that prevailed on EDF to shutdown the reactor.

Table 5 : Physical characteristics of EDF UNGG virgin graphite [17] [27].

Reactor		Origin of petroleum coke used for graphite manufacturing	Ashes (ppm)	Cross section (mbarn)	Density g/cm^3)	Compressive strength (MPa)	
						(//)	(⊥)
CHA1		Lockport L 1 impregnation	178	3.86	1.696	40	-
CHA2	Stack	Lockport L 1 impregnation	106	3.75	1.671	36.5	36.3
	Sleeves	Lockport L 2 impregnations	89	-	-	-	-
CHA3	Stack	Lockport L 1 impregnation	107	3.75	1.700	40.1	-
		Lockport M 1 impregnation	121	3.74	1.669	-	-
	Sleeves	Lockport L 2 impregnations	118	-	-	-	-
SLA1	Stack	Lockport M 1 impregnation	80	3.69	1.683	38.5	-
	Sleeves	Lima 2 impregnations	162	-	-	-	-
SLA2	Stack	Lima 1 impregnation	98	3.76	1.684	43.7	-
	Sleeves	Lima 2 impregnations	162	-	-	-	-
BU1	Stack	Lima L 1 impregnation	98	3.74	1.685	40.6	38.0
	Sleeves	Lima 2 impregnations	162	-	-	-	-

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

Table 6 : Chemical impurities in EDF UNGG graphite [17][27].

	Stack						sleeves		
Impurities (ppm)	CHA1	CHA2	CHA3	SLA1	SLA2	BU1	CHA2	CHA3	SLA1-SLA2-BU1
Boron (10^{-3})	137	118	110	91	110	110	130	120	176
Hydrogen	-	20	-	-	20	20	20	20	-
Lithium	0.15	0.03	-	0.08	0.07	0.01	0	0	0.100
Vanadium	30	26.6	30	-	-	-	17.6	7.6	-
Calcium	23	0.63	-	-	-	5	5.2	4	-
Sodium	14	2	-	-	-	11	0.9	0.8	-
Potassium	-	-	-	-	-	11	1.4	0.9	-
Magnesium	-	0	-	-	-	-	2.1	-	-
Iron	0	4.1	-	-	-	7	3.7	5	-
Cobalt	0.04	-	0.54	-	0.05	-	0.029	-	0.08
Titanium	0	0.75	-	-	-	-	0.2	-	-
Chlorine	6	10	-	-	-	-	12	-	-
Dysprosium (10^{-3})	0.65	1	-	-	0.75	0.45	0.85	0.22	-
Europium (10^{-3})	-	0.3	-	-	0.5	0.62	0.12	0.8	-
Samarium (10^{-3})	25.2	5.2	-	-	1.5	3	1.8	0.73	-

Table 7 : Radioactive inventory for graphite waste according to [3].

Assessed activity in 2020 (Bq)	Sleeves in EDF silos	Sleeves in AREVA silos	Sleeves in CEA silos	EDF graphite stack	G1 (CEA) graphite stack	G2-G3 (CEA) graphite stack	EL2-EL3 (CEA)
α emitters	$1.2\ 10^{12}$	$5.6\ 10^{11}$	$4.3\ 10^{11}$	$3.4\ 10^{13}$	$3.0\ 10^{10}$	$5.8\ 10^{9}$	$1.1\ 10^{10}$
Short-lived β and γ emitters	$7.6\ 10^{14}$	$3.6\ 10^{14}$	$2.8\ 10^{14}$	$1.1\ 10^{15}$	$8.1\ 10^{13}$	$1.6\ 10^{13}$	$4.4\ 10^{12}$
Long-lived β and γ emitters	$8.4\ 10^{14}$	$4.0\ 10^{14}$	$3.1\ 10^{14}$	$5.3\ 10^{15}$	$6.3\ 10^{13}$	$1.3\ 10^{13}$	$2.9\ 10^{12}$

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

5.2.2 Radioactive inventory

5.2.2.1 Preliminary radioactive inventory

A preliminary radioactive inventory has been established by Andra in 2004 with the help of the producers. It has been published in 2006 and it can be downloaded at the Andra website [3]. This radioactive inventory was based on various radiochemical measurements on samples. The inventory of each graphite stack has been assessed from the highest measured values. When the value for one radionuclide was unknown, the highest value measured in other reactors was taken into account. A summary of the published values concerning graphite waste is presented in Table 7. This inventory will be updated in 2009.

When considering into more details the measurements that were used to establish this inventory, one can notice that the radionuclides of significant importance taken into account are activation products H-3, C-14, Cl-36, Ni-63, and Co-60. Very small amount of fission product have been found. A major focus has been made on long-lived radionuclides C-14 and Cl-36.

CEA radioactive inventory of graphite concerns all its reactors but a major focus has been made on G2 reactor where H-3 is the main radionuclide (146 kBq / g), followed by C-14 (45 kBq / g), Ni-63 and Co-60 (≈ 6 kBq / g each). The amount of Cl-36 is 1.3 kBq / g. These values measured in 2007 are reported in Table 8.

For graphite sleeves, the situation is slightly different due to the steel wires (fils de selle) that were not, in most cases, separated from the graphite sleeves. The activation product of steel, Co-60, is the main radionuclide with a maximum value of 21 kBq / g . For the other radionuclides (Table 9), activity values are slightly lower than those measured in the graphite stack : these differences could originate from the origins of graphite, the irradiation history or the storage environment in silos.

For EDF UNGG reactors, a summary of maximum values measured in each reactor is presented in Table 10. Except for Co-60, measurements carried on BU1 reactor samples led to

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

the highest activity level of activation products. This result is not surprising because BU1 was the reactor with the highest specific energy.

Critical review of a radioactive inventory method based on the highest measurements

The method of global inventory assessment based on the highest activity values measured lead to a preliminary overestimated inventory. For final studies on disposal, it needs to be refined on more realistic basis for at least three main reasons :

Deleted: .

- The maximum values measured in one reactor are not representatives of other UNGG reactors. For example, for BU1 measurements on Cl-36, the mean value measured is of about 200 Bq / g whilst only of about some tens of Bq / g for SLA2 and CHA3;
- The measurements on graphite samples from a reactor present very important variations; this is the case for Cl-36 in BU1 where the measured values range from 1 Bq / g to 900 Bq / g. These variations originate both from physical phenomena (the graphite activation products depend on the localization in the stack - e.g. neutron flux) and from sampling conditions, unavoidable variations can occur only because the size of the samples (cores) is not representative of the material or because of the very low level of activity (traces) measured in some cases;
- It does not explain graphite activation behavior in the reactor.

Deleted: ¶
¶

Refining the radioactive inventory using a data assimilation method

The preferred way to assess the radiological inventory should be a direct activation calculation using the neutron flux cartography and a known amount of impurities inside the graphite. Unfortunately, this method can not be used, most of time because of a lack of data; this is the case for Cl-35 that gave rise to Cl-36. It was not measured on virgin graphite used in most UNGG reactors in the 70’s and the 80’s. When some data exist, the method leads in some cases to results that do not fit with measurements on i-graphite samples. This is the case for some radionuclides produced from volatile impurities such as N-14, the values measured many years ago on virgin graphite are certainly not representatives of the impurity level remaining in the graphite in operating conditions. Moreover, because C-14 originating from N-14 is mainly located on the external and internal (pores) surface of the graphite and not in the graphite lattice, C-14 loss in the cooling gas happened during operation [37].

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

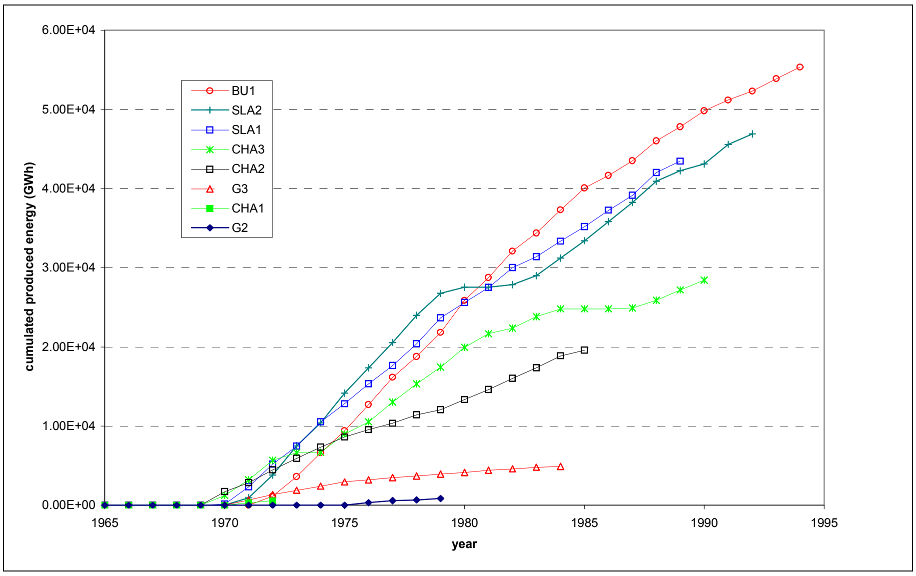

Figure 14 : Cumulated produced energy (GWh) for French GCR

A new method based on data assimilation and activation calculation is being developed in EDF [30]. It consists in an assessment of impurities levels in virgin graphite that gave rise to the remaining radionuclides within graphite stack after shutdown. It is based on statistical analysis of measurements carried out on samples and on the adjustment of activation calculation taking into account the neutron flux cartography (calculated - operating history) and an assessed initial concentration of impurities (N-14 for C-14 for example) able to explain the measurements.

Preliminary results obtained with this method for BU1 stack radioactive inventory are presented in Table 11. For each radionuclide, the result is expressed in term of mean calculated value ("TBq centré" in Table 11) for which the initial concentration of impurity is assessed in order to minimize the difference between measurements (mean value) and calculation. A second calculated value named as "TBq majoré" in Table 11, corresponds to an overestimated value of the initial impurity concentration taking into account the margin of variation of the measurements.

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

Table 8 : G2 reactor activity measured values in January 2007 [21].

	Activity (Bq/g)	Contribution to the total activity (%)
^{3}H	146520	71.3
^{14}C	44790	21.8
^{63}Ni	6480	3.2
^{60}Co	5720	2.8
^{36}Cl	1300	0.6
$^{152}Eu + ^{154}Eu + ^{155}Eu$	324	0.2
^{137}Cs	178	0.1
^{133}Ba	111	< 0.1
α emitters	105	< 0.1
^{90}Sr	28	< 0.1
Total	**205556**	**100.0**

Table 9 : CHA2 / CHA3 graphite sleeves activity measured values in January 2007 [21].

	Activity (Bq/g)	Contribution to the total activity (%)
^{60}Co	20670	53.8
^{3}H	9080	23.6
^{14}C	8283	21.6
^{63}Ni	98	0.3
^{90}Sr	81	0.2
^{137}Cs	70	0.2
$^{152}Eu + ^{154}Eu + ^{155}Eu$	59	0.2
α emitters	52	0.1
^{133}Ba	28	< 0.1
^{36}Cl	4	< 0.1
Total	**38425**	**100.0**

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

Table 10 : EDF graphite stack activity measured on samples (highest measured value for each reactor in 2007) - in bold the highest values taken into account for EDF graphite inventory assessment.

Radionuclide (kBq / g)	CHA1	CHA2	CHA3	SLA1	SLA2	BU1
H-3	140.0	60.0	247.0	-	426.0	**1,300.0**
C-14	14.0	44.0	47.8	64.6	-	**153.0**
Co-60	**1,470.0**	369.0	212.0	-	210.0	1,330.0
Ni-63	-	-	81.2	-	46.5	**243.0**
Cl-36	-	-	0.043	-	0.102	**0.900**

As shown in Figure 16, Figure 17 and Figure 18, this method enables the variation of radionuclide measurements depending on the sample localization (neutron flux) and lead to an assessment of radionuclide inventory able to take into account graphite activation behavior. The use of this method is in progress with a new sampling campaign on CHA2 and CHA1 reactors.

Deleted: Figure 16

Lessons to be learned from a data assimilation method : the example of ^{14}C

The interest of a data assimilation method to determine the graphite radioactive inventory does not only lie in a more accurate determination of the inventory, but it can also teach crucial information about the origins and the behavior of some radionuclide remaining within the graphite of a shutdown reactor. C-14 is a very typical example.

Deleted:

According to theoretical calculations, it is said in recent publications [17] [37], that the most significant element that give rise to C-14 in graphite in GCR is N-14. These calculations are based on various assumed N-14 content within graphite; for example, when the N-14 content is quoted to be 10 ppm, it is calculated that nitrogen accounted for 61 % of the total C-14 activity. When assuming such N-14 content (some tens of ppm) within the graphite of the BU1 reactor, calculation results does not fit at all with the measured total C-14 activity remaining in the graphite stack. The reason is certainly that, if such a N-14 content within graphite during operation is assumed, a vast majority of the C-14 that could have been produced by N-14 activation could have migrate and been lost in the cooling gas. The only way to take into account the different phenomena leading to the remaining C-14 activity in the stack (unknown

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

initial amount of impurities, activation, loss of radionuclides) is to carry out activation calculation with the known amount of C-13 (supposed to be non volatile because inserted in the graphite lattice) and a fitted amount of N-14 that can explain the amount of C-14 remaining inside the graphite stack after the reactor shutdown. With this method, it is shown that N-14 account for less than 4 % of the remaining C-14 in the graphite stack of BU1 [38].

5.3 I-graphite wastes retrieval

Published information on graphite reactor dismantling and graphite waste retrieval in France concerns EDF UNGG reactors and more especially, the Bugey 1 (BU1) reactor [26]. BU1 is intended to be the first of the UNGG reactors to be dismantled, followed by others at St Laurent and Chinon.

EDF strategy for these reactors is to achieve AIEA level 3 decommissioning status (reactor vessel and internals removed) as soon as possible. The methodologies and techniques being developed for BU1 are intended to be applied across the other reactors where practicable to do so (CHA3, SLA1 and SLA2).

The safety report describing the proposed methodology and setting out the safety and environmental justifications for the methodology and equipment design has been submitted to the French safety organization. The license to dismantle BU1 is expected to be issued in the second half of 2008. The beginning of the dismantling operation (flooding of the main pressure vessel) is intended to start in 2012.

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

Table 11 : BU1 inventory assessment based on data assimilation and activation calculation [31]

RN	TBq centré	TBq majoré
_H__3	1,82E+02	5,42E+02
_C_14	1,17E+02	2,85E+02
NI_63	9,88E+01	1,89E+02
CO_60F	5,71E+01	1,63E+02
FE_55	5,10E+00	1,01E+01
AR_39	1,28E+00	4,76E+00
CS137	3,78E-01	2,01E+00
EU154F	4,54E-01	1,62E+00
NI_59	7,84E-01	1,45E+00
CA_41	2,44E-01	1,02E+00
CM244	9,22E-02	7,33E-01
EU155	2,17E-01	7,23E-01
BA133F	3,36E-01	6,45E-01
CL_36	1,55E-01	5,93E-01
SR_90	7,69E-02	4,00E-01
CS134F	2,59E-01	3,12E-01
SB125	4,42E-02	1,94E-01
BE_10	4,43E-02	1,75E-01
PU241	1,59E-02	6,89E-02
NB_94F	3,28E-02	6,44E-02
HF178N	1,73E-02	5,76E-02
NB_93M	3,23E-02	5,14E-02
KR_85F	8,59E-03	5,02E-02
PM145	8,33E-03	3,13E-02
PM147	5,39E-03	2,54E-02
AM241	5,40E-03	2,31E-02
TL204	3,73E-03	2,11E-02
SN121M	4,70E-03	1,97E-02
PT193F	1,50E-03	1,62E-02
CD113M	4,71E-03	1,59E-02
PU238	3,55E-03	1,57E-02
SM151	3,63E-03	1,14E-02
PU240	2,10E-03	8,90E-03
KR_81F	1,14E-03	5,97E-03
HO166M	1,28E-03	4,69E-03
PU239	1,14E-03	4,66E-03
MO_93F	1,76E-03	3,86E-03
CD109	6,89E-04	2,97E-03
AG108M	7,44E-04	2,52E-03
AM243	4,01E-04	2,26E-03
LA137	5,09E-04	2,01E-03
TC_99F	3,13E-04	1,32E-03
_K_40	1,65E-04	6,50E-04
RU106	9,31E-05	4,75E-04
CM246	3,06E-05	3,71E-04
CS135F	6,55E-05	3,07E-04

RN	TBq centré	TBq majoré
EU152F	5,03E-05	2,81E-04
CM243	4,75E-05	2,20E-04
TM171	2,08E-05	2,04E-04
PM146	5,28E-05	2,02E-04
TB157	4,36E-05	1,43E-04
PU242	2,30E-05	1,13E-04
ZR_93	2,21E-05	9,87E-05
TB158F	2,46E-05	8,70E-05
AM242M	1,68E-05	7,38E-05
CM245	7,48E-06	6,10E-05
SN126	7,36E-06	3,98E-05
TC_97F	9,61E-06	3,95E-05
NB_91F	2,42E-06	1,37E-05
SI_32	1,85E-06	1,12E-05
TC_98	1,53E-06	8,00E-06
PD107F	1,35E-06	7,07E-06
NA_22	6,73E-07	5,36E-06
SE_79F	5,89E-07	4,47E-06
AR_42	4,34E-07	3,31E-06
CF250	1,83E-07	3,23E-06
BI210M	6,46E-07	2,77E-06
HF182F	3,71E-07	2,22E-06
CF252	7,59E-08	1,41E-06
_U234	3,36E-07	1,33E-06
IR192N	1,38E-07	1,15E-06
BI207F	1,23E-07	1,09E-06
_I129	1,51E-07	7,52E-07
_U233	2,16E-07	7,15E-07
_U238	1,80E-07	6,63E-07
CF249	3,66E-08	6,30E-07
RE186M	1,35E-07	5,81E-07
BI208F	7,40E-08	3,28E-07
RH102F	5,47E-08	2,87E-07
TH228	6,45E-08	2,45E-07
_U232	6,28E-08	2,39E-07
NP237	5,26E-08	2,27E-07
FE_60	2,23E-08	1,40E-07
_U236	3,61E-08	1,35E-07
LU174F	3,38E-08	1,25E-07
OS194	9,46E-09	1,16E-07
CM248	3,46E-09	5,76E-08
RE187	1,26E-08	4,31E-08
AL_26F	4,61E-09	4,14E-08
PO209	5,18E-09	2,95E-08
EU150F	4,41E-09	1,84E-08
TA179	2,85E-09	1,47E-08

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

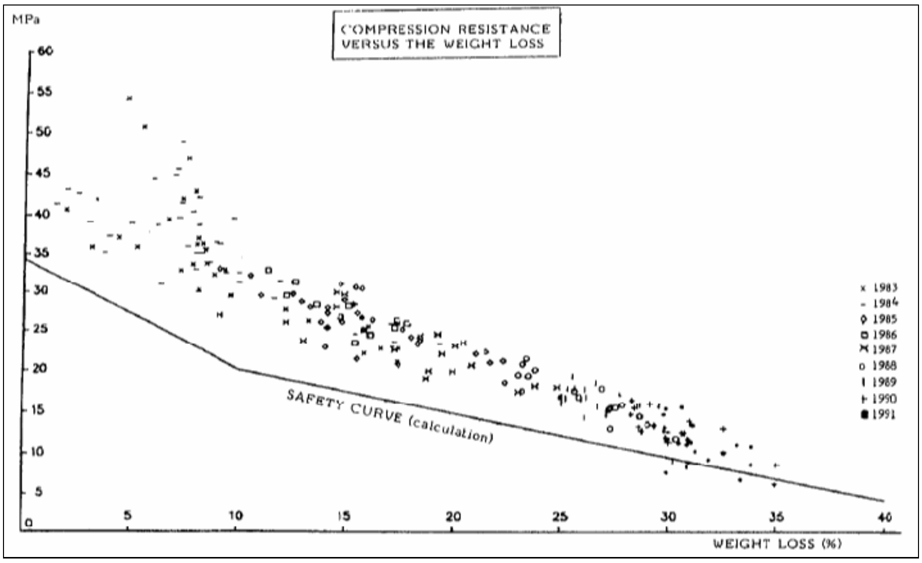

Figure 15 : Compressive strength of BU1 stack samples versus weight loss [24].

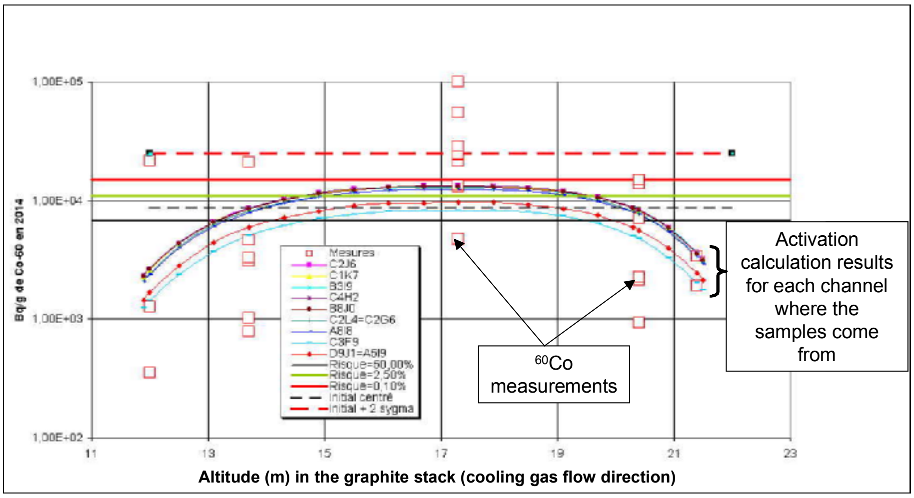

Figure 16 : Activation calculation results on BU1 stack with a data assimilation method : Co-60 [38].

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

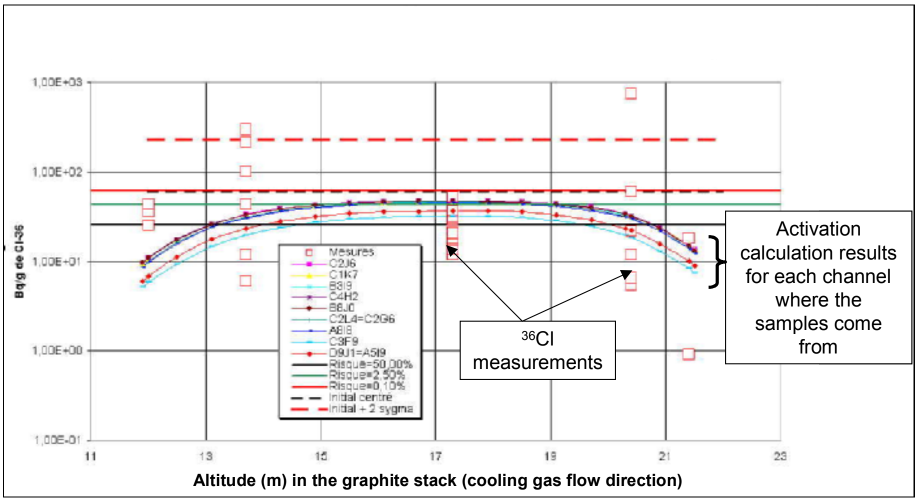

Figure 17 : Activation calculation results on BU1 stack with a data assimilation method : Cl-36 [38].

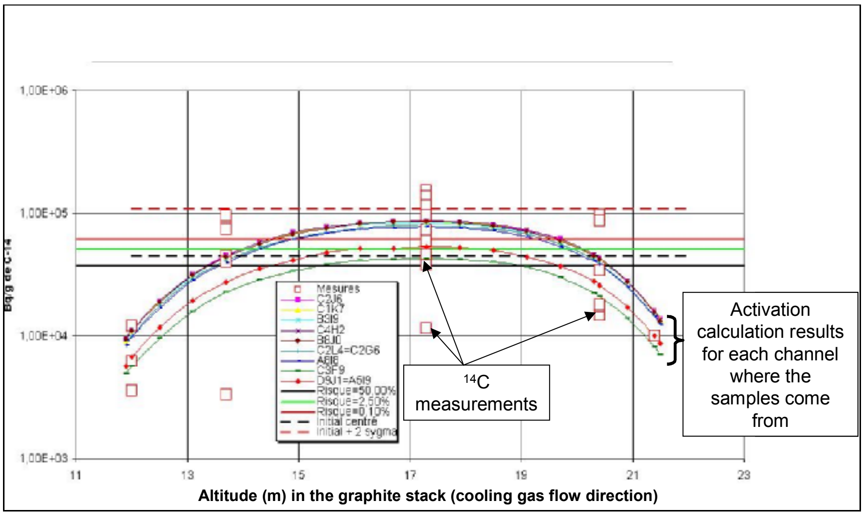

Figure 18 : Activation calculation results on BU1 stack with a data assimilation method : C-14 [38].

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

5.3.1 Underwater dismantling

The decision was taken by EDF that the dismantling works would be done underwater for the reactors for which water tightness can be assured and where the geometrical configuration enables this operation. It corresponds to reactors whose main pressure vessel is made of pre-stressed concrete, CHA3, SLA1, SLA2 and BU1. For the dismantling of CHA1 and CHA2, dry conditions will be used due to the specific geometry of the reactor vessel.

For underwater dismantling, the reactor is to be flooded before any intrusive work is undertaken. The reasons for this are several, and include:

- that the shielding afforded by the water would enable a more "hands on" approach to be adopted rather than a fully remote operation;
- greater flexibility to respond to unforeseen technical problems during dismantling, seen as particularly important when the size and the complexity of reactors is taken into account.

The amount of water required to fill in the reactor vessel is approximately 7,000 m^3. The time period during which the graphite stack will be under water, will last at least 2 years. A water treatment facility will be provided, located in the basement of the reactor building, in order to maintain water clarity, to remove dissolved activity leached from the graphite and also to regulate the chemical composition of the water.

The water treatment facility will produce additional wastes containing Cl-36 coming from the leaching of irradiated graphite. The studies of final disposal of these wastes (anionic ion exchange resins, filters and sludge) are in progress.

5.3.2 Reactor dismantling scenario

The different steps identified in the dismantling scenario are the following:

- *Previous works in order to make the main pressure vessel watertight* : sealing the gas circulator penetrations and the many other service penetrations for water, steam, instrumentation including cores which were cut through the sidewalls of the concrete wall to determine its activity profile.
- *Top Cap opening*: Following flooding, the top cap of the main pressure vessel will be

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

opened using a combination of coring and diamond wire sawing. The cap will be removed in two main stages; first the top 7 meters of cap will be removed and will then allow the dismantling platform (Figure 19) to be installed in the partial opening. Then the final 1.2 meters thickness will be removed, working from the dismantling platform. These two stages methodology provides for a separation of the none active upper layer of concrete from the activated concrete on the inner face of the caisson top cap, and also ensures the platform can be installed without excessive dose to construction workers.

- *Upper internals removal including graphite stack* : the sequence of the dismantling of upper internal removal is 1/ guide tubes, 2/ steel bricks which sit on top of the graphite core, 3/ steel links which tie the "corset" structure together and finally graphite core, corset bricks and corset support tubes coincidentally. The graphite bricks will be removed one by one, handle with semi-remotely deployed tools and placed in metallic baskets.
- *Sidewall concrete removal*: Once the entire reactor internals above the core support floor have been removed, a layer of concrete some 0.8 meters thick will be cut from the inside face of the caisson, including with it the steel liner and insulation. The water level in the caisson will be lowered as the concrete sidewall sections are removed.
- *Core support dismantling*: After removal of the sidewall concrete down to approximately 3 meters above the core support floor, the water level in the caisson will be established some 3 meters above the core support floor, and the platform lowered to just above the water level. The core support structure will then be dismantled working from the platform and using a variety of hand tools and semi-remotely deployed tools. Much of the structure requires to be cut up into relatively small sections in order to export them from the caisson through the platform. There is also a substantial quantity of graphite, the core support structure contains some 450 tons of graphite waste in the form of rods which must be extracted and separated from the steel components.
- *Second phase of sidewall concrete removal*: A further 8 meters high section of sidewall concrete, some 0.6 meters thick must be removed once the core support structure has been removed. The methodology to be used is identical to the first phase.
- *Lower internals*: The lower internals comprise the heat exchangers, pipe work, support skirt and support floor. Whilst obviously contaminated, they are not activated, and activity levels have been such that man access has been possible into the lower part of the caisson

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

several times during operations and after shutdown. Therefore it is intended to drain the caisson and perform the dismantling of all the lower internal structures manually.

- *Final demolition*: Once the caisson has been cleared of all internal components the intent is that the structure will be decontaminated using mechanical means, to be able then to monitor the remaining structure and declare it non-radioactive. Demolition using conventional means would then follow.

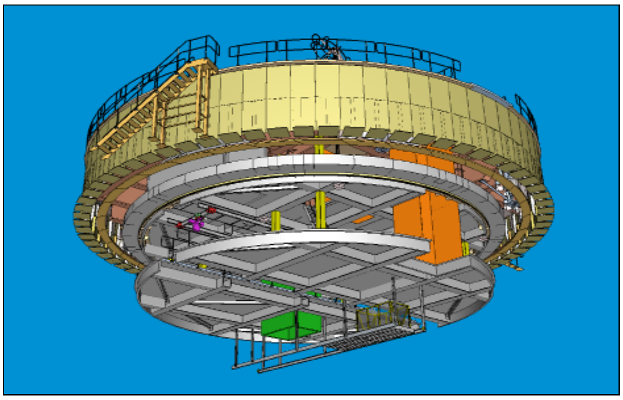

Figure 19 : 3D view of the dismantling platform: the support gantry is 25 meters diameter and weighs approximately 800 tons - the upper and lower decks below the support gantry are 18 meters diameter and weigh approximately 450 tons [26].

5.3.3 Containment requirements

The main shielding during dismantling operation is the water flooding the reactor. Shielding is also provided on the lower deck to ensure dose rates are low enough for unrestricted access. Additional shielding is provided on the support gantry to limit dose rates outside the site boundary in the unlikely event of a total drain down of the caisson, either accidentally or for maintenance reasons.

The dismantling platform comprises a gamma gate which gives access to a shielded shaft which connects through the lower deck to the waste basket carried on rails below the water level where the retrieved irradiated graphite is placed.

5.3.4 Graphite wastes management

EDF retrieved i-graphite will be grouted onsite without any preliminary treatment. The retrieved graphite wastes will be grouted onsite in concrete boxes in a waste grouting facility at

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

ground level. A temporary store for more than 100 packages is envisaged as a buffer between processing and transport off site to the disposal facility.

The package for graphite wastes direct disposal is being developed by EDF according to Andra technical specifications. It is a 10 m^3 concrete box (**Figure 20**) whose concrete formula is based on the F44 concrete formula already used in EDF for radioactive wastes disposal, with steel rebar or steel fibbers reinforcement. The graphite wastes in the metallic basket (Figure 21) will be loaded in the concrete box and grouted prior to capping. According to the shape of the graphite wastes to be packaged and their radioactivity level, it is assumed that 3 to 5 tons of irradiated graphite will be encapsulated in each package.

Figure 20 : Prototype of concrete package for irradiated graphite disposal : Total weight 24 tons - External volume 9.2 m^3.

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

Figure 21 : Metallic basket with graphite (non irradiated for development tests here) to be grouted inside the concrete package.

5.4 Treatment / conditioning techniques / process used on retrieved i-graphite

5.4.1 Leaching during underwater dismantling

The use of an underwater dismantling process will lead to the leaching of some radionuclides from the graphite stack during retrieval and to a decrease of the initial radioactive level of graphite stack. Some preliminary results aiming at assessing the impact of underwater dismantling on radioactive inventory of graphite stack are presented in this section. The objective is to assess the amount of radionuclides, particularly, long-lived radionuclides still remaining inside i-graphite after underwater retrieval and the amount that will be captured inside ion exchange resins used in the water treatment facility during retrieval.

Studies on graphite leaching have been carried out by CEA on EDF graphite sleeves and on EDF and CEA graphite stack samples. One must consider separately graphite sleeves from graphite stack because both the origins of graphite, its physical characteristics (density, open porosity) and the irradiation and temperature history are slightly different. This section is focused on graphite stack leaching experiments (graphite sleeves will not be submitted to underwater dismantling). Results on CEA graphite are reported because even if it is not known if these reactors will be dismantled underwater, the results obtained contribute to a better understanding of results obtained on EDF graphite stack samples leaching experiments.

For G2 graphite stack, CEA has carried out leaching experiments in pure water and in lime saturated water (pH = 12.5) at ambient temperature [32]. These experiments were carried out in

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

sealed reactor (Figure 22) in Pyrex glass completed with argon to prevent contamination, carbonatation and isotopic exchange. The samples used were coming from one core taken in 1989 in G2 reactor. They were homogeneous in term of spatial localization, radionuclides distribution (Figure 23) and porosity.

For the two long-lived radionuclides considered, Cl-36 and C-14, similar leaching results were obtained whatever the leaching solution (pure water or lime water) and whatever the sample considered. C-14 exhibits very low leaching rates (< 0.1 %) whereas for Cl-36, about 80 % of initial Cl-36 is leached in only 2 month (Figure 24, Figure 25). When considering the shape of the curve obtained for Cl-36, one can think that above this short period the Cl-36 leaching rate will not evolve anymore.

For BU1 graphite stack, leaching experiments were carried out on nine samples from various localization in the reactor [33]. Because of the different localization (irradiation history, temperature), a wide range of sample activities resulted. Different leaching conditions were used in term of leaching water (pure water, lime saturated water) and temperature (20°C, 40°C).
The results obtained for C-14 on BU1 graphite stack samples are very similar to those obtained on G2 samples (Figure 26). C-14 leaching rate is very low, the release fraction is less than 0.1 % and sometimes below the detection limits. These results accounts with the hypothesis of 2 existing kind (chemical form?) of C-14 in i-graphite:

- A C-14 produced from N-14 activation as an impurity from the cooling gas on the surface of graphite - This C-14 can be easily leached;
- A C-14 produced from C-13 activation or N-14 activation as an impurity in the graphite itself. This C-14 is inserted inside the structure of the graphite (sp^2) and can not be leached easily.

The very small amount of “leachable” C-14 found in experiments on BU1 graphite stack samples, is consistent with EDF activation calculation results showing that the remaining C-14 originates mainly from C-13 activation. Indeed, according to this activation calculation method

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

(section 5.2.2), the amount of acceptable N-14 impurity in graphite able to explain C-14 radiochemical measurements carried on samples, needs to be very low.

The results obtained on Cl-36 on BU1 graphite stack samples (Figure 27) are slightly different from those obtained on G2 samples. The cumulative released fraction of Cl-36 is very variable from one sample to another. It varies between about 20 % of the initial Cl-36 content in i-graphite to 80 % in some cases. Among the hypothesis that can be proposed to find an explanation, the temperature history of i-graphite seems to be one of the more interesting track. The results obtained on Cl-36 release on BU1 samples are reported in Figure 28, versus the localization, the height, of the samples, that is to say, versus the graphite working temperature.

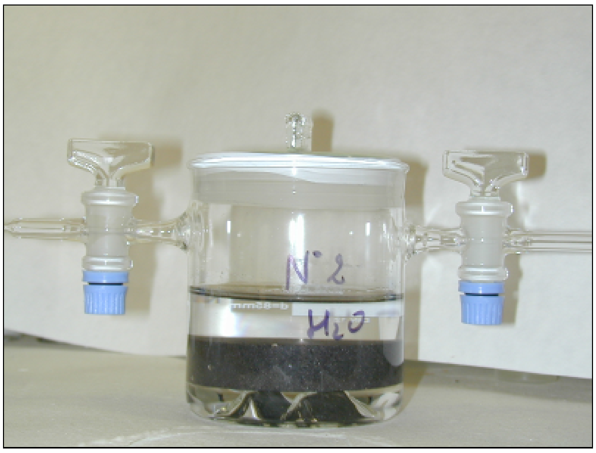

Figure 22 : Sealed reactor used for G2 leaching experiments [32].

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

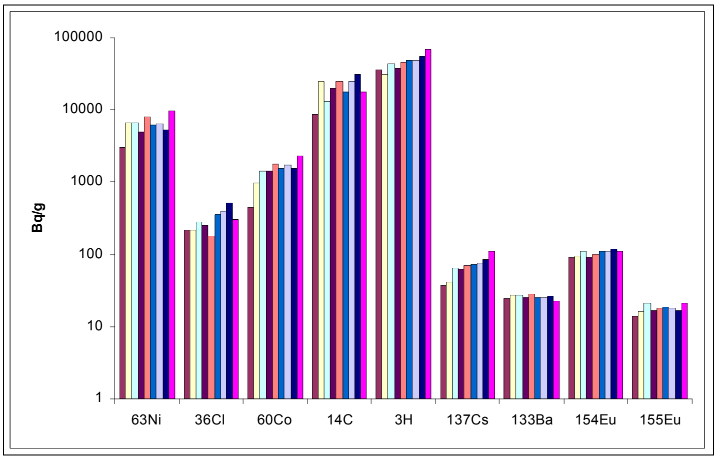

Figure 23 : Radionuclides distribution in G2 samples used in CEA leaching experiments [32].

One can notice a general trend in Cl-36 release versus working temperature: the lower the working temperature is, the higher is the Cl-36 release.

More studies are needed to explain the observed variation on Cl-36 leaching rate in order to assess the impact of underwater retrieval on graphite stack radioactive inventory. Studies on SLA2 graphite stack samples leaching are on the course.

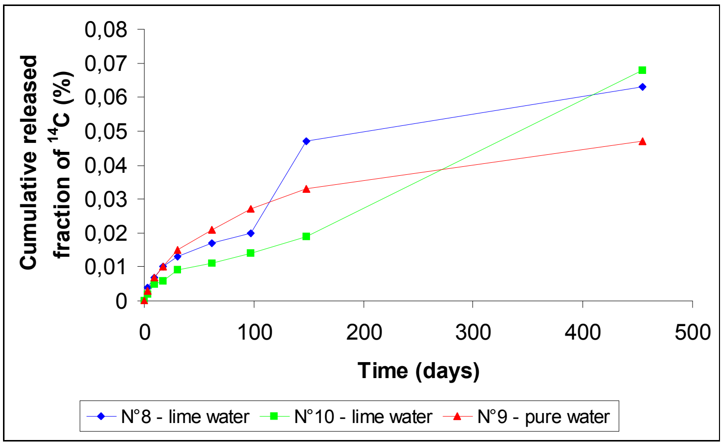

Figure 24 : Cumulative released fraction of C-14 in G2 leaching experiments [32].

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

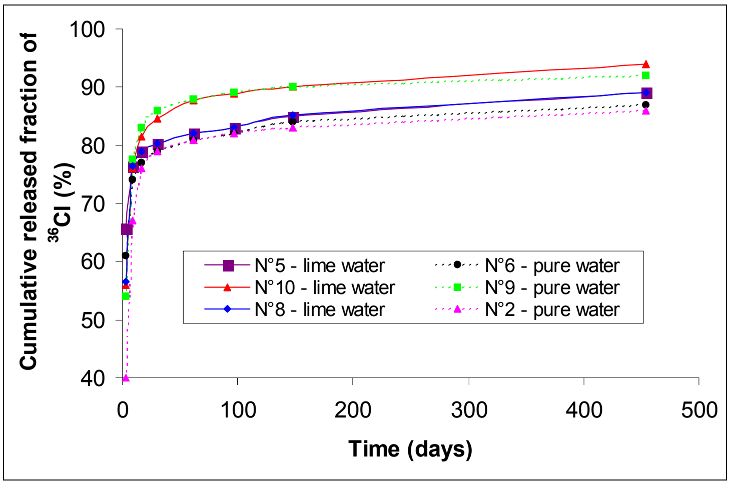

Figure 25 : Cumulative released fraction of Cl-36 in G2 leaching experiments [32].

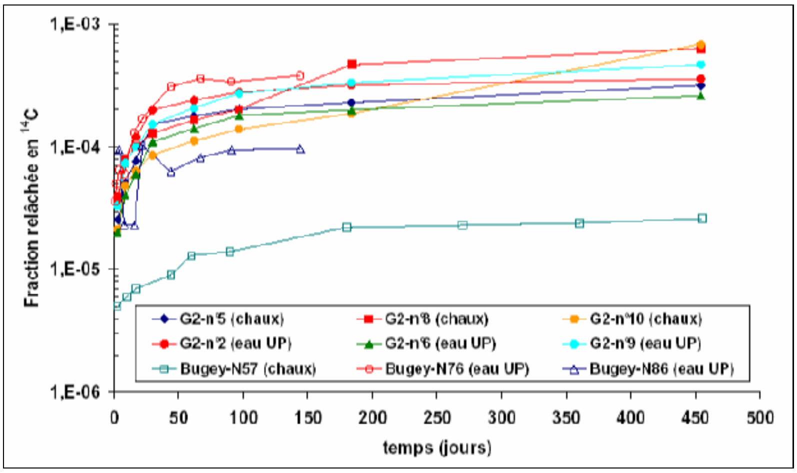

Figure 26 : Synthesis of C-14 cumulative release fraction for G2 and BU1 graphite

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

stack samples [33].

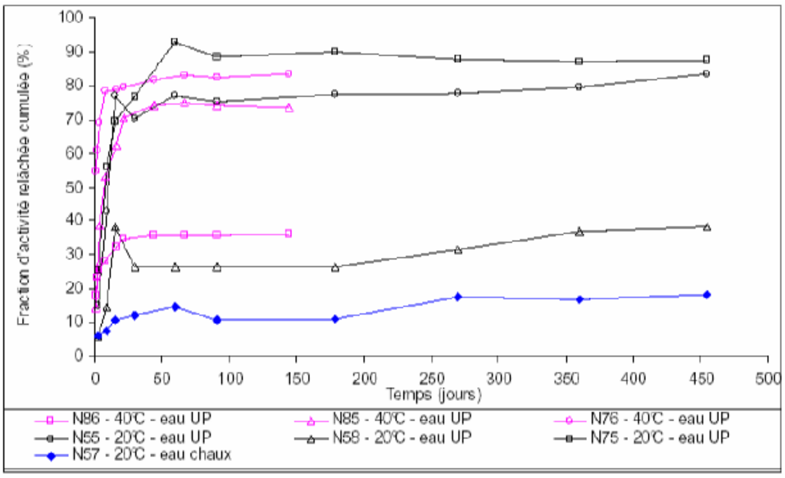

Figure 27 : Cumulative release fraction of Cl-36 in BU1 graphite stack samples [33].

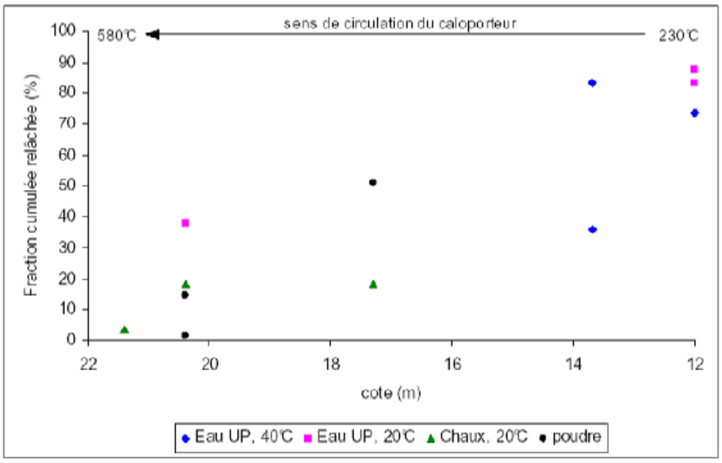

Figure 28 : Cumulated released fraction of Cl-36 in BU1 graphite stack samples versus the localization (height) of the sample in the reactor - 0 m corresponds to the top cap of the reactor - the top of the graphite stack (12 m) corresponds to the lower working temperature as the cooling gas flows from the top to the bottom of the reactor in BU1 (arrow “cooling gas flow”) [33].

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

5.4.2 Graphite impregnation with organic compounds

CEA has carried out research into immobilization of radioactivity on graphite using impregnation with epoxy resins and bitumen [34]. These studies were carried out in the eighties on samples from G2 reactor graphite stack.

The impregnation with epoxy resin was realized with a preliminary evacuation of air under vacuum followed by immersion at a pressure of up to 10 bar. A cure of the resin at 150°C followed. An increase in mass of around 12% indicated successful impregnation of the internal pore structures down to diameters of approximately 0.1µm, confirmed by X-ray tomography. For bitumen products, again, the graphite was evacuated before and then impregnation was realized at a pressure of about 2 bars, temperature 200°C during 30 minutes to achieve 3 to 7 % bitumen penetration.

Leaching tests carried out after impregnation have shown reduction in leaching rate by water of up to two orders of magnitude for the principle radionuclides. Even if this technique is considered to be able to immobilize the great majority of the radionuclides in short term experiments, it is very difficult to demonstrate its efficiency on long term disposal.

5.4.3 Graphite destruction by heat treatment

An incineration fluidized bed process has been studied by Framatome (Areva group) for graphite wastes incineration. A prototype incinerator was installed in Le Creusot and some tests were carried out in order to fit the best parameters needed to achieve complete incineration of graphite [36]. According to incineration efficiency, complete and controlled incineration was obtained with crushed graphite samples.

The incineration issue is dominated by the quantities of C-14 which are potentially released in the environment. Even if, provided some precautions, calculated radiological impact is low enough in term of the limited exposure of 1 mSv / year established for the general public [35], the use of incineration leading to the direct release of radionuclide in the environment remains not acceptable in France.

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

6 References

[1] Available in English on ASN website at http://www.asn.fr/sites/default/files/files/radioactive-waste-management-act-280606.pdf

[2] Available in English on Andra website at http://andra.12.kernix.biz/publication/produit/loi-VA-12122006.pdf

[3] Andra, "Catalogue descriptif des familles de déchets radioactifs" (in French), Dec.2006, http://www.andra.fr/pages/fr/menu1/l-andra/nos-missions/realiser-l-inventaire-42.html

[4] O.Ozanam, "Progress for the disposal project for LL-LL radioactive waste in France", 7th EPRI Int. decommissioning & radioactive waste workshop - Carbowaste session in Lyon, oct.2008.

[5] French Parliament website : http://www.parlement.fr/

[6] French government website : http://www.premier-ministre.gouv.fr/fr/

[7] DGEMP website : http://www.industrie.gouv.fr/energie/sommaire.htm

[8] ASN website : http://www.asn.fr/

[9] CEA website : http://www.cea.fr/

[10] CNE website : http://www.cne-recherches.prd.fr/

[11] IRSN website : http://www.irsn.fr/en/

[12] EDF website : http://www.edf.com/the-edf-group/presentation-of-the-group-95038.html

[13] CEA website : http://www.cea.fr/english_portal

[14] AREVA website : http://www.areva.com/servlet/group-en.html

[15] F.Berenger, « Projet graphite - document de base - connaissance générale des réacteurs UNGG », rapport EDF CIDEN ELIER0700644, 2007.

[16] P.Cornuault, « Génie nucléaire - modérateurs graphite », Techniques de l'Ingénieur B 3680.

[17] JP.Bonal, I.Bresard, S.Parraud, « Caractérisation du graphite des réacteurs de la filière UNGG », Matériaux, pp.1-5, 2002.

[18] MT.Pascal, « le démantèlement des centrales UNGG - gestion du graphite », conférence SFEN, 13 sept. 2007.

[19] A.Corcuff, A.Dionisio Gomes, « Le programme de déconstruction des centrales de première génération d'EDF », Revue Générale du Nucléaire (ISSN 0335-5004), 6, 34-39, 2003.

Deleted: http://www.asn.fr/sections/main/documents-available-in/act-no-2006-686-of-13

Deleted: http://www.andra.fr/publication/produit/loi-VA-12122006.pdf

Deleted: http://www.andra.fr/publication/produit/FichesFamilles.pdf

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

[20] D.Bastien, « French activities on gas cooled reactors », IAEA-TECDOC-899, pp:51-53, 1995.

[21] JP.Bonal, M.Butez, « Radiological inventory of CEA reactors graphite », 6th International decommissioning and radioactive wastes management workshop, Vienna (Austria), October 24th , 2007.

[22] http://www.cea.fr/energie/gestion_des_dechets_radioactifs_au_terme_de_la_l/un_demi_siecle_de_programmes_nucleaires_francais

[23] http://www.energethique.com/energie/NRGTiC_elnuc3083A.htm

[24] A.Petit, C.Phalippou, M.Brié, "Radiolytic corrosion of graphite surveillance and lessons drawn from the opération of the Bugey 1 reactor", Specialists' meeting on the status of graphite development for gas cooled reactors. Tokai, Ibaraki (Japan). 9-12 Sep 91 IAEA-TECDOC-690,265-272, sept.1991.

[25] D.Bastien, « Réacteurs à uranium naturel-graphite-gaz », Techniques de l'Ingénieur, B3180, 1993.

[26] F.Tardy, P.Lefevre, A.Willis, " Dismantling of Bugey 1", Communication SFEN Congress in Avignon (France), 29 september-1st October 2008.

[27] I.Bresard, JP.Bonal, « Caractérisations mécanique, chimique et radiologique des graphites de la filière UNGG », Article scientifique et technique DEN, 2000.

[28] http://www.iaea.org/cgi-bin/db.page.pl/pris.powrea.htm?country=FR&sort=NetElecCapacity%20DESC,GrossElecCapacity%20DESC,&sortlong=By%20Power%20Output

[29] JP.Bonal, F.Berenger, « La filière graphite gaz en France - connaissance du graphite », Réunion avec le Haut-Commissaire à l'énergie atomique, 28 janvier 2008.

[30] B.Poncet, « Data assimilation and dismantling wastes : a methodology customized for radioactive inventory assessment purposes », Waste Management 08 conference, Phoenix, February 24-28, 2008.

[31] F.Berenger, B.Poncet, L.Rahmani, « Inventaire radiologique Bugey 1, Graphite de l'empilement, des réflecteurs et de la protection biologique », Technical report EDF CIDEN ELIER/06 01237, jan.2007.

[32] C.Pichon, C.Guy, J.Comte, « ^{36}Cl and ^{14}C behavior in UNGG graphite during leaching experiments », Poster session Atalante conference 2008, 19-23 may 2008.

Deleted: to be presented at the

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc

[33] C.Pichon, « Behavior of radionuclides in UNGG graphite in aqueous media - State of the art - Review and program proposal », Technical report CEA NT SA3C/LARC 08-02, sept.2008.

[34] JR.Costes, C.De Tassigny, H.Vidal, « Conditioning of graphite bricks from dismantled gas-cooled reactors for disposal », Waste management, 10, pp.297-302, 1990.

[35] M.Dubourg, « The carbon-14 cycle », IAEA-TECDOC-901 pp.233-237, 1995.

[36] J.J.Guiroy, « Graphite waste incineration in a fluidized bed », IAEA-TECDOC-901 pp.193-203, 1995.

[37] BJ.Marsden and col., « The chemical form of carbon-14 within graphite », Serco Assurance technical report SA/RJCB/RD03612001/R01, march 2002.

[38] B.Poncet, « Data assimilation and dismantling wastes : a methodology customized for radioactive inventory assessment purposes for activated metal, concrete or graphite », 7th EPRI Int. decommissioning and radioactive waste workshop in Lyon, oct. 2008.

Deleted: CARBOWASTE D1.1.4 - FRANCE Issue 1 Draft for Review.doc