Study of Treatment as an Alternative WM Pathway: Examples of I-Graphite and Metallic Components such as Steam Generators – 15212

Jérémy Buttin *, Anne Deudon *, Rémi Duvivier *
* EDF

ABSTRACT

EDF is working continuously to optimize and secure its waste management pathways, leading in some case to develop appropriate treatment solutions that could be implemented parallel to reference solutions, complementary or as alternative solutions to already existing paths. This approach is illustrated by the examples of steam generators replaced during the operation of French PWR power plants, and of irradiated graphite (stacks and sleeves), coming from the decommissioning of UNGG reactors, which have been shut down.

Concerning graphite wastes, the near surface disposal facility intended for this waste does not exist yet, and is currently giving rise to the research of a suitable site, and an evaluation of its performances. While accompanying ANDRA (French national agency for nuclear waste management), in charge of the development of the facility, and while waiting for more data to be available, EDF is continuing the consolidation of graphite wastes radiological inventory, and is working on the development of an alternative management solution, based on a thermal treatment process.

Depending on the nature of the treatment, and on the choice of operating parameters, this treatment may contribute to reduce graphite wastes volume and/or promote its acceptance in a disposal facility. Reducing the radiological inventory by decontamination is one of the means to achieve these objectives.

Concerning replaced steam generators, nearly 200 of them will have been removed by 2030, and stored on site waiting for their treatment, which represents about 65 000 tons of metal. Initially intended to be cut and packed on NPPs' sites during their dismantling, EDF is studying the economic value of having the steam generators treated in a centralized facility, which allows reducing the volume of final wastes by optimizing cutting operations, while considering recycling after melting. However, this studied pathway implies a number of difficulties to be overcome. In particular, the shipment of such large components to a centralized facility may be complex. Moreover, French regulatory framework does not allow recycling outside nuclear industry, therefore limiting recycling opportunities to storage containers for radioactive wastes.

INTRODUCTION

From the 90's, in order to reduce the volume of its final wastes, EDF has chosen to develop and exploit, through its subsidiary SOCODEI, a suite of waste treatment methods (incineration and melting) which also allows the recycling of part of the metallic wastes. This real world example shows how treatment solutions can be fully integrated as part of the EDF industrial model of radioactive waste management.

With this experience, EDF is studying new waste treatment solutions with two complementary objectives:

- Optimize the management of a waste type and offer, when possible, an added value management option, as shown through the example of the recycling of metallic components,

- Secure a management path for a waste type by working on the development of alternatives to the reference solution, as shown through the example of graphite waste treatment.

MANAGEMENT OF GRAPHITE WASTE

Origins of Irradiated Graphite

The first generation of French nuclear plants _9 UNGG reactors, Natural Uranium Graphite Gas_ was operated from 1956 to 1994. These reactors were moderated with graphite, cooled with carbon dioxide (except G1 which was air cooled) and fuelled with natural uranium. Among these 9 reactors, distinction can be made between:

- 3 smaller reactors (G1, G2, G3), which belong to the CEA and were used for plutonium production,
- 6 reactors (Chinon A1, A2, A3 – Saint Laurent A1 and A2 – Bugey 1), which were operated by EDF for electricity production.

The dismantling of EDF's 6 UNGG reactors will generate about 15 000 tons of graphite waste, coming from the stacks and reflectors (see Table 1).

Table 1. Main characteristics of EDF UNGG reactors

UNGG reactor	Chinon A1	Chinon A1	Chinon A1	St Laurent A1	St Laurent A2	Bugey 1
Commissioning date	June 1963	February 1965	August 1966	March 1969	August 1971	Avril 1972
Decommissioning date	April 1973	June 1985	June 1990	April 1990	May 1992	May 1994
Electrical power (MW)	70	210	365	480	515	540
Mass of graphite (tons)	1050	2200	2670	3270	3220	2585

In addition, 2000 tons of graphite fuel sleeves, which were produced throughout Saint-Laurent reactors operation, belong to EDF. The total amount of graphite waste in France, that includes CEA reactors, is closed to 23 000 tons.

In France, graphite waste is classified as Long-Lived LLW. The radiological inventory of irradiated graphite (i-graphite) results of the activation under neutron flux of graphite itself (i.e. formation of C-14 from C-13 stable isotope), or from the activation of the very low amount of impurities that remains in graphite matrix after the manufacturing process. The two main long lived radionuclides contained in i-graphite and considered in France as important regarding long term safety are C-14 and Cl-36.

I-graphite Management Routes

In compliance with IAEA and French Nuclear Safety Authority (ASN) recommendations, EDF choose to dismantle these facilities as soon as possible. Therefore, a management solution for the graphite waste generated by dismantling operations has to be found, to allow EDF to safely dismantle these facilities within industrial and economical acceptable conditions.

According to the French law, since 2006 the reference for graphite waste management is direct disposal in a dedicated repository, whose conception, construction and operation is ANDRA's responsibility. In 2009, based on the initial radiological inventory of graphite waste, a first concept of 100 m depth disposal was envisaged by ANDRA.

Since recent EDF work on graphite characterization, based on a statistical approach, has lead to a significant reduction of its radiological inventory (especially Cl-36 which was previously widely overestimated due to the lack of data), a new concept of near surface disposal (15 m depth) is now studied. Development of such a facility is giving rise to the research by ANDRA of a suitable site, and evaluation (still ongoing) of its performances. Graphite waste producers (mainly EDF and CEA) are currently working, together with ANDRA, to demonstrate the feasibility from a long term safety point of view, of disposing of the graphite in a near-surface repository, through leaching behavior tests and contributions to a realistic evaluation of the long term impact of the facility (including transfers phenomena in both biosphere and geosphere). To support this reference route for graphite waste management, waste owners pursue their efforts to improve their knowledge of graphite waste inventory.

The National Plan for the Management of Radioactive Waste and Material (PNGMDR) provides that ANDRA have to submit in mid 2015 a report, which should contain a feasibility study of a near surface repository, the schedule associated with its implementation, and the nature of radioactive wastes that would be acceptable in it.

While waiting for this report, EDF, as a responsible waste owner, is studying alternative routes for graphite waste management. One of these alternative routes is based on a thermal treatment of i-graphite.

Thermal Treatment of I-graphite: Principle

In this context, a thermal treatment process may achieve two different objectives:

- Promote the acceptance of graphite waste in a sub-surface facility, by reducing the radiological inventory of graphite waste and / or have a beneficial effect on the release of long-lived radionuclides that have an impact on long-term safety.

- If this first step of decontamination is efficient enough, it may be followed by the full gasification of graphite matrix, with acceptable gaseous emissions. This option implies to achieve high decontamination, and is independent of the availability of a sub-surface disposal compatible with graphite waste.

The principles of graphite waste thermal treatment process is briefly summarize on the following figure.

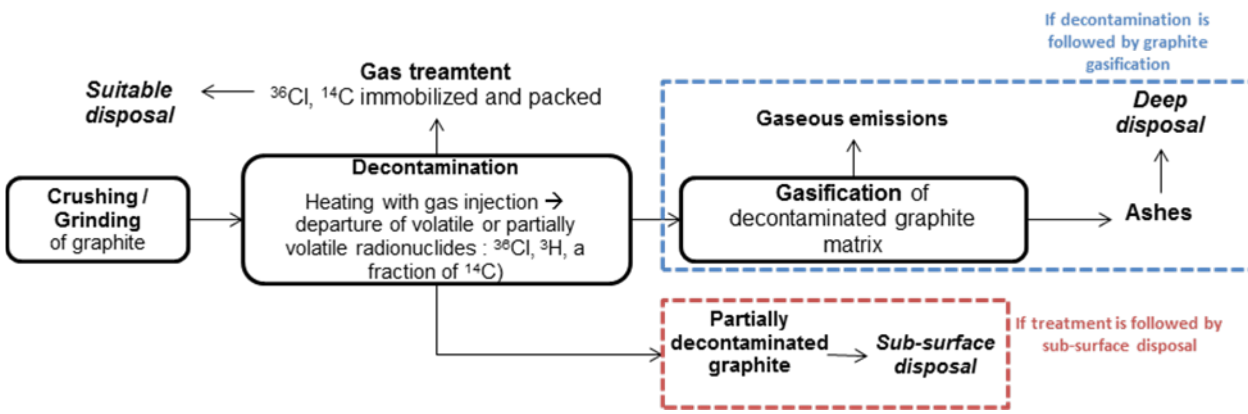

Fig. 1. Thermal treatment of irradiated graphite: principle

The first step consists of crushing the graphite. Reducing graphite particle size allows the thermal treatment to be more efficient by increasing the specific surface of graphite, create access for treatment gas into internal porosity and improving both mass and heat transfer in the thermal treatment reactor.

Then, the crushed i-graphite is sent into the thermal treatment reactor, wherein decontamination occurs. In this equipment, the combination of high temperature (≥ 1000°C), and the injection of a suitable treatment gas results in the release of part of the volatile (H-3, Cl-36) or partially volatile (C-14) radionuclides. During the decontamination step, these radionuclides have to be efficiently trapped, in order to minimize releases of radioactive species at the stack. Trapping the radionuclides lead to generation of a secondary waste that has to be packaged for disposal.

At these temperatures, graphite easily reacts with oxidizing gases and can be gasified into CO or CO2. If an oxidizing treatment gas is selected, its amount has to be precisely determined and controlled. The more the graphite is gasified, the more the C-14 released is diluted in the off-gas. Since the implementation of isotopic separation between C-14 and carbon stable isotopes may not be realistic, for both technical and economical reasons, reaching a high C-14 decontamination ratio while minimizing graphite gasification is necessary to avoid the generation of an unacceptable amount of secondary wastes. Achieving such a selective decontamination is the main challenged associated with the graphite treatment process.

A last step of full gasification of the graphite matrix could be the best way to drastically reduce the volume of waste. However, because during the gasification, trapping all the radionuclides (especially C-14 that has not been released during decontamination step) would generate huge amounts of secondary waste, off-gases treatment equipments would have to be partially by-passed. If the prior decontamination is not efficient enough, this would lead to unacceptable gaseous emissions at the stack. Therefore, depending on gaseous discharge permits that may be obtained from the Nuclear Safety Authority, implementation of full-gasification probably achieves high decontamination.

Ongoing Studies

To demonstrate the feasibility of a thermal treatment of irradiated graphite, and its industrial transposition, a project was initiated by EDF. In the scope of that project, and to support this demonstration, a contract was signed in 2009 between EDF and Studsvik Inc., for the realization of laboratory tests on French irradiated graphite. Other research programs, involving major French nuclear industry stakeholders, have been led parallel to this contract. These programs are focused on three main topics:

- The research on the best operating conditions to achieve the highest decontamination possible,
- The research (through a contract with ANDRA), on the best management options for secondary wastes.
- The demonstration of the industrial feasibility of the treatment of i-graphite (through the preliminary design by Studsvik Inc. of a thermal treatment prototype).

Current results show that a substantial decontamination in Cl-36 and a partial C-14 decontamination may be achievable. C-14 decontamination, within the operated conditions tested, is not selective enough to have confidence in the feasibility of a full-gasification of graphite waste. The main objective of the thermal treatment is thus to be a support to the acceptability of graphite in the near-surface disposal, when available.

Disposing of untreated graphite in a near-surface repository remains EDF’s reference for graphite waste management, and all the effort are being made to support this route.

MANAGEMENT OF METALLIC COMPONENTS

Origin and Inventory of Replaced Steam Generators

In a Pressurized Water (PWR) Nuclear Power Plants (NPP), steam generators insure the thermal exchanges between the water of the primary circuit, brought to high temperature in the reactor core, and the secondary circuit water, which is fed to the turbines after its vaporization.
Stress corrosion and mechanical wear phenomena are observed on tube bundles; therefore an important monitoring and maintenance program has to be implemented, to insure a satisfactory

level of safety, and a maximal operating ability. However, despite the implementation of technological improvements on French nuclear fleet steam generators, the replacement of part of the steam generators is considered as eventually inevitable. That is why EDF began in the 1990s a replacement program of its steam generators.
Currently, this program covers all the thirty four 900 MW reactors (corresponding to 102 steam generators – 3 for each reactor). In September 2014, 26 out of these 34 have already been performed (corresponding to 78 components), the last operation being planned in 2030.

Replacement program of steam generators of 1300 MW reactors has not been started yet, but should lead eventually to the replacement of 80 additional steam generators, corresponding to 4 steam generators for each of the 20 1300 MW reactors.
All replaced steam generators from both 900 and 1300 MW reactors will represent a mass of 65 000 tons, including about 80% steel and 20% Inconel®. Steam generators coming from the dismantling of NPPs at the end of their life time will be added to these 65 000 tons of metal.

Treatment Routes for Steam Generators

Once replaced, steam generators are stored on NPPs site in dedicated buildings equipped with reinforced concrete walls. The reference for replaced steam generators consists in safely storing these components until the dismantling of the NPP, to take advantage of radioactive decay, and of the presence on site during dismantling of treatment facilities dedicated to primary loops. The envisaged treatment consists in decontaminating, cutting and packaging the steam generators before their final disposal, in existing French repositories.
However, in order to preserve these facilities, and based on the international return on experience regarding recycling of metallic waste, especially steel coming from steam generators, EDF in mid 2011 began to look for a recycling management route for its replaced steam generators.

Recycling / Fusion Route: Principles

The route studied by EDF is based on the treatment of steam generators within a centralized facility, geographically accessible to large components, which would allows:

- Optimization of cutting operations (different scenarios, involving or not decontamination prior to cutting are envisaged),
- Melting of steel, for its recycling.

Because the French regulatory framework has no release threshold, recycling opportunities are limited to be within the nuclear industry. Currently, the primary target is the recycling through the manufacture of containers made of recycled cast iron, which could be used to pack Low and Intermediate Level Short Lived Waste.

The choice of a centralized treatment facility implies that transport operations from NPPs have to be made with neither decontaminated nor cut steam generators. Such operations may be challenging, because of the size and the radiological inventory of these components.

Ongoing Studies

Ongoing studies are focused on the establishment of all the technical and economical data required to decide whether or not EDF should implement a recycling route for steam generators. The realization of a full scale test in an existing facility in Sweden could be the first step to partially validate the feasibility and the associated challenges of such a waste management path.

Securing the outlet opportunities, through the demonstration of the acceptability of having recycled iron cast containers in a disposal and the study of technical and regulatory constraints associated with their use, is also necessary to validate the potential interest of this management path.

Studying the opportunity of extending the use of the centralized facility to other metallic waste is the next step beyond.

CONCLUSION

EDF is continuously working on the development of safe, sustainable and optimized new management routes for nuclear waste. In this framework, innovative waste treatment processes can be of great interest.

The first step to achieve an adapted and optimized solution is to have a good knowledge of the waste to be treated, in particular of their radiological inventories. Based on these data, EDF experiment and develop innovative treatment methods and processes.

Implementing a new treatment process also requires the development of an integrated industrial solution, which requires new infrastructures, to solve both industrial and logistics challenges, while evaluating the economical interest of the new path and taking in account regulatory constraints. EDF, as a major nuclear industry stakeholder, has the overall vision required to integrate each link of this complex chain. The two examples of graphite and metallic waste treatments are representative of this approach.

For graphite and steam generators, reference routes have already been identified, but until they are fully available, EDF’s will is to evaluate trough this approach, in relation with the stakeholders, every options to finally implement the best management path possible, within a constant concern about safety.