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Treatment of Residual Sodium and Sodium Potassium from Fast Reactors

Review of Recent Accomplishments, Challenges and Technologies

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TREATMENT OF RESIDUAL SODIUM AND SODIUM POTASSIUM FROM FAST REACTORS

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TREATMENT OF RESIDUAL SODIUM AND SODIUM POTASSIUM FROM FAST REACTORS

REVIEW OF RECENT ACCOMPLISHMENTS, CHALLENGES AND TECHNOLOGIES

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Marketing and Sales Unit, Publishing Section
International Atomic Energy Agency
Vienna International Centre
PO Box 100
1400 Vienna, Austria
fax: +43 1 2600 29302
tel.: +43 1 2600 22417
email: sales.publications@iaea.org
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For further information on this publication, please contact:

Waste Technology Section
International Atomic Energy Agency
Vienna International Centre
PO Box 100
1400 Vienna, Austria
Email: Official.Mail@iaea.org

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FOREWORD

In addition to the usual radiation and conventional hazards present during the decommissioning of disused nuclear installations, the presence of residual sodium or the alloy sodium potassium — used in primary, secondary and support systems in reactors using liquid metal as a coolant — presents additional technical, safety and cost challenges for decommissioning. This results from the propensity of these materials to react exothermically with water and moisture in the air potentially resulting in toxic and explosive reactions.

This publication discusses a variety of treatment methods to be considered when dismantling components that still contain residual quantities of sodium or sodium potassium, several of which were presented as contributed papers to the IAEA session during the 5th International Conference and Exhibition on Decommissioning Challenges, in Avignon, France, 7–11 April 2013. The publication provides a synthesis of information presented during the session, which was developed further at a consultants meeting held in Vienna, 2–6 December 2013. Decommissioning challenges faced at eight different facilities in five different countries are discussed, as well as the achievements and lessons learned that are of value to the worldwide decommissioning community.

The special session at the Avignon Conference on the treatment of residual sodium and sodium potassium was organized by the IAEA International Decommissioning Network, whose members also contributed to the drafting and review of this report. The IAEA officer responsible for this publication was P.J. O’Sullivan of the Division of Nuclear Fuel Cycle and Waste Technology.

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1. INTRODUCTION

1.1. BACKGROUND

Fast reactor development programmes in many countries were at their peak at the beginning of the 1980s. At that time, fast test reactors were operating in: France (Rapsodie); Germany (KNK-II); India (FBTR); Japan (JOYO); United Kingdom (Dounreay Fast Reactor); the Soviet Union (BR-10, BOR-60), and United States (Experimental Breeder Reactor, EBR-II, Fermi 1, Fast Flux Test Facility, FFTF, and Santa Susana Field Laboratory). Commercial size prototype reactors under construction or coming on line at that time included: Phénix, Superphénix (France); SNR-300 (Germany); MONJU (Japan); Prototype Fast Reactor (PFR) (United Kingdom); BN-350 (Kazakhstan/Soviet Union); and BN-600 (Russia/Soviet Union).

Subsequently, during the Eighties and Nineties, fast reactor development began to decline. By 1994 the Clinch River Breeder Reactor (CRBR) in the US had been cancelled and EBR-II had been shut down. The FFTF and BR-10 were permanently shut down in 2002. As of 2014, Phénix, Superphénix, BN-350, and PFR have also been shut down and are in various stages of decommissioning. SNR-300 was completed but never operated, and the site has been fully restored. Only the BOR-60 and BN-600 are currently operating at full power. The main recent development in this field has been the start-up and commissioning of the BN-880 fast reactor at Beloyarsk in the Russian Federation.

The use of liquid metal as a coolant in fast nuclear reactors[1] presents safety advantages as the need to keep the coolant under pressure is avoided and higher power densities may be achieved compared with the use of non-metal coolants. The high vapour temperature resulting from the high temperature of the liquid metal also leads to a higher thermodynamic efficiency as compared with water-cooled reactors. Disadvantages with the use of liquid metals result from the difficulty to inspect and repair the vessel, as the metal is opaque, fire hazard risks in the case of alkali metals, and the production of radioactive activation products in the coolant.

Freshly exposed sodium has a bright, silvery lustre that rapidly tarnishes, forming a white coating of sodium hydroxide (NaOH) and sodium carbonate (Na_2CO_3). Sodium belongs to the group of alkali metals, the most highly reactive inorganic elements; it does not exist in nature, but instead must be prepared from its compounds. Sodium is an excellent heat conductor, and like all alkali metals, reacts exothermically with water and moisture in the air, to the extent that, when exposed, sufficiently large pieces melt to a sphere and may explode. When in direct contact sodium and water generate heat, caustic smoke, and flammable hydrogen gas; such a combination can result in dangerous and pressure-producing reactions.

Sodium potassium (NaK) is an alloy of potassium and sodium that reacts in the same way as sodium when exposed to air or water, the main difference from sodium being that it takes a liquid form at ambient temperatures. In addition to its uses as a heat transfer medium, the NaK alloy is also used for gas purification, wave transducers, and as a barrier in hydraulic pressure relief valves.

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[1] Also used in submarine reactors.

The radioactivity of bulk sodium or NaK from primary circuits has several causes:

- Activation by neutron flux from the core and formation of radioelements, mainly ^{22}Na and ^{40}K (^{24}Na is also produced by activation, but it decays rapidly);
- Contamination by fission products from the fuel due to initial external contamination, fuel cladding failures during operation or use of leaking fuel pins: typically radioisotopes of actinides (plutonium, americium, curium and uranium), caesium and tritium;
- Contamination by activated corrosion products from fuel cladding and primary circuit structures, such as ^{54}Mn and ^{60}Co.

The specific activity of radioactive products in the primary coolant is highly dependent on the operational history of the reactor. Major contributors to the radiological inventory, as indicated above, are generally ^{137}Cs, ^{22}Na (^{40}K in NaK) and tritium [1].

Decommissioning experience relevant to the treatment of sodium and NaK used in liquid metal cooled reactors is generally concerned with one or more of the following three situations, with different treatment technologies generally being more effective depending on which situation applies:

(1) Bulk sodium (the fluid drained from the main cooling systems);

(2) Residual sodium (material left after draining); and

(3) Sodium in special components (such as cold traps, sodium vapour traps and caesium traps).

In 2007 IAEA published *'Radioactive Sodium Waste Treatment and Conditioning—Review of Main Aspects'* [2], discussing challenges arising during the maintenance and decommissioning of liquid metal fast reactors. Treatment and conditioning methods presented in that report described the best available technologies at the time of issue.

In 2009 IAEA published *'Decommissioning of Fast Reactors after Sodium Draining'* [3], which focused mainly on decommissioning of active loops, vessels, and components that have been drained of sodium, but had residual films and heels. This report addressed experience prior to that time for:

- Decommissioning and sodium treatment process development;
- Reactor and facility decommissioning experimental feedback;
- Sodium cooled fast reactor waste characterization and treatment;
- Decommissioning strategy for liquid metal cooled fast reactors.

1.2. OBJECTIVE

The purpose of this report is to document several treatment methods for sodium and NaK-contaminated waste types. These were presented in contributed papers to the International Atomic Energy Agency (IAEA) sessions at the French Nuclear Energy Society Conference at

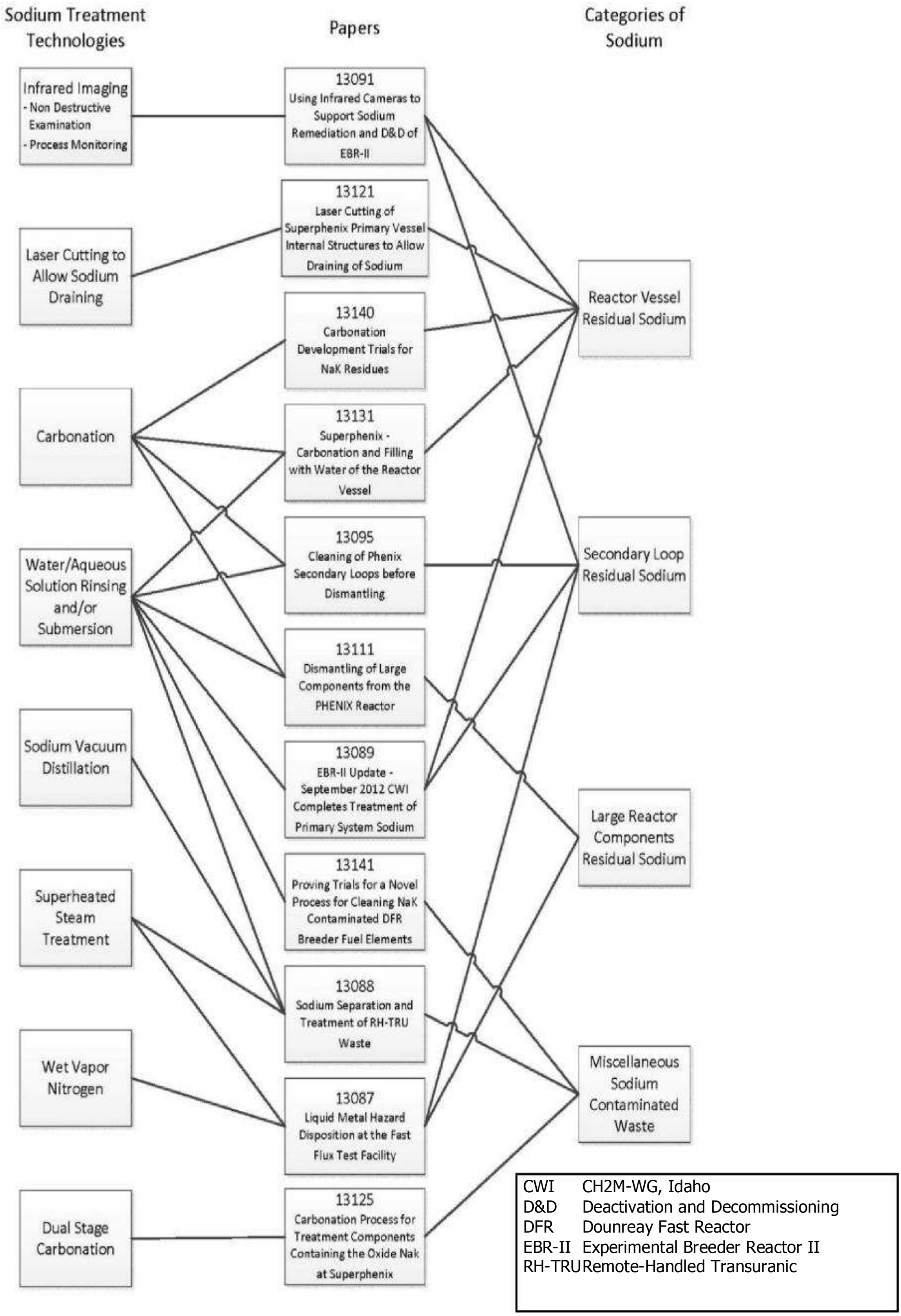
Avignon (April 2013) and developed further at a subsequent consultancy meeting at IAEA (December 2013). Figure 1 provides a visual relationship showing the categories of sodium and NaK treatment technologies cross-referenced to those IAEA session papers presented at the Avignon Conference. The report will assist those involved in the decommissioning of liquid-metal cooled reactors by providing an update on current practices being used for dealing with any residual sodium or NaK remaining in the reactor following the removal of the bulk coolant and by summarizing lessons learned from recent projects.

1.3. SCOPE

This publication covers a range of topics related to removal of residual radioactive sodium and sodium potassium (NaK) eutectics from reactor vessels and components in connection with decommissioning of liquid metal cooled reactors. The testing and application of relevant key technologies in five countries at a number of facilities are described. The technologies include passivation of the residuals, methods to control and monitor the processes, and also gaining access to difficult to reach locations where residual sodium may be found. Non-radioactive sodium waste is not specifically considered in this report. However, the approach, procedures, and recommendations that apply to safe management of radioactive sodium waste are also largely applicable to non-radioactive sodium. In addition to technical topics, lessons learned for management of removal and disposal of residual sodium and NaK are also presented.

1.4. STRUCTURE

Section 2 first defines categories of liquid metal contaminated wastes and provides a list of the treatment technologies, which are linked to the papers presented at the Avignon Conference. Section 2 also provides a brief summary of nuclear facilities by country with liquid metal cooled facilities and the sodium treatment technologies used at those facilities. Section 3 provides a brief description of facilities using liquid metal coolant and Section 4 provides an overview of the treatment technologies used to disposition the liquid metal hazardous waste. In addition to processing technologies, laser cutting techniques (4.9) and infrared imaging (4.10) are also discussed. Section 5 presents lessons learned related to management practices for decommissioning liquid metal cooled reactors. Appendices I and II, respectively, describe decommissioning experience at the BR-10 research reactor and BN-350 power reactor, providing additional material to that discussed at the Avignon Conference.



CWI	CH2M-WG, Idaho
D&D	Deactivation and Decommissioning
DFR	Dounreay Fast Reactor
EBR-II	Experimental Breeder Reactor II
RH-TRU	Remote-Handled Transuranic

FIG. 1. Treatment methods as related to waste categories.

2. SODIUM WASTE FORMS AND TREATMENT TECHNOLOGIES

Contaminated sodium and NaK may be found in several different locations or situations during the decommissioning of liquid metal cooled fast reactors:

- Reactor drained sodium and NaK;
- Drainable sodium from cold traps;
- Spent cold traps;
- Reactor vessel residual Na;
- Secondary loop residual Na;
- Large reactor components residual Na;
- Miscellaneous Na-contaminated waste.

The safe treatment of components containing residual sodium and NaK relies on having a thorough understanding of the mechanisms for sodium- and NaK-water reactions. Figure 2 shows the relationship between different types of Na-bearing waste and relative segregation, treatment, and disposal pathways.

This section of the report provides an overview of the technologies most widely applied for the management of sodium and NaK waste and gives a brief description of their use in the reactor decommissioning projects considered in this report. Details of the application of the treatment technologies are described in Section 4.

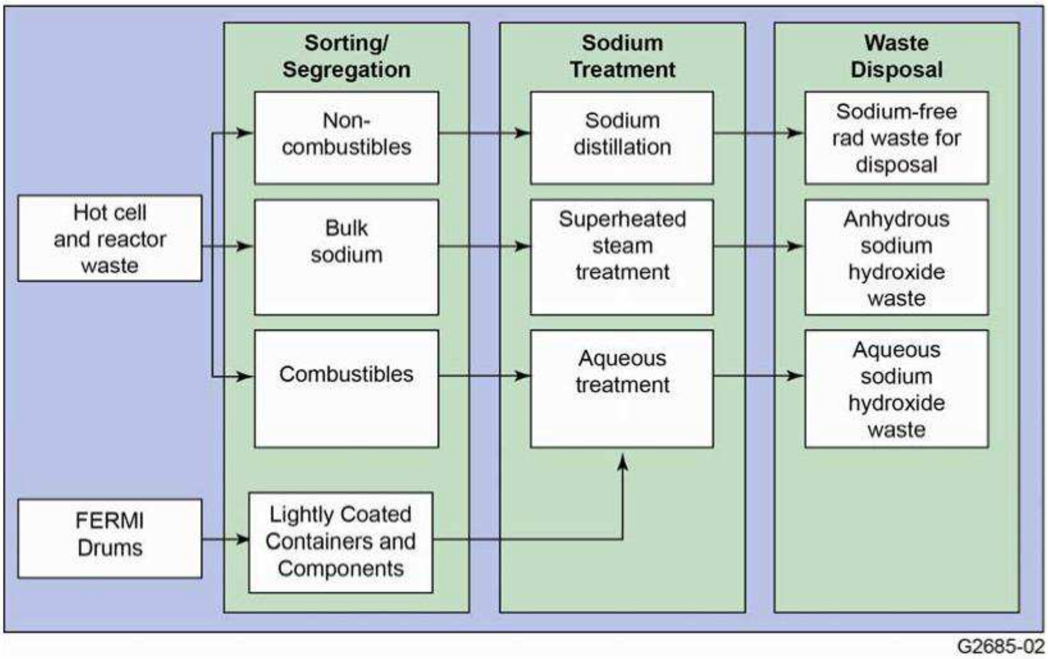


FIG. 2. Sodium-bearing waste sources to disposal pathways.

2.1. SODIUM TREATMENT TECHNOLOGIES

There are currently several sodium treatment technology options for treatment of different forms of sodium-bearing waste; those presented at the Avignon Conference included:

- Carbonation;
- Water/aqueous solution rinsing and/or submersion;
- Na vapour distillation;
- Superheated steam treatment;
- Wet vapour nitrogen (WVN);
- Dual stage carbonation;
- Solid-phase oxidation;
- Gas-phase oxidation by N_2O;
- Infrared imaging, non-destructive examination, and process monitoring;
- Laser cutting to allow sodium draining.

Treatments used by each of the subject facilities, by country, are briefly summarized below. Descriptions of the application of these technologies at reactors and other experimental facilities are presented in Section 4.

United States of America

Experimental Breeder Reactor (EBR-II), Idaho, USA. Separation and treatment of remote-handled intermediate level (transuranic) waste using sodium vapour distillation, superheated steam, and an aqueous spray wash system while monitoring material behaviour using infrared imaging.

Fast Flux Test Facility (FFTF), Washington, USA. Wet vapour nitrogen and superheated steam methods.

France

Phénix Nuclear Power Plant, Marcoule, France. Cleaning of secondary loops using carbonation and vaporous water circulation in secondary loops to generate Na_2CO_3, followed by rinsing the pipes to clean the Na_2CO_3.

Superphénix Reactor, Creys-Malville Nuclear Plant, Lyon, France. Laser cutting of primary vessel internal structures to allow draining of sodium retentions, development of a carbonation process for treating components containing oxidized NaK, and carbonation and flooding of the reactor vessel.

United Kingdom

Prototype Fast Reactor (PFR), Dounreay, Scotland (UK). The wet vapour nitrogen (WVN) process performed on secondary circuits at Dounreay prior to dismantlement was based on small Na-contaminated components cleaned in purpose built WVN facility at Birchwood.

Dounreay Fast Reactor (DFR), Dounreay, Scotland. Proving trials for a novel process for cleaning NaK-contaminated DFR breeder fuel elements comprising a sequence of atomized water spray, pulverization water spray, full water immersion, and a drying cycle, all carried out in a dedicated reaction vessel. In addition, the WVN process, carbonation process, carbonation development trials for NaK residues were performed.

Russian Federation

Research reactor BR-10, Institute for Physics and Power Engineering (IPPE), Obninsk, Russia. Solid-phase oxidation of bulk Na, NaK by slag. Gas-phase oxidation of residues Na, NaK (e.g. in cold and hot traps) by N_2O.

Kazakhstan

Fast reactor BN-350, Aktau, Kazakhstan. A six-loop 1000 MW(t) liquid metal cooled power reactor—steam/nitrogen process, carbonation process.

2.2. BRIEF SUMMARIES OF THE ELEVEN AVIGNON PAPERS

A summary of the eleven sodium treatment technologies presented in contributed papers to the special IAEA session at the 2013 Avignon conference, together with the additional material provided in Appendices I and II to this report, is provided below. This represents the source information for this report.

2.2.1. Liquid metal hazard disposition at the fast flux test facility (Paper 13087) [3]

A variety of technologies for remediation of residual sodium were evaluated for use at the Liquid Metal Test Reactor (FFTF), located at Hanford, USA, among them being wet vapour nitrogen, superheated steam (SHS), carbonation, incineration, and ammonia solutions. This paper presented FFTF's experience with sodium metal processing techniques and how they have advanced the efforts to select the methodology for proceeding with the residual sodium metal cleanout. The paper describes the procedure and details of the cleanout of the Composite Reactor Components Test Activity (CRCTA) vessel that is one third mock-up of the FFTF reactor vessel. This treatment used the SHS with nitrogen inerting for processing the residual sodium to produce sodium hydroxide and hydrogen gas.

The success of the CRCTA cleaning demonstrated that SHS is a major consideration for sodium removal at FFTF assuming that systems and components can be adequately preheated prior to introducing steam. Superheated steam was subsequently used to clean residual NaK in the FFTF Fuel Storage Facility NaK cooling system. The NaK residuals were completely reacted.

2.2.2. Sodium separation and treatment of remote-handled transuranic waste (Paper 13088) [4]

Sodium treatment technologies were evaluated to determine the best methods for treating the various categories of waste. Sodium vapour distillation was selected to remove sodium from non-combustible waste; SHS was selected to treat bulk sodium; and an aqueous spray/wash was selected to treat combustibles, containers, and components that are lightly coated with sodium.

The various designs described in the paper will be effective for treating about 90% of the sodium-bearing mixed waste in the Idaho National Laboratory (INL) site inventory. The same technologies could be applied to the remaining 10% of waste forms with significant modifications.

2.2.3. EBR-II update – September 2012, CWI completes treatment of primary system sodium (Paper 13089) [5]

Initial treatment of sodium residues in the EBR II systems using moist CO_2 gas safely treated much of the alkali metal; however, a significant volume still remained in the pipes, covered by a blanket of sodium bicarbonate. An aqueous solution of citric acid was used to dissolve the sodium bicarbonate layer, react the sodium, and finally neutralize the sodium hydroxide by-product. Treatment of the partially passivated sodium and NaK within the primary coolant tank and within its various auxiliary components was completed without incident. It was observed during these operations that that adding the citric acid treatment solution to an excess of sodium resulted in no significant pressure excursions.

Treatment of the secondary sodium system met with mixed results. Attempts to treat sodium in a network of pipes led to excursions of increasing intensity in conjunction with movement of treatment solution back and forth between branches. Ultimately, this resulted in a larger reaction of sodium and water moving a slug of water rapidly down a pipe and against a dead end, causing a breach of the piping by water hammer. Detailed lessons learned from this excursion are described in the paper.

2.2.4. Using infrared cameras to support sodium remediation and D&D of EBR-II (Paper 13091) [6]

This paper describes the development and application of infrared technology during sodium treatment at EBR II. One application was the nonintrusive identifications of the locations of residual sodium, including any complete blockages within the pipes. A second was safely monitoring the treatment process and verifying completion of the sodium reaction. Four infrared cameras were used, and the resulting pictures provided valuable research data, increased safety, and improved process feedback throughout each of the phases. Details of these applications are presented. The success at EBR-II demonstrated the benefit of using infrared cameras for these decommissioning tasks.

2.2.5. Cleaning of Phénix secondary loops before dismantling paper (Paper 13095) [7]

The Phénix sodium cooled experimental Fast Breeder Reactor at Marcoule operated from 1974 until its shutdown at the end of 2009. This paper describes decommissioning program phases for the three secondary loops: 1) their draining and other actions to prepare for carbonation; 2) the carbonation process consisting of nitrogen, carbon dioxide and water vapour circulation to generate sodium carbonate; and 3) final rinsing with water. The main challenges for each of these phases are described in the paper.

2.2.6. Dismantling of large components from Phénix reactor (Paper 13111) [8]

The PHENIX reactor was permanently shut down in 2009. The cleaning and dismantling preparation operations Phénix reactor were underway as of 2013. These operations include the removal and treatment of large removable components, such as primary coolant pumps, intermediate heat exchanger, and a heat exchanger blanking device, which is a dummy intermediate heat exchanger called the "DOTE". When installed, the DOTE provides a radiation shield and a leak tight argon seal between the primary and secondary sodium.

To reduce the dose and the cost, DOTE will be disposed of as a single part. The paper describes the major steps to accomplish this objective, which include removing the internal thermal barrier; decontamination by washing; characterization; gaining acceptance from the disposal site authority; preparation for disposal; and shipping to the *Centre Industriel de Regroupement, d'Entreposage et de Stockage* repository for very low level waste.

The DOTE resided for several years within the reactor. To be disposed as a single item, the DOTE dose rate and contamination needed to be compatible with very low level waste specifications. This was successfully achieved.

2.2.7. Laser cutting of Superphénix primary vessel internal structures to allow draining of sodium (Paper 13121) [9]

The draining of sodium coolant from the Superphénix reactor primary vessel will leave several sodium retention zones that cannot be drained by the main electromagnetic pumping system. Some of them, located in the area of the junctions between the primary pumps and the core feeding plenum (called “LIPOSO”), represent a significant quantity of sodium that could compromise the further water immersion of the reactor block internal structures. For this reason, a method to cut openings in the structures to drain the sodium retention zones was developed.

This paper describes the physical configuration and locations of the retained sodium, the choice of laser cutting technology, accessibility of the laser to the locations, considerations of the environmental conditions within the LIPOSO on the laser equipment, and the equipment design and testing.

As the laser technology for this application was novel, safety standards and rules were developed for the equipment and working methods, followed by training and conduct of their application. The operations in the conditions present were unique and first of a kind; testing revealed many problems that were previously unrecognized. Many lessons about the possibilities and limits of laser cutting were learned. Upon completion of the full-scale trials of the robotic handling of the laser equipment, the entire operation will be thoroughly rehearsed prior to use within the LIPOSO.

2.2.8. Carbonation process for treatment components containing the oxide NaK at Superphénix (Paper 13125) [10]

An experimental qualification program on the chemical reactions involved in the oxidized NaK treatment process was carried out from 2007 to 2010 through two sets of trials. The first trials verified dry carbonation reactions and evaluated their kinetics with NaK oxides and hydrated hydroxides. The aim of the second trials was to confirm the results of the first trials for potassium dioxide (KO_2) and for dry behaviour of sodium peroxide (Na_2O_2) and its hydrates. They also studied interactions with species combinations (oxides + hydroxides + hydrates) and with NaK. The second campaign completed a series of trials on the whole treatment process (dry and moist carbonation) on oxidized NaK at various temperatures.

The paper provides details of both sets of trials, the results, and lessons learned. Of particular interest was the finding that KO_2 preferably reacts during moist carbonation, contrary to what one might infer prior to having conducted these trials.

2.2.9. Superphénix–carbonation and filling with water of the reactor vessel (Paper 13131) [11]

Following removal of the reactor core, the Superphenix vessel was drained of bulk sodium and then flooded to eliminate sodium risks during dismantling. Flooding the vessel with its very complex internal structures necessarily involves management of the hydrogen produced (i.e., collection, dilution, confinement, etc.). The strategy of limiting reactions solely by modulating the reduction in the sodium inventory was found to be expensive and time consuming.

2.2.10. Carbonation development trials for NaK residues (Paper 13140) [12]

Flooding the Superphénix vessel with water provides shielding for dismantling of the very complex internal structures within the vessel described in the paper. A major concern was controlling the rate of hydrogen generation so as to eliminate the potential for ignition. This approach minimizes residual sodium, carefully controls the water injection rate, and renders the sodium inert with nitrogen.

Uncertainties with theoretical process rates and carbonation results, as well as the difficulty with modelling and limited operating experience to substantiate theory, would cast doubt on analytical safety analyses. Therefore, methods were developed to manage hydrogen and more quickly and completely wash the vessel. These include the important means for access to and washing of the space behind internal baffle plates and approximately twenty small component penetrations within the vessel. Combined with short carbonation durations, this approach makes dismantling the vessel with no hydrogen risk achievable with continuous human presence above the vessel water pool.

2.2.11. Trials for a novel process for cleaning NaK contaminated DFR breeder fuel elements (Paper 13141) [13]

As of 2013, approximately 1000 breeder fuel elements remained in the core of the Dounreay Fast Reactor. All of the fuel has a coating of residual NaK following drainage of the reactor. Some of this fuel is known to have failed during reactor operations. Specialist fuel element retrieval equipment and a dedicated process plant to remove this residual NaK and repackage the fuel has been designed and built in preparation for the campaign to deal with this fuel. This paper explains the process testing for cleaning of the fuel elements and the equipment designed for the testing. Details of the testing and lessons learned to be applied to the cleaning campaign are also described. Conclusions are presented regarding hydrogen control, effectiveness of a hydrogen monitoring systems, water misting injection nozzle design, and process duration, among others.

3. FACILITIES USING LIQUID METAL COOLANT

3.1. EXPERIMENTAL BREEDER REACTOR II, IDAHO, USA

The Experimental Breeder Reactor-II (EBR-II) was designed as a 62.5 MW(t), metal-fuelled, pool reactor with a conventional 19 MW(e) power plant. EBR-II operations began in 1964, and the plant was closed in 1994.

FIG. 3. EBR-II containment with the boiler building in the foreground.

The EBR-II project served to demonstrate the fast reactor fuel cycle with on-site fuel reprocessing. It was used as an irradiation facility for testing fuels and materials, and to show the passive safety of the design. It was on its way to completing a second fast breeder fuel cycle when it was shut down in October 1994 following the national decision to end the Integral Fast Reactor programme.

The early shutdown of EBR-II resulted in the absence of a planning phase that looked ahead to its decommissioning activities. Nevertheless, it was completely defueled by December 1996. By March 2001, sodium from both the primary and secondary circuits had been reacted to solid NaOH (73% by weight). The reactor EBR-II circuits were then passivated using carbonation. Fermi-I primary sodium was treated during the same campaign. All the passivated material was disposed as 1450 tonnes of low level solid radioactive waste.

The reactor system is now in a long term monitoring phase (safe enclosure), which is planned to last at least 50 years to allow radiation levels to drop. During this time the reactor is stored under an argon atmosphere and is constantly monitored.

3.2. FAST FLUX TEST FACILITY REACTOR, HANFORD, USA

The Fast Flux Test Facility (FFTF) was a 3-loop liquid metal cooled fast reactor located at the Hanford Site in Richland, Washington. It was designed for a thermal output of 400 MW and operated successfully from 1982 to 1992. The primary and secondary systems contained liquid metal sodium with several support systems containing NaK. All fuel was removed from the reactor in 1995.

All of the sodium and NaK have been drained with the NaK having either been reacted for disposal or mixed with the sodium inventory. The sodium systems contain less than 38 m3 of residual sodium that is maintained at an ambient temperature under an argon buffer gas blanket. Approximately 1136 m^3 of metallic sodium are located at the Hanford Site; this material is being retained for potential future use.

FIG. 4. The Fast Flux Test Facility at Hanford.
Photograph courtesy of the US Department of Energy, Richland Operations Office.

3.3. PHÉNIX, MARCOULE, FRANCE

The Phénix was a pool type reactor with three secondary loops located in Marcoule, France. It was designed for an output of 563 MW(t) and 250 MW(e) and operated successfully from 1974 to 2009. The primary and secondary systems contained liquid metal sodium with several support systems containing NaK. All fuel is scheduled to be removed from the reactor by 2018.

The primary system still contains bulk sodium (as of 2014) but the secondary system has been drained, with 450 m^3 of sodium having been removed. All but 6.1 m^3 have been processed. Approximately 1.3 m^3 NaK currently remains in support systems; it will be mixed with sodium when removed.

FIG. 5. Phénix. Photograph courtesy of CEA.

3.4. SUPERPHÉNIX, CREYS-MALVILLE, FRANCE

Superphénix, located in Creys-Malville, was a pool type reactor with four secondary loops. It was designed for an output of 1200 MW(e) and operated successfully from 1986 to 1997. The primary and secondary systems contained liquid metal sodium with approximately 20 subsystems containing NaK. All sodium has been drained from the secondary systems (325 m^3 for each of the four loops), the residuals removed by carbonation and dismantlement of the secondary loops is in progress. The bulk sodium has been drained from the primary system (3200 m^3) and treatment of the retention areas is underway. Carbonation will be used to remove the residuals followed by reactor vessel flooding in the 2016 time frame.

FIG. 6. Superphénix. Photograph courtesy of EDF.

3.5. DOUNREAY FAST REACTOR, DOUNREAY, SCOTLAND

The Dounreay Fast Reactor (DFR) was the first fast neutron reactor in the United Kingdom and was the first in the world to supply electricity to a national grid. It was built between 1955 and 1958, and in 1959 was commissioned and went critical. Although the DFR was an experimental reactor, it was built at a significant scale and was finally shut down for decommissioning in 1977.

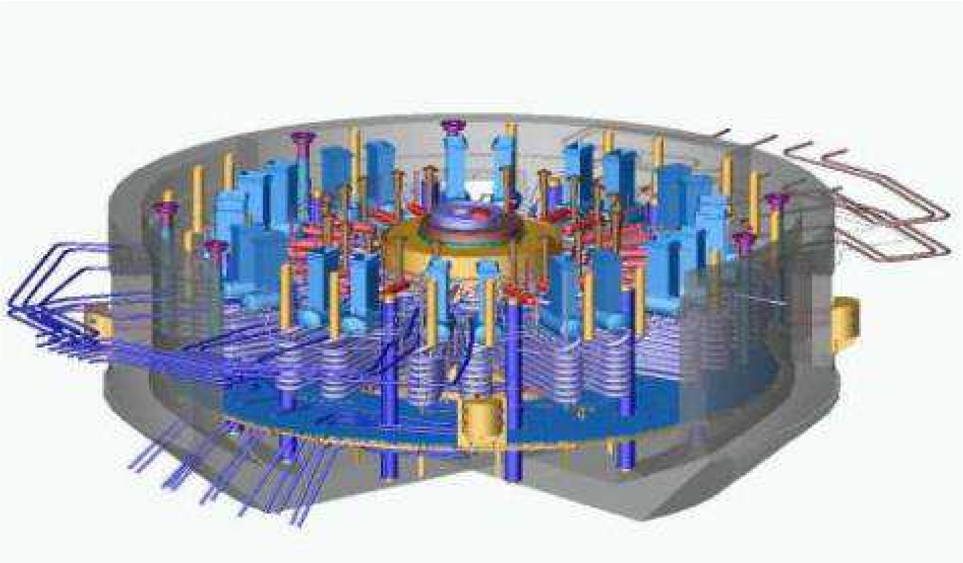


FIG. 7. Dounreay fast reactor. Photograph courtesy of Nuclear Decommissioning Authority (NDA) and Dounreay Site Restoration Limited (DSRL)

The DFR was a 24-loop liquid metal fast reactor located in the north of Scotland and was designed with a thermal output of 60 MW. Each loop has an associated secondary heat exchanger and decay heat removal heat exchanger. All the primary pipework is double contained either by the secondary and decay heat removal systems or by a leak jacket. In all there is over 9 km of pipework contained inside a biological concrete vault. The DFR coolant in the primary and secondary circuits was a mixture of NaK in the ratio of 70% sodium by weight; however, during commissioning some gas entrainment problems led to the addition of more sodium to the primary circuit and the composition is now approximately 75% sodium by weight. The decay heat removal system contained eutectic NaK, which has a composition of 78% K by weight and remains in liquid form down to around -12°C.

In 1967, following a leak from the primary circuit into the leak jacket, a repair was made in which the primary pipework was modified. At the same time, all the primary cold traps were removed and discarded since they were not very efficient and an external cold trap was fitted. These two activities resulted in more than expected NaK remaining after bulk removal. In the late 1970s the NaK was drained from both the secondary systems and the decay heat systems and neutralized by aqueous treatment. In the early 1980s, the project was halted and the reactor kept in care and maintenance until the late 1990s.

At the time of writing (2014) the bulk NaK has been removed from the primary circuit and treated using the same method as for the secondary and decay heat systems and a programme of work has started to remove and dismantle the breeder fuel. The secondary and decay systems were cut and plugged at the biological vault wall but are still contaminated with residual alkali metal. The primary circuit contains a ring main around the vault connecting all of the circuits and each circuit contains an auxiliary vessel. Seven of these extend down below the ring main and as a result of the 1967 repair they do not drain. There is a current project to minimise the residual NaK from the pools inside the reactor vessel and from the auxiliary vessels. Before any further treatment of residual NaK can start the breeder elements will need to be removed and the residual NaK minimised. However, three of these vessels contain hot traps which are made from zirconium foil and will not drain very well during minimisation even if the NaK is removed from the vessels.

3.6. PROTOTYPE FAST REACTOR, DOUNREAY, SCOTLAND

The Dounreay Prototype Fast Reactor (PFR) was a 600 MW thermal fast neutron reactor. This pool-type reactor contained six intermediate heat exchangers and three pumps and associated pump valves. The reactor was commissioned in the early 1970s, went critical in 1974, supplied electricity to the national grid, and was operated until it was finally shut down for decommissioning in 1994. All of the bulk sodium from the primary and secondary systems has been removed and destroyed. The bulk NaK from the decay heat systems and NaK loop heating system has also been destroyed by adding it to the bulk sodium during disposal operations. The residual sodium in the secondary circuits was removed using the WVN method and was successful in completely cleaning the circuits. The secondary circuits were dismantled and removed from the turbine hall to the intermediate heat exchanger stubs. The primary vessel contained approximately 900 m^3 of sodium of which less than ~8 t remains as residues in pools, heels in pipes and films.

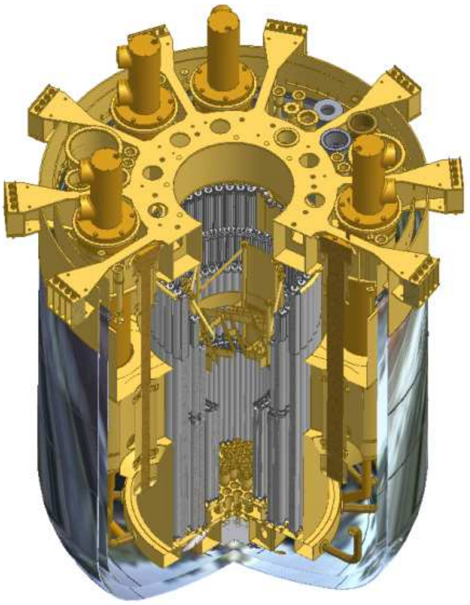


FIG. 8. Schematic view of the Prototype Fast Reactor at Dounreay. Photograph courtesy of Nuclear Decommissioning Authority (NDA) and Dounreay Site Restoration Limited (DSRL).

3.7. RESEARCH REACTOR BR-10, RUSSIAN FEDERATION

Research reactor BR-10 is located in the State Scientific Centre of the Russian Federation ‘A. I. Leipunski Institute for Physics and Power Engineering (IPPE)’, Obninsk. BR-10 operated from 1959 to 2002, with a maximum power 8 MW (t). Although it was a small research reactor, it accumulated a significant amount of radioactive waste in the form of liquid metal coolant.

FIG. 9. Interior view of BR-10, Obninsk. Photograph courtesy of State Scientific Centre Institute of Physics & Power Engineering (IPPE).

The sodium coolant of the primary circuit was changed three times and the cold traps were regenerated, with the resulting spent sodium being drained into the long-term storage tanks. The tanks also received coolant from the secondary circuit (NaK eutectic alloy), contaminated with mercury as a consequence of depressurization of the double-wall steam generator tube with a mercury layer, which was used as part of the BR-2 reactor (see Appendix I).

The waste liquid metal is currently in storage tanks, cold traps, and within the BR-10 systems. Approximately 18 $m^3$of alkaline coolant radioactive waste is temporarily stored within BR-10, with activity totalling in excess of 1000 Ci (37 tBq). The exact amount of radioactive coolant is impossible to ascertain as an unknown quantity of sodium still remains in the intact equipment of the primary and secondary circuits, i.e., pipelines, tanks, heat exchangers, and wave compensators. This sodium cannot be drained; it is coolant residue that will need to be neutralized before the equipment it which it is contained can be dismantled.

3.8. FAST REACTOR BN-350, KAZAKHSTAN

The BN-350 reactor facility with the fast neutron reactor with liquid metal coolant operated 1973–1999, on the Mangyshlak peninsula near Aktau city in western Kazakhstan. The reactor was designed for production of weapons-grade plutonium and to supply steam for a water desalination facility and the turbine generators of a thermal power station. The designed thermal power of the reactor was 1000 MW (t) and the designed electrical power of 350 MW (e). The maximum thermal power achieved during operation was 750 MW (t).

FIG. 10. Exterior view of the BN-350 reactor building. Photograph courtesy of Nuclear Technologies Safety Centre (NTSC).

The main reactor features 3 circuits with 6 loops in each circuit. The total volume of sodium in the primary and secondary circuits was 1180 m^3. In addition, 20 m^3 of NaK were in the support systems.

During decommissioning all of the spent fuel was discharged from the reactor, characterized, and packaged into the stainless steel canisters filled with argon gas. Canisters were loaded into heavy metal-concrete casks and sent to the former Semipalatinsk test site in eastern Kazakhstan for long term storage. The casks are dual-use casks and are used for transportation and long term storage of spent fuel. The transportation of the spent fuel was accomplished in December 2010.

Primary sodium was drained from the loops and the reactor vessel into the storage tanks. It will be processed into NaOH at the sodium processing facility that was constructed near BN-350 reactor facility. Secondary sodium was sent to Ulba metallurgical plant (Kazakhstan) for other uses. Residual sodium treatment is discussed in the Section 4.

4. TECHNOLOGIES FOR TREATMENT OF LIQUID METAL COOLANT

4.1. EXPERIMENTAL BREEDER REACTOR II — CARBONATION

The EBR-II reactor was shut down in 1994. Initially the reactor was defueled and then drained. The bulk sodium was processed at Argonne National Laboratory—West[2] into solid NaOH, so that it could be disposed as solid low level waste.

The third and final phase of the decommissioning activity was the placement of reactor and non- reactor systems in a radiological and industrially safe condition. The planning for this phase included a detailed system-by-system evaluation to determine necessary actions based on the following definition:

> *Radiologically- and industrially-safe is the placement of equipment and facility in a condition that does not pose any unusual, unexpected or additional industrial safety risk and does not pose a radiation or contamination risk beyond normal EBR-II levels for controlled access areas.*

As the system-based planning was developed, necessary and newly identified surveillance activities were identified. Surveillance provides the regulatory assurance that EBR-II systems would not deteriorate.

A major component of the radiologically and industrially safe strategy was treatment of the residual radioactive sodium remaining within system piping and components. The conversion of the exposed residual sodium surfaces to a non-reactive layer of sodium carbonate (Na_2CO_3) was accomplished through a process called passivation. Passivation (or carbonation) is achieved through the controlled humidification of the carbon dioxide (CO_2) cover gas. The presence of water vapour in CO_2 forms a layer of Na_2CO_3. The carbonate layer remains porous and does not impede reaction rates until a layer of greater than 20 cm has been achieved. All exposed surfaces of sodium remaining within both the primary and secondary systems have been passivated.

After draining the primary sodium a closed-circuit television camera was inserted into the EBR-II primary tank. The objectives of this visual inspection included confirming completion of the sodium coolant draining process; providing a visual understanding of the draining process and residual sodium deposition locations; and finally providing a visual assessment of tank and component integrity.

Visual examination provided evidence of the draining process and confirmed the removal of all but very small quantities of residual Na. An additional visual examination followed the completion of passivation of all primary systems.

4.2. FAST FLUX TEST FACILITY — SUPERHEATED STEAM

Liquid metal disposition at Hanford has historically used the WVN technique. This method has been successfully used in the disposition of several sodium test loop facilities. The most recent sodium processing effort involved the use of superheated steam to remove sodium in the Composite Reactor Components Test Activity (CRCTA) vessel. The CRCTA was used to

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[2] Now part of Idaho National Laboratory.

test initial operation of FFTF reactor equipment in a sodium environment. The vessel is a cylindrical tank, 4.6 m in diameter and 11 m tall, which replicated one tri-sector of the FFTF reactor vessel. The upper portion of this vessel consisted of a sodium flow baffle plate (which was sodium wetted during operation) and a series of 25 thin thermal baffles spaced 25 mm apart (which resided in an argon environment but were subject to sodium frost deposition). The planning effort assumed that as much as 1900 litres of metallic sodium remained as residual in the vessel, with as much as 1136 litres retained in the thermal baffle area. The location of steam injection points are shown in Figure 12.

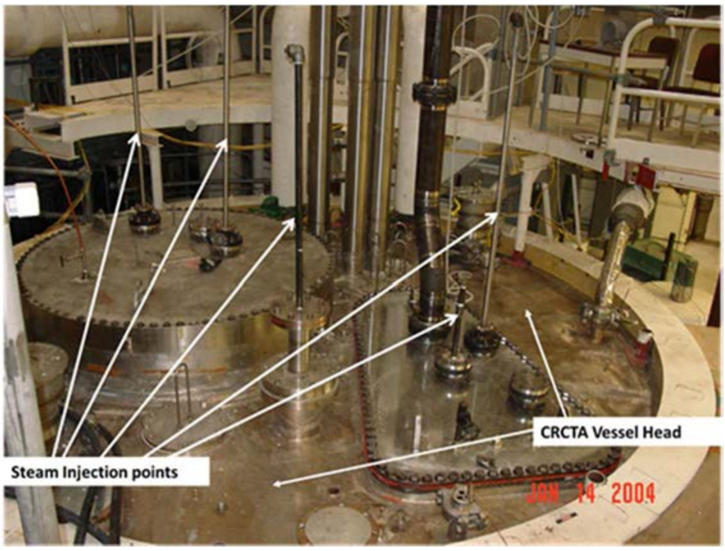

FIG. 11. Exterior view of the CRCTA vessel at Hanford. Photograph courtesy of the US Department of Energy, Richland Operations Office.

The entire vessel was heated to 200°C prior to the introduction of steam. The tank was then inerted with nitrogen and a steam flow of ~23 kg/h was initiated. The reaction was monitored by hydrogen generation and temperature and the steam injection probes were manually moved to new locations as the reaction proceeded. The sodium reaction took about 8 hours, with the vent gas containing 30–36% hydrogen by volume. A total of ~350 litres of sodium was reacted. The vessel was then filled with water and drained twice to ensure removal of the reacted product.

This reaction of the residual sodium in a mechanically complex environment was performed safely and effectively utilizing superheated steam without the drawbacks of the WVN process of potential ‘rollovers’ (i.e. sudden heavier than normal sodium-water reactions resulting in pops, bangs, and pressure excursions) and lengthy processing time. The primary constraint to successful use of superheated steam method at FFTF is the need to preheat the systems to 200°C prior to initiating the reaction. This preheat would require re-energizing the preheat system on the piping, as well as utilization of other methods to preheat the reactor vessel.

4.3. PHÉNIX — CARBONATION

Before 2009, all large Phénix components were cut by means of plasma and packaged into smaller sections in order to meet the low level and intermediate level short lived waste specification. This manpower intensive effort was expensive and exposed personnel to increased radiation dose and industrial hazards. Therefore, for safety and financial reasons, technical and regulatory evaluations were performed to allow waste disposal of large components without physical segmentation. Based on operational need, a dummy (false) intermediate heat exchanger (DOTE) is now used to replace an intermediate heat exchanger to provide a radiation shield and a seal between the hot and cold pool with an argon cap. The DOTE is a cylinder ~7 m long and 1.7 m in diameter weighing ~16 tonnes. The location of the DOTE in the Phénix is shown in Figure 13, as well as a photograph of the DOTE during handling (Fig. 14).

After removal from the reactor, dose rate and contamination mapping is performed to determine hot spots and how to process the component. The residual sodium is then removed from the DOTE by carbonation. Following direct spraying by steam and rinsing, the DOTE is decontaminated with sulphuric and phosphoric acid. The lead shot (shielding) that was previously removed to ease handling is then reinserted into the DOTE. The significant amount of empty space remaining in the DOTE was loaded with waste.

The final product is a large reactor component that can now be delivered directly to the waste repository, thereby avoiding expensive sectioning and mitigating radiation exposure to personnel.

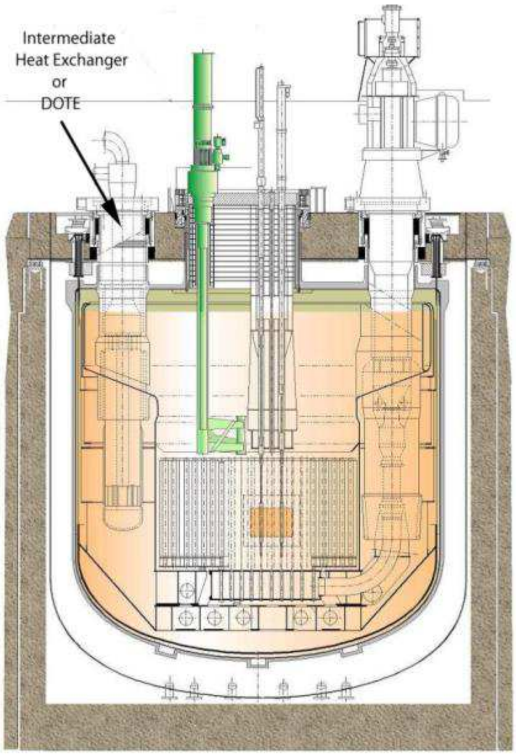

FIG. 12. Location of the Dummy Heat Exchanger (DOTE), Phénix reactor. Image courtesy of CEA.

FIG. 13. Removal of the DOTE. Image courtesy of CEA.

4.4. SUPERPHÉNIX — CARBONATION

Following draining of the reactor vessel as much as 800 kg residual sodium is expected to remain, with another ~900 kg sodium as a wetting film on the component surfaces. Following carbonation of the reactor vessel, it will be flooded with water to ensure that no metallic sodium residuals remain and also to provide radiation shielding from the highly activated components in the lower part of the reactor vessel. The possibility of hydrogen generation during flooding and potential explosions will require the flooding process to maintain a hydrogen concentration below the lower explosive limit.

There are three components at the Superphénix facility that contain a total of <3 kg of NaK, and < 182 kg of NaK oxides, as well as other chemical forms of sodium and NaK. From 2007 to 2009, a series of trials was performed to determine the chemical reaction rates, morphology, and risks involved in reacting the remaining NaK material.

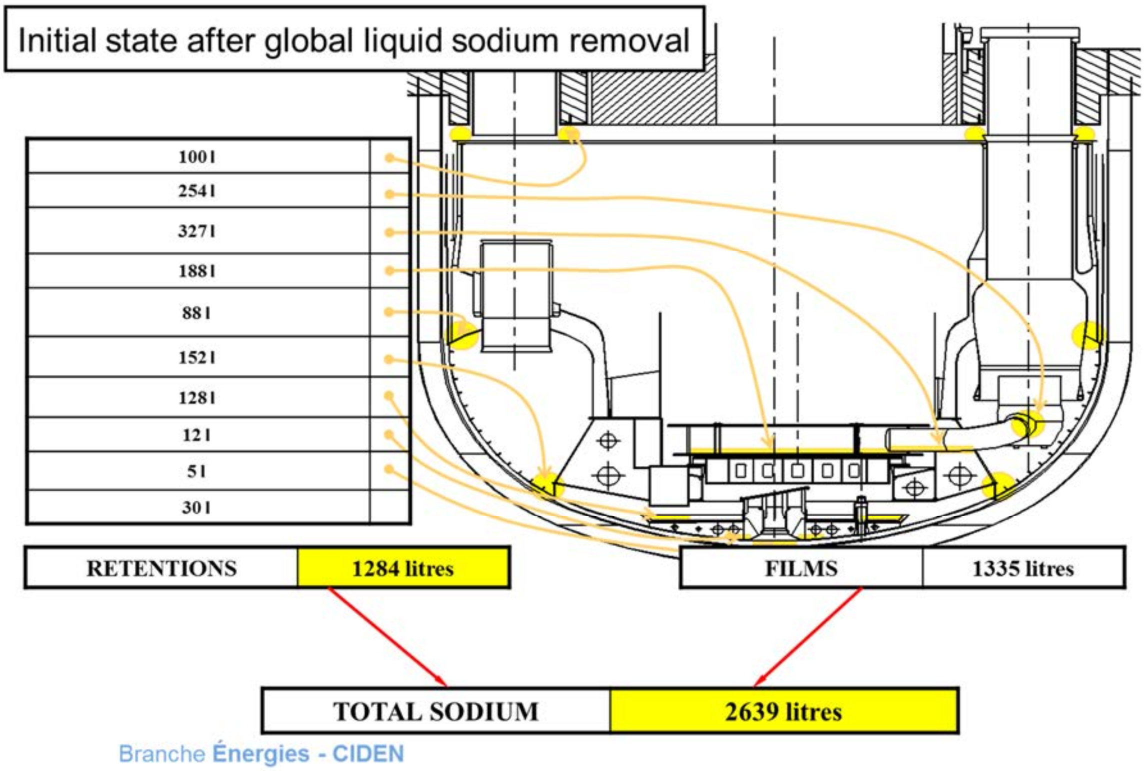

FIG. 14. Lower part of the reactor vessel at Superphénix. Image courtesy of EDF.

4.5. DOUNREAY FAST REACTOR — METHODS UNDER CONSIDERATION

The DFR was shut down in 1977 and a programme of decommissioning was begun very soon afterwards. This started with defueling and then draining of the thermal siphon circuits and secondary circuits. All drained NaK was passed through a dedicated treatment plant that sprayed the NaK into a nitrogen jet and then into aqueous hydroxide solution. Because the secondary and thermal siphon circuits were lightly contaminated (mainly tritium), they were also treated using an aqueous process and cut back to the biological vault of the primary circuits and reactor vessel.

Earlier in the reactor life the cold traps in the auxiliary vessels were removed before they became too contaminated and were cut open to clean the contents of residual NaK. In the early 1980s, the first batch of primary NaK was passed through the NaK treatment plant when the decommissioning activities on DFR were stopped due to external constraints. At this time the NaK treatment plant was examined and beads of Hg were found in the base of the NaK treatment plant reaction vessel. During operation of DFR, the NaK had been contaminated with mercury (Hg) from the rotating shield dip seals and a NaK-Hg amalgam was formed. The DFR was then placed in care and maintenance until the late 1990s, when the NaK treatment plant was re-commissioned and brought up to the then-prevailing standards for nuclear safety.

Several processes were examined to determine the best method of removing the residual NaK. Initially, WVN was proposed, but unsteady reaction behaviour led to moving to a process that formed dry hydroxide. To attain reasonable reaction rates this had to be done at 120°C with a water vapour concentration of 4% by volume in the nitrogen. Even though this appeared to be successful there was also an issue of potassium superoxide being present, which decomposes and could even explode when in contact with the NaK. Following worldwide experience, a sodium carbonation experimental programme was undertaken to investigate the use on NaK. This also gave some unexpected results because the NaK liquid migrated through the bicarbonate to form worm like structures due to the hydraulic force exerted on the liquid NaK during the carbonation process. These worm-like structures contained a core of NaK surrounded by a bicarbonate crust. With little mass, the NaK–water reaction can elevate the bicarbonate temperature such that it can thermally decompose to carbonate releasing more water for reaction with further NaK. Although the decomposition of bicarbonate is endothermic, the NaK water reaction is exothermic and initiates a positive feedback system, elevating the temperature significantly. Because the atmosphere was CO_2-rich, the NaK started to react with the CO_2 when the local temperature became high enough, which led to a runaway reaction. The carbonation process was thus rejected for use on DFR. This process was not observed on sodium or potassium reactions since the metal was in the solid phase during normal carbonation reactions and the worm-like structures were not formed.

Presently it is proposed to remove as much of the held up NaK as possible and to process the pipework using either superheated steam (SHS) for the larger hold ups or low temperature WVN for the lightly contaminated pipework.

4.6. PROTOTYPE FAST REACTOR — METHODS UNDER CONSIDERATION

The PFR was shut down in 1994, followed by the use of WVN on the secondary circuits. A programme of work was subsequently initiated to investigate using WVN for removal of the residual sodium within the reactor systems. Initially, experimental work was undertaken to determine the reaction characteristics of sodium with WVN under various different conditions in order to set the parameters for clean-up of the reactor vessel. At the same time design and construction of the sodium disposal plant was initiated. It was known that to remove as much sodium as possible, intervention was required to drill through the strongback to allow the hot pool to drain into the cold pool where the bulk sodium was to be extracted, and also to drill the low pressure plenum and pierce the high pressure pipework. At this time it was understood that the WVN technique would deal with the rest of the residue. However, the DFR and PFR projects were running in parallel and when the instability as described in Section 4.5 was better understood, the risk of instability was thought to be unacceptable.

The process was then changed to one in which the hydroxide was produced at 120°C as a dry hydroxide solid product. Alternatives were evaluated, followed by a conceptual design. This approach was abandoned due to the uncertainties in being able to successfully perform the process.

Presently, it is likely the main retention area of the heel pool and core melt-out tray will be drained and the sodium treated in a separate facility using superheated steam. All the major components are to be removed, size-reduced where necessary, and treated using either WVN or superheated steam. The main vessel and remaining structure is then likely to be treated using carbonation or low temperature WVN.

4.7. RESEARCH REACTOR BR-10 — SOLID PHASE OXIDATION

Two main treatment methods are used for the BR-10: solid-phase oxidation is applied to bulk liquid metals (see also Appendix I) and neutralization is applied to non-drainable residues (see also Appendix II).

4.7.1. Solid-phase oxidation method

The solid-phase oxidation method was successfully demonstrated 16 times at the Mineral-3 laboratory installation with 3–5 litres of alkaline metal processed at a single time (sodium, NaK, NaK-Hg, and radioactive sodium) into a mineral product. It was established that the product did not contain any un-reacted alkaline metal. The process of solid-phase oxidation does not result in any release of ^{137}Cs outside the product material, which may also contain 40–50% of mercury admixture. Release of hydrogen into the envelope gas is insignificant, so that the risk of a hydrogen explosion may be discounted as a safety concern.

At the completion of the experimental series, 10 litres of alkaline metal had been successfully treated using an improved version of the solid-phase oxidation process. The method of solid-phase oxidation of alkaline metals is protected by patent[3].

The product of alkaline metal radioactive waste processing is a mineral-like sintered solid of the reaction products contained inside a steel reaction container. It is a stable matrix that ensures containment of all radionuclides (which the exception of tritium) from the alkaline coolant and all alkaline elements as chemically stable compounds. The product material is the first barrier preventing the release of radionuclides into the environment; the second barrier being the steel shell of the reaction vessel. It is immediately shipped for dry storage in a solid radioactive waste storage facility.

4.7.2. Neutralization of non-drainable residues

The IPPE has developed a method neutralizing non-drainable residues of alkaline liquid metal coolants (Na, NaK alloy) with a gaseous suboxide of nitrogen (dinitrogen oxide, N_2O), which is characterized by the absence of hydrogen generation, improving the safety of the technology.

Research and experimentation established a mechanism of interaction between sodium, caesium and eutectic NaK alloy with gaseous suboxide of nitrogen. It was demonstrated that the intermediate compound that ensures complete consumption of the alkaline metal (Me) by the reaction is $Me_2N_2O_2$, hyponitrite of alkaline metal. Initial conditions of interaction were determined along with temperature effects and the composition of final product (a mixture of oxide, nitrate and nitrite of alkaline metal in various combinations) as a function of the duration of the neutralization process. The kinetics of the nitrogen suboxide absorption by the alkaline metal was established, composition of the gaseous phase was studied, and dependence on other parameters was determined.

Research has been performed to develop a conditioning technology to neutralize the final product by placing grout inside the cold trap itself. It was demonstrated that the most promising and workable method is incorporation into a geocement compound. Cold traps act

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[3] Russian Federation patent No. 2200991, issued in 2003.

as packaging for solidified radioactive waste, which may be sent to long-term storage or disposal. The method will be subjected to further testing on the cold traps of the first loop of the second circuit of the BR-10 reactor.

4.8. BN-350 REACTOR — CARBONATION

The comparative analysis of the different technologies from the points of view of safety, cost effectiveness and applicability showed—for the conditions of BN-350—the most applicable technologies are steam-gas washing and passivation with moist CO_2.

The physical-chemical basis of the hydrocarbonation technique consists of interacting sodium and moist gas. The positive results of laboratory testing and full-scale usage of the technology at EBR-II provided assurance that the technology would successfully result in passivated sodium residuals that remained in the “pockets” and bottom of BN-350 reactor vessel. Passivation through hydrocarbonation technology was carried out with additional equipment, such as special reagents producing system, exhaust gases control system, and supply/drainage pipelines.

At present this method can be considered as the safest among other known methods of residual alkali metals passivation. The experiments, which mostly repeated those performed by EBR-II researchers, tested the BN-350 specific configurations that could be difficult for passivation.

After drainage of sodium coolant from the equipment and the systems of BN-350 reactor facility some sodium in the form of films remained on the internal surfaces of equipment and pipelines. The depth of the sodium film depends on the temperature mode during and after drainage, the sodium purity and the location of these surfaces. Some non-drainable equipment cavities also have sodium residuals in the form of ‘puddles’ or ‘pools’ of various depths.

The sodium residuals removal system was erected in the central hall of BN-350. The system used the hydrocarbonation technique; it also provided a means for ‘post-washing’ of the hydrocarbonized systems with water. All of the main internal surfaces of the BN-350 primary circuit pipelines and equipment were completely carbonized, as was the secondary sodium. Intermediate heat exchangers and other large equipment were disconnected and plugged to make the process possible. Over 380 kg of primary sodium and ~200 kg of secondary sodium was converted into carbonate form.

FIG. 15. Sodium residues in the pipelines of the secondary circuit of BN-350. Photograph courtesy of Nuclear Technologies Safety Centre (NTSC).

4.9. SUPERPHÉNIX — LASER CUTTING TECHNOLOGY

The draining of sodium coolant from the Superphénix reactor primary vessel will leave several sodium retention puddles that cannot be drained by the main electromagnetic pumping system. These are generally inside large diameter pipes and in plenums deep inside the reactor system. Eight junction pipes 850 mm in diameter fed the sodium from the primary pumps to the core structure. In order to access these areas, the cutting device has to proceed downwards through the reactor lid and to worm its way into the internal structures down to the cutting site without breaking the containment of the vessel. After technological cutting tests, the laser process was selected. However, due to the specific environmental conditions in the primary vessel, development was necessary to validate the suitability of the system for all the conditions it would have to endure.

The sodium retention puddles are situated in the primary vessel of the Superphénix reactor in the pipework connecting the pumps to the core. There could be heels up to 30 mm deep and up to 160 mm in the bellows around each pipe that contain up to 2.7 m^3 sodium. This quantity prevents the immersion of the internal structure without prior treatment. Because of the uncertainty in carbonation penetration in these areas it was decided to cut the steel structure in order to let the sodium drain into the base of the vessel.

Trials were conducted in 2007 on full-scale models of the retention geometries, but the sodium was replaced by a low melting tin-lead temperature alloy. Different methods of cutting were tested, of which laser proved to be the most efficient. Laser technology was also chosen for its capacity to cut without contact and with minor mechanical load, and for its ability to cut thick and heterogeneous structures. This technology has made great progress over the last 15 years. The power of the laser systems has increased while the ruggedness has improved and the size reduced.

The selected approach to get to the retention areas is through the plugs that now replace each primary pump and enter the reactor vessel and worm its way into the 850 mm diameter pipe up to the bellows. The sodium retention puddles are distributed among eight areas so the trajectory will be repeated twice for each reactor loop. The distance of about 20 m and the width of the hole make access to the operating zone problematic. Further, there are other obstacles to overcome around reach the retention area. In addition to the accessibility difficulties, the internal environment represents some other challenges like the temperature, radiation level, inert gas cover and sodium aerosols.

‘Charli’ is a small remote-operated vehicle equipped with a robotic arm fitted with a laser cutting head and equipped with multiple cameras. It is designed to worm its way into the internal structures down to each Superphénix "LIPOSO," which are the pipes in which it is to be deployed. It carries the laser cutting head, camera and sensors to help it to get into the operating position. Due to the limited load capacity of the robotic arm, a specific laser cutting head was designed to reduce its weight to less than 2.5 kg in all (with coolant, camera, deflector protection against sodium and aerosols projections, head dropping device).

One of the questions before the beginning of the trials was the behaviour of laser cutting through sodium with a departure of the laser beam directly on the sodium surface. The very low melting temperature of sodium could result in flow onto the cutting zone, possibly creating a back reflection of the laser light from its surface that supposedly could damage the laser itself. This phenomenon did not appear during tests or operations, but another point was noticed: the production of sodium aerosols was massive, far more than expected. From the beginning of the cutting, the sodium splashes all around, and also evaporates in a thick fog from which particles seep everywhere, even inside the robot body. Therefore, splashing and aerosol proof protections had to be designed for the robot.

4.10. IDAHO NATIONAL LABORATORY — INFRARED VIZUALIZATION TECHNOLOGIES

Thermographers often use infrared cameras to troubleshoot equipment and to conduct preventive maintenance inspections. When evaluating an electrical power system, a thermographer using an infrared camera can help determine whether a component is in danger of failing due to excessive heating from a loose connection or an unbalanced load. In an inspection of rotating machinery, a comparison of thermal images over time can help predict problems before they occur; images showing significant heat increases are often a sign that bearings or seals are about to fail.

Safety has long been a major advantage of using infrared cameras for inspecting equipment; infrared inspections usually can be conducted at a safe distance while the equipment is powered up and running. This capability is a primary reason why these cameras have been used in the civilian world for the past 40 years. And as infrared cameras continue to become more sophisticated and less expensive, they are providing value in an ever-increasing variety of unique applications.

One of the most novel industrial uses of infrared cameras is in decommissioning of nuclear facilities. These cameras are being used at the Idaho National Laboratory site to provide process feedback and to monitor safety while residual sodium is removed from the primary vessel and cooling loops of the sodium-cooled EBR-II. Primarily, infrared camera techniques were used, followed by verification of levels in some areas by running a visual camera down the pipe to the same locations.

The most common infrared cameras use a lens to focus infrared radiation onto a focal plane array, a two-dimensional crystal array that is sensitive to infrared wavelengths. The electronic signal from the focal plane array is processed by the camera’s electronics to produce a thermal image on a video monitor, typically with lighter colours representing warmer temperatures, as shown in Figure 19 using the FLIR M-626L camera. More expensive cameras can perform precise temperature calculations if the surface characteristics of the viewed object are known.

FIG. 16. Real-time sodium reaction monitoring via camera in the EBR-II reactor vessel.

5. LESSONS LEARNED FOR MANAGEMENT OF LIQUID METAL COOLED REACTOR DECOMMISSIONING

This section presents key lessons learnt from experience gained in the implementation of decommissioning of liquid metal cooled reactors, particularly in relation to dealing with residual sodium and NaK.

5.1. EARLY REGULATORY INTERACTION

Discussions about decommissioning strategies with the regulators should begin at an early stage of planning. Engagement in the formative stages of major activities is extremely important. This early involvement promotes a teaming concept, brings additional perspectives and input from the regulatory technical review, and provides insights into public acceptance and involvement.

Regulators were involved in the case of WVN treatment of the secondary circuits of PFR and bulk removal of sodium at both PFR and DFR. Informal discussions with the regulators on the WVN project helped their understanding the process and also enhanced the ability of the designers and operators to meet the regulatory requirements.

5.2. DECOMMISSIONING STRATEGY SELECTION AND TIMING OF IMPLEMENTATION

Selecting a strategy for decommissioning reactor facilities containing contaminated liquid metal coolant is based on several competing considerations. These include, in particular, the level of risk presented to the safety of the workforce and to the general public, and the public policy considerations on the remediation of contaminated sites.

Experience shows that decisions concerning the timing of facility dismantling and the disposition of contaminated components are constrained by the availability of waste disposal routes, the socio-economic impact on local communities and the availability of funding. Two specific examples that illustrate contrasting timing approaches are:

- The FFTF facility is remote from population centres and, therefore, presents a low safety risk to the public. As a result, available financial resources are applied to other projects on the site that represent a higher level of risk. There may be a significant time delay before the facility is completely decommissioned. During this delay, residual sodium in FFTF is being maintained in a safe state by means of an argon blanket;

- The current strategy being followed at the PFR and DFR is to proceed with early dismantling of the plants, due to the greater weight given to a public policy preference that the Dounreay site should be remediated over a period of one to two decades.

5.3. DECOMMISSIONING PLANNING

Prior to commencement of the removal of liquid metal from shutdown reactors, a full decommissioning plan should be prepared. This plan should specify the disposition strategy for all contaminated components, the intended end state of facility, and recognition of uncertainty of funding.

Experience shows that, in cases where the removal of bulk liquid metal is undertaken without giving adequate consideration to the management strategy for the contaminated components and removal or passivation of the residual liquid metal, there may be unintended consequences for the latter activity. Two specific examples at FFTF include:

1) The primary system cold traps and vapour traps do not have a defined path to disposition;

2) The core components pots used to hold fuel assemblies in the spent fuel pool were cut in half and stored in containers. The loss of funding for decommissioning of FFTF and the lack of timely environmental documentation created an additional storage issue—namely the need to store these containers on-site for an indefinite period of time.

5.4. OPTIMIZE THE CONTRACTING STRATEGY

Active measures should be taken to optimize the contracting strategy and to manage the supply chain for the decommissioning of liquid metal reactors.

The experience gained at the BN-350 reactor with the supplier of the CO_2 gas demonstrated the importance of understanding the business relationship between subcontractors. This situation was context specific and probably unique: the gas supplier had debts to the electricity supplier, which resulted in the electricity supplier terminating supply. As a result the gas supplier could not meet its obligations to the BN-350 project, even though the electricity supplier was itself the operating company for the BN-350 facility.

5.5. THE ROLE OF THE INFORMED OPERATOR

When preparing procedures for sodium treatment, it is extremely important that those who are creating the procedures utilize technical expertise and system engineers, especially when the new operation is being applied for the first time. In this statement, "new" not only refers to the process, but also the physical conditions and configurations for each case of applying the process. Furthermore, those preparing the procedures must be indoctrinated in the reactions that are taking place and the consequences of errors.

An example of the lack of this understanding was at the Janetstown off-site test facility near Dounreay where tanks were being cleaned using WVN. To reduce the risk of instability occurring, the tanks were tilted to expose more sodium. However, this increased the risk when the inventory of hydroxide solution became much higher and caused the sodium reaction to 'stall'. The stalling of the reaction was because most or all of the sodium became submerged by the hydroxide solution. The operators continued to add WVN for several hours, which continued to be absorbed at the surface of the hydroxide solution. The operators eventually decided to sparge nitrogen through the hydroxide liquid. This resulted in rapid mixing of diluted and concentrated solution that ended in a large reactive excursion that ruptured the bursting disc on the discharge line.

In another example, while preparing to conduct hydrocarbonation, it was discovered that in some places the quantity of residual sodium was greater than anticipated, and in other places the quantity was less than expected. This was due to:

- Not checking the "as built" documentation (particularly drawings) to determine the real dimensional parameters of the installed equipment and pipelines;
- Insufficient knowledge of the design and lack of knowledge transfer of the design;
- Inadequate calculation of the coolant residuals resulting from a lack of detailed characterization of the facility during decommissioning planning before operations commenced.

As an example, a simple check of slopes of pipelines can help in understanding the real situation and prompt preparation of recovery measures to decrease the volume of residuals of the coolant.

5.6. PROCESS DESIGN FOR DECOMMISSIONING OPERATIONS

Examples of considerations for designing decommissioning processes include:

- All drainage systems must be checked for gas permeability and be heated as high as technically reasonable during the draining procedure so as to avoid plugging by impurities of sodium;
- The presence of dead legs in liquid metal circuits at nuclear power plants is unavoidable. Consider the use of vacuum distillation during system heating up to 500^{o}C and collect the sodium vapours in the vacuum traps;
- Deleting or neutralization of sodium residuals in equipment and subsequent deactivation of the system must use the initial design of the circuit as modified during construction and subsequent design changes; and
- The final cutting of clean equipment has to take place under a neutral gas environment (nitrogen, argon) to avoid accidental situations.

5.7. SPARES RETENTION FOR MOCKUPS

The decommissioning activity at the reactor is often carried out by specialized organizations with appropriate licences. In cases where decommissioning is undertaken using the operating organization's own staff, equipment spares should be retained following the closure of the reactor. These spares will aid the construction of mock-up equipment to be used for checking the technologies and training the workforce. Examples of components that could have been of use at FFTF were the spare instrument tree and the spare in-vessel handling machine.

5.8. WASTE STREAM OPTIMIZATION

All fast reactors have NaK systems — some of which are only lightly contaminated. In the planning stage for the bulk removal it should be considered that this NaK could be mixed with the bulk sodium before its destruction as a means of optimizing the waste stream. This can alleviate the need for a specially built NaK passivation facility since the potassium in the bulk sodium can effectively be considered as chemical contamination. This is not feasible for all waste streams; for example, some may be constrained by as radioactivity or mercury contamination. This method was used at EBR II and FFTF where the NaK systems were mixed with the sodium systems prior to draining. In this way all the NaK was flushed out of the pipes leaving a solidified, slightly potassium-contaminated pipe, for residue removal. A similar method was used at PFR where the system NaK was mixed to attain 3% potassium prior to passing through the sodium disposal system. In this way a specially built NaK destruction facility was not necessary.

At EBR II the sodium was drained and stored at Argonne National Laboratories—West[4] awaiting its final disposal. The process was chosen based on the final form of the waste rather than the easiest or least expensive process; however, there were cost savings in the end since the waste form produced could be stored as low level solid waste and negated the need for further downstream processing.

5.9. WASTE PACKAGE OPTIMIZATION

Preparing liquid metal reactor components for waste disposal can be a complex operation when there is residual sodium on and/or within the items. It is important that the organizational elements responsible for dismantling coordinate closely with those responsible for waste management. In particular, questions should be addressed as to how the presence of sodium or NaK will affect personnel and equipment for all steps between preparation for removal and packaging for disposal.

As a specific example, size reduction of metal for disposal presents hazards to workers when they are directly cutting or shearing metal, not only from the tool operations but also from the residual sodium. If the components are in radiation areas or they are radioactive, the situation becomes complicated with the potential for exposure to humans. The specific example is described in Section 4.3. Before 2009, all large Phénix components were being size reduced into smaller sections in order to meet the waste disposal packaging specifications. This manpower intensive effort exposed personnel to increased radiation dose as well as industrial hazards. A waiver was sought and granted to allow disposal in larger sized containers.

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[4] now Idaho National Laboratory.

APPENDIX I. BR-10 REACTOR SODIUM AND SODIUM-POTASSIUM REMOVAL AND PASSIVATION[5]

I.1. BR-10 CONDITIONS BEFORE PASSIVATION

Research reactor BR-10 (Fig. 17) is located in the State Scientific Centre of the Russian Federation ‘A.I. Leipunski Institute of Physics and Power Engineering’ (IPPE), Russian Federation, Obninsk. BR-10 operated from 1959 to 2002, during which time a significant amount of radioactive waste in the form of liquid-metal alkali coolants was accumulated by the research institute. Figure 18 is a schematic illustration of the BR-10 systems.

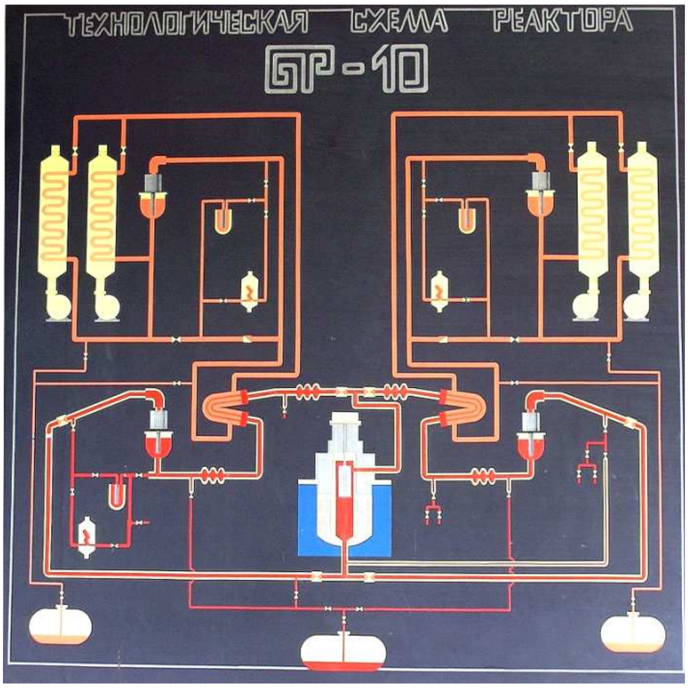

FIG. 17. The BR-10 reactor.

FIG. 18. Primary and secondary circuits of the reactor.

The sodium coolant of the primary circuit was changed three times, and the cold traps were regenerated with the resulting spent sodium drained into tanks for long-term storage. The tanks also received sodium potassium (NaK) eutectic alloy coolant from the secondary circuit, contaminated with mercury. This was the result of de-pressurization of a double-wall steam generator tube with a mercury layer, which was part of the BR-2 reactor. The BR-2 reactor was located in the building where the BR-10 reactor was built. Tables I.1 and I.2 indicate the volumes and activities of the drained material.

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[5] All photographs in this appendix are courtesy of State Scientific Centre Institute of Physics and Power Engineering (IPPE).

TABLE I.1. STORAGE TANK MATERIAL VOLUMES

Tank	Volume of Material	Sodium	Potassium	Mercury
First Set Storage	4.5 m^3	44%	48.5%	7.5%
Second Set Storage	4.5 m^3	95%	5%	0.02%
Primary Circuit Tank	2 m^3	100%	-	-

TABLE I.2. STORAGE TANK ACTIVITY

Tank	Caesium-134	Caesium-137	Tritium
First Set Storage	not reported	$7*10^8$ Bq/kg	$2*10^8$ Bq/kg
Second Set Storage	not reported	$3*10^8$ Bq/kg	$2*10^8$ Bq/kg
Primary Circuit Tank	up to 10^6 Bq/kg	up to 10^8 Bq/kg	up to 10^8 Bq/kg

In addition to these materials, the reactor had 15 spent cold traps for oxides that were cut out of the primary circuit. During operation they each accumulated between 1.5 TBq (40 Ci) and 3.7 TBq (100 Ci) of Cs-137 activity. The combined amount of radioactive sodium in the cold traps is approximately 3 m^3.

In addition to the primary circuit material, processing is also required for some 4 m^3 of sodium drained into the tank from one loop of the secondary circuits, which is also classified as radioactive waste ($2*10^9$ Bq/kg of tritium). This first loop's secondary cold trap was cleaned by gaseous N_2O some years ago. The second loop of that circuit still contains one cold trap with 200 litres of sodium contaminated with tritium, which is being processed using N_2O.

The total amount of alkaline coolant radioactive waste currently in temporary storage is approximately 18 m^3, with activity totalling in excess of 37 TBq (1000 Ci). The exact amount of radioactive coolant is impossible to identify as an unknown quantity of sodium remains in the still intact equipment of the primary and secondary circuits, which includes pipelines, tanks, heat exchangers, and wave compensators. This sodium cannot be drained; it is coolant residual that requires a special technology for its neutralization (passivation) before the equipment can be dismantled.

In 1997 it was decided to convert BR-10 into an experimental testing ground (polygon) for verification of decommissioning technologies for fast-neutron reactors[6]. In accordance with Minatom Order No. 204 of 30 March 1999, BR-10 was shut down in December 2002.

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[6] Order No. 130 dated 5th March 1997, Ministry of Atomic Power of the Russian Federation.

I.2. METHODS FOR REPROCESSING OF SODIUM AND NA-K (BULK AND RESIDUALS)

I.2.1. Solid-phase oxidation method

Analyses were performed for various processing methods of alkaline metals. The method chosen for further research in consideration of production effectiveness and safety was solid-phase oxidation of alkaline metal using slag from the copper-smelting industry. The method is based on a spontaneous exothermic chemical reaction of alkaline metal with iron oxides and silicon oxide, which are the key components in the slag. The volumetric mixture ratio prior to processing is one part alkaline metal mixed with 2.5 parts slag.

The solid-phase oxidation method was successfully demonstrated 16 times at the 'Mineral-3' laboratory setup using 3 to 5 litres of alkaline metal processed separately with Na, NaK, NaK-Hg (with as much as 40% to 50% mercury admixture), and radioactive sodium. It was established that the mineral product did not contain any un-reacted alkaline metal. It was further determined that solid-phase oxidation retains all ^{137}Cs within the mineral product. Also, the hydrogen released into the cover gas is insignificant; which eliminates the hydrogen safety problem. When process improvements were finalized, 10 litres of alkaline metal had been successfully treated. This method of solid-phase oxidation of alkaline metals is protected by patent[7].

After these successful experiments with Mineral-3, the Mineral-30 rig (Fig. 19) was fabricated and used to carry out seven large-scale demonstrations of the process with as much as 30 litres of alkaline metal (Na, NaK) in each batch.

FIG. 19. The Mineral-30 apparatus.

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[7] Russian Federation patent No. 2200991, issued in 2003.

The Mineral-50 rig was next fabricated to process about 50 to 60 litres in a batch. Two large-scale batches of sodium were processed. This processing demonstrated the following advantages of the solid-phase oxidation process:

- Unlike water-based technologies, neutralization of alkaline metal in the solid-phase oxidation process occurs in a single phase;
- The product of radioactive alkaline metal processing is solid mineral-like, sintered reaction product, shown in Figure 20 (with the steel reaction vessel removed);
- The mineral product consists of chemically stable compounds that ensures containment of all radionuclides (which the exception of tritium) and all alkaline elements within its matrix;
- By design, the product is contained inside a steel reaction container that can be immediately placed in a dry storage radioactive waste facility. It is necessarily fabricated with carbon steel because stainless steel (grade X18H10T) turned out to be prone to cracking. The carbon steel results in less costly vessels;
- The mineral product matrix is the first barrier preventing the release of radionuclides into the environment; the second barrier being the steel reaction vessel;
- The relatively high temperature (1100–1350°C) shown in Figure 21 that develops during the solid-phase oxidation process ensures that the alkaline metal reacts completely with the slag.

FIG. 20. Product of solid-phase oxidation process 30 L NaK at Mineral-30.

The final amount of processing product that is sent to dry storage exceeds the amount of the original radioactive waste by a factor of 2.5–3. The total activity of the final product will equal that of the original radioactive waste, with the specific activity being, respectively, some 2.5 times less.

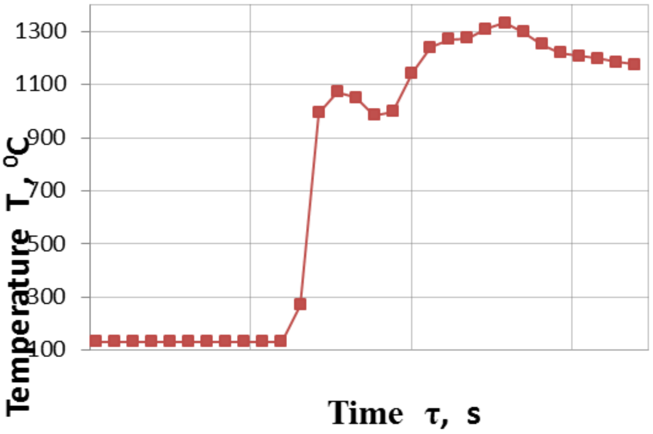

FIG. 21. Temperature-time profile during the solid-phase oxidation processing of sodium.

The solid-phase oxidation process does not result in the generation of any gases that would require the application of special safety precautions. No intermediate or final forms of liquid radioactive waste are generated. In order for the reagents to be mixed (metallurgical slag and alkaline metal), no mechanical means are used: the process takes place inside the reaction container under the effect of gravity after heating the source materials to 200–250°C. The absence of mechanical devices contributes to the reliability of the installation.

Currently, the MAGMA experimental-industrial plant (similar to the Mineral test rig) is being installed in the reactor building. It will be used to process accumulated radioactive alkaline metals. When this technology is implemented for the BN series of reactors, consideration should be given to designing and constructing a system for processing up to a 1000 litre batch of sodium at each time.

I.2.2. Neutralization (passivation) of non-drainable residues

The IPPE has developed a method neutralizing non-drainable residues of alkaline liquid metal coolants (Na, NaK alloy) with a gaseous suboxide of nitrogen (dinitrogen oxide, N_2O), which is characterized by the absence of hydrogen generation, improving the safety of the technology.

Research and experimentation established a mechanism of interaction between sodium, caesium and eutectic NaK alloy with gaseous suboxide of nitrogen. It was demonstrated that the intermediate compound that ensures complete consumption of the alkaline metal (Me) by the reaction is $Me_2N_2O_2$, hyponitrite of alkaline metal. Initial conditions of interaction were determined along with temperature effects and the composition of final product (a mixture of oxide, nitrate and nitrite of alkaline metal in various combinations) as a function of the duration of the neutralization process. The kinetics of the nitrogen sub-oxide absorption by the alkaline metal was established, composition of the gaseous phase was studied, and dependence on other parameters was determined.

Testing of the method was performed using NaK alloy residues in the transport tank and in the oxide hot trap of a terrestrial prototype of a spacecraft nuclear installation. The results of studies were positive: the entire alloy that remained inside the vessel (approximately 1.5 litres) and in the oxides hot trap (approximately 2 litres) was fully reacted and converted into salt and oxide compounds—$NaNO_2$, KNO_2, $NaNO_3$, KNO_3, Na_2O, K_2O_2. The compounds were then safely dissolved in water, also generating no hydrogen. The salt solutions were drained and disposed of, resulting in a final safe condition for the equipment.

In order to test the new technology for neutralization of non-drainable sodium residues and admixtures in the secondary circuit cold traps of the BR-10 reactor, the Luiza-T test rig (Fig. 22) was designed, fabricated and commissioned for trial operation. It was used for experimental verification of the neutralization technology for sodium residues present in the cold traps of the second loop of the secondary circuit. The amount of sodium with admixtures present in the cold traps was not known precisely in advance. Radioactivity of sodium residues was determined by the content of tritium, the specific activity of which was 4.0×10^6 Bq/kg.

The process of sodium residues neutralization inside the filtering filler and in the setter occurred in acceptable temperatures (maximum temperature levels did not exceed 340–350°C), without any pressure surges of the gaseous phase. Neutralization of non-drainable sodium with oxides consumed approximately 33 kg (about 16.8 m^3) of gaseous nitrogen oxide. This known quantity was used to determine that cold traps originally contained approximately 17 kg of metallic sodium. As a result of neutralization, all of sodium was converted primarily into salt forms (sodium nitrate, nitrite, hyponitrite), the contents of which in the final product measured 75–80% by weight.

It was also established that approximately 99.9% of radioactive tritium contained in non-drainable residues as sodium tritide (NaT), as a consequence of interaction with nitrogen suboxide, was eventually found in the final product, tritiated sodium hydroxide, represented as NaOT below:

$$N_2O + NaT \rightarrow NaOT + N_2\uparrow$$

Research has been performed to develop a conditioning technology to neutralize the final product by placing grout inside the cold trap itself. It was demonstrated that the most promising and workable method is incorporation into a geocement compound. Cold traps act as packaging for solidified radioactive waste, which may be sent to long-term storage or disposal. The method will be subjected to further testing on the cold traps of the first loop of the second circuit of the BR-10 reactor.

FIG. 22. The Luiza-T test rig.

APPENDIX II. BN-350 REACTOR SODIUM AND SODIUM-POTASSIUM REMOVAL AND PASSIVATION[8]

Removal of the physical and chemical hazard represented by the liquid metal coolant of a fast reactor is a key element of the initial decommissioning of this type of nuclear power plant. At BN-350, work has been in progress to achieve this objective since the final shutdown of the reactor in 1999. The overall aim of the BN-350 Liquid Metal Coolant decommissioning project is to process or passivate all the major inventories of sodium (Na) or sodium-potassium alloy (NaK) to allow the plant to be put into a “Safe Store” condition—a long period of storage with surveillance to allow radioactive decay before final dismantling.

The strategy of reducing activity levels by removing caesium from the primary sodium and for maximising the amount of bulk sodium able to be drained from the reactor simplifies the processing of bulk and residual sodium. Much of the reactor’s low activity secondary circuit sodium was re-used in other industrial processes, thereby minimizing the amounts of solid radioactive waste which will be produced. The Sodium Processing Facility is now operational and can be used in trial mode until the planned Geocement Stone Facility becomes available to allow processing of primary sodium to begin in earnest. Residual sodium in the main coolant circuits was passivated and studies are in hand to develop plans for the remaining Na- or NaK-contaminated plant items.

International collaboration has been a major feature of BN-350’s liquid metal coolant decommissioning activities. This initially brought in experience from the US EBR-II project, together with substantial US funding assistance for caesium removal, sodium draining, sodium processing and residual sodium process development. Latterly, it has been supplemented with United Kingdom experience from decommissioning of the Dounreay reactors, as well as funding assistance from the British Government. The combination of knowledge from abroad with the expertise of nuclear specialists in Kazakhstan has been an important factor in the overall success of the Liquid Metal Coolant project.

II.1. REACTOR DECOMMISSIONING STRATEGY

On April 22, 1999 the Government of the Republic of Kazakhstan adopted the Decree No 456 on Decommissioning of BN-350 Reactor Facility. The goals to be maintained while putting the reactor BN-350 in SAFSTOR status are the achievement of nuclear- and radiation-safe conditions and an acceptable level of industrial safety. Three basic stages for BN-350 reactor decommissioning are envisaged: transfer of BN-350 reactor facility to the state of long-term safety storage, long-term safe storage during 50-years, partial or complete dismantling of the equipment, of buildings, structures and burial of wastes. The goal of each stage is achieved by satisfying corresponding criteria.

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[8] All photographs used in the appendix are courtesy of Nuclear Technologies Safety Centre (NTSC).

During the stage of placing the BN-350 in safe long-term storage state, the main task in the area of handling the liquid metal coolant is its removal from the reactor, heat-exchange loops and equipment, bringing the liquid metal to a chemically inert, explosion-proof and fire-safe condition, and placing the products of processing in long-term storage. The strategy for performing this task includes:

- Purification of sodium from radionuclides of caesium;
- Drainage of bulk sodium;
- Removal of residues;
- Processing.

The sodium is to be processed using a facility similar to one at the Argonne National Laboratory West (ANL-W, USA) in the United States by injecting a melt of alkali metal (Na or NaK) into a concentrated alkaline solution. Then the alkaline solution will be processed into geocement stone, which produces a solid radioactive waste that meets the Republic of Kazakhstan's regulatory requirements for disposal.

II.2. LIQUID METAL SODIUM AND NAK REMOVAL

The status of removal, treatment, and handling of the various sodium and NaK materials are described below.

II.2.1. Secondary loops

The sodium from the secondary loops was drained, and placed into 100-litre drums. They were transported for recycling at the Ulba metallurgical plant in Kazakhstan.

II.2.2. NaK materials

The major volume of NaK alloy will be diluted into the sodium and stored with the primary loop sodium. The major volume of the NaK alloy from the cold trap cooling system will be drained into the primary sodium storing system and mixed into the sodium. For this purpose, it is necessary to modify the reactor facility technological scheme. Draining will be completed before the processing of primary sodium starts.

The NaK alloy residues will be removed from the cold traps by replacing NaK with sodium, thereby cutting off the traps from the system. The mixture of NaK alloy contaminated with oil will be removed from the tanks of the spent fuel bath cooling system, similarly to the technology used to remove the same mixture from spent fuel subassemblies drums. Presently, the technology of removing other non-drainable residues of NaK alloy from the system (system washing) has not yet been determined.

II.2.3. Primary coolant

The draining of sodium coolant from the primary circuit involved these steps:

- The primary circuit coolant was cleaned of caesium nuclides using caesium traps. A caesium trapping system for cleaning caesium radionuclides from the BN-350 reactor primary sodium was designed, fabricated, installed and successfully operated in

Aktau, Kazakhstan. This activity was performed within the framework of international collaboration between the USA and the Republic of Kazakhstan. As a result of this joint team work, about 255 300 GBq (6900 Ci) of caesium were trapped and the ^{137}Cs specific activity in the BN-350 primary sodium was decreased by a factor of 800 down to less than 0.37 MBq/kg (10 mCi/kg) with seven traps containing a total of about 16 kg of reticulated vitreous carbon.

- The project design does not involve draining the lower part of the reactor vessel (down from the inlets in the area of the high pressure plenum reflector). There are areas in the reactor vessel from which coolant discharge by the projected means is impossible. Complete draining required a number of measures, e.g., drilling holes in the central sleeve of the discharge manifold. A unique method for drilling of metal structures inside the BN-350 reactor vessel was carried out for liquid metal sodium at a depth of 13.4 m at a temperature of 280–300ºC. There is no analogue of such operation in the world practice. Draining of the primary sodium was successfully completed on December 1, 2003.
- About 610 m^3 of sodium was drained from the vessel of the reactor and loops of the primary circuit, and about 150 m^3 of sodium was drained from the reactor vessel; these were placed in safe storage prior to processing. This operation filled the drainage tanks with the primary sodium. The rate at which the reactor vessel cooled was slow enough such that there was no need to operate the gas heating system. One result of the draining was that the radiation dose rate decreased in the vicinity of the primary circuit location, which will benefit dismantling of the reactor. The estimated volume of the residual sodium in the primary circuit after draining is 18.4 m^3, 16 m^3 of which is located in the cold filter traps of the primary sodium cleaning system. Approximately 0.056 m^3 sodium comprising a layer with a thickness of 24 mm was left at the bottom of the reactor vessel.

The method selected for sodium dissolution involves injecting liquid sodium into an alkaline solution of NaOH. Construction of the sodium processing facility (SPF) and SPF startup activity with secondary circuit sodium has been completed. The system and integrated startup activities at SPF with secondary circuit sodium were completed at the end of 2008. About 610 m^3 of sodium and 20 m^3 of NaK alloy is projected to be processed into caustic, followed by conversion into geocement stone. The resulting product is compliant with Kazakhstan regulations for long-term storage and subsequent disposal of radioactive waste. The geocement stone facility is planned to be constructed in future.

II.3. SODIUM RESIDUALS PASSIVATION METHOD SELECTION

The comparative analysis of the different technologies from the points of view of safety, cost effectiveness and applicability showed that for the conditions of BN-350 the most applicable are the technologies of the steam-gas washing and technology of passivation with moist CO_2. The physical-chemical basis of hydrocarbonation technique consists of interaction of sodium and moist gas. Sodium bicarbonate $NaHCO_3$ (or sodium carbonate, Na_2CO_3) and hydrogen are formed as a result of this reaction in accordance with the following equations:

$$2Na + 2CO_2 + 2H_2O \rightarrow 2NaHCO_3+H_2$$

$$NaHCO_3+NaOHNa_2CO_3+H_2O$$

$$2Na + CO_2 + H_2O \rightarrow Na_2CO_3+H_2$$

The main technology selection criteria recommended for BN-350 was the necessity to passivate relatively deep (about 10 cm) layers of the sodium. The positive results of the passivation of the residual sodium of EBR-II proved the applicability of the hydrocarbonation technology for the BN-350 reactor.

The positive experiences of the laboratory tests and a full scale usage of the technology at EBR-II confirmed that the technology will successfully transfer in a chemically passive condition (passivate) the sodium residuals that are to remain in the “pockets” and on the bottom of BN-350 reactor vessel. The passivation using the technology of hydrocarbonation was carried out with a use of additional equipment such as special reagents producing system, exhaust gases control system, supply/drainage pipelines and other system features.

II.3.1. Laboratory scale development testing

The hydrocarbonation method was tested by experts at Argonne National Laboratory-West (ANL-W) for EBR-II reactor residual sodium coolant removal. Also, the method of residual sodium removal, using the wet CO_2, was developed by the French experts as the basic method for cleaning of the equipment of reactors Phénix and Rapsodie, and by Russian experts for residual liquid-metal coolants removal from the reactor equipment components. Results of passivation showed that the only limiting factor for present technology is the passivation rate reduction with an increased sodium hydro-carbonate layer thickness. At the present time this method can be evaluated as the safest one among other known methods of residual alkali metals passivation. The experiments at the development test facility mostly repeated the experiments performed by ANL-W researchers; they were conducted for the BN-350 application to test specific configurations that are difficult for passivation.

The development test facility consisted of an experiment chamber and several engineered systems and equipment for testing the residual sodium removal technology by hydrocarbonation method. The experimental chamber was a stainless steel vessel with observation windows, where the research samples were located. Carbonic gas was supplied to the system using a bubbler to obtain required relative humidity.

II.3.2. Development testing

Testing on residual sodium removal by wet CO_2 was conducted in four stages:

- Stage 1: Sodium sample passivation at environment (room) temperature and constant relative humidity of supplied carbonic gas (temperature in bubbler was maintained similar to environmental temperature). The samples were damaged due to uncontrolled temperature increase and next stages were carried out with a new set of samples;

- Stage 2: Sodium sample passivation (new set of samples) with step by step increase of temperature in the experimental chamber and constant relative humidity of supplied carbonic gas (temperature in bubbler was maintained at environment (room) temperature);

- Stage 3: Sodium sample passivation with step by step increase of temperature and relative humidity of supplied carbonic gas without condensation (water temperature in bubbler was kept 10°C lower than samples temperature in the experimental chamber to avoid condensation);

– Stage 4: Sodium sample passivation with step by step increase of temperature and relative humidity of supplied carbonic gas (water temperature in bubbler was kept 10°C higher than samples temperature in the experimental chamber to provide for water condensation and to react as much as possible sodium).

The highest reaction rate was observed at Stages 2 and 4, but it was attributed to cracking of glass with large quantity of sodium (Stage 4) and free surface of sodium at the experiment beginning (Stage 2). The Stage 3 reaction rate was lowest due to the large thickness of sodium hydrocarbonate. However, as the reaction at Stage 3 was restored after falling at Stage 2, the parameters of experiment Stage 3 were optimal for removal process. The conditions of Stage 4 can be also applied to react sodium layers at the final stages of passivation.

Samples of elements of equipment before filling with sodium and in the process of hydrocarbonation (Stage 2) are shown on Figure 23 and Figure 24.

FIG. 23. The samples, represented elements of BN-350 reactor equipment, which are considered to be difficult for passivation.

FIG. 24. Samples after 1566 hours after start of experiment.

Maximal sodium layer, reacted during the experiments, was 69 mm and the maximum thickness of the generated sodium hydrocarbonate layer was 125 mm.

Unlike the steam-gas washing, the hydrocarbonation method is safer and does not cause the fluctuation of temperature and pressure in the washed equipment. The hydrogen concentration did not exceed 3.5% by volume. This peak was a result of glass cracking and exposure of large sodium surface to wet CO_2 at elevated temperatures. The hydrogen concentration usually did not exceed 2.5% and it decreased exponentially in the course of testing. The development testing showed that the hydrocarbonation method can be used for residual sodium removal in the BN-350 conditions.

This development testing confirmed the INL laboratory studies that showed the Na_2CO_3 layer forms a porous structure allowing moist carbon dioxide (CO_2) to reach the metal sodium, which is located under the developing layer of Na_2CO_3. In the process of hydrocarbonation Na_2CO_3volume is increased due to the increase of molar volume and the porous structure of the product.

II.4. SODIUM RESIDUALS REMOVAL FROM THE REACTOR VESSEL AND PRIMARY CIRCUITS

The project for sodium residual removal by steam nitrogen technique and hydrocarbonation took place following spent nuclear fuel of the BN-350 reactors being removed, packed, stabilized, and placed in the reactor's cooling pond. Also liquid metal coolant was drained from the primary and secondary circuits, except for secondary sodium retained in the U-tube bundles of the intermediate heat exchangers. These steps were necessary prior to, removing the sodium residuals from the vessel and circuits of the reactor to ensure nuclear safety and to minimize the amount of residual sodium to be passivated.

The residual sodium is in the form of films left on the internal surfaces of equipment and pipes. The thickness of the sodium film depends on the temperature during and after drainage, the sodium purity and the physical configuration in the vicinity of these surfaces.

II.4.1. Sodium residuals passivation steps

The sodium residuals removal system using the hydrocarbonation was installed in the central hall of the BN-350 facility. The system included a feature for "post-washing" of the hydrocarbonized systems with water. Passivation of the residual sodium was conducted with the following steps:

- Before supplying moist CO_2 to the primary circuit, the equipment and pipelines of the dioxide gas system and off-gas system were purged with nitrogen followed by dry CO_2. This was necessary to remove residuals of air and to prevent ingress of air oxygen into the systems with residual sodium prior to introducing moisture;
- Dry CO_2 was moistened by injection into the bubbler from a gas cylinder. As CO_2 was moistened by passing through a water layer in the bubbler, it was sent into the equipment to be washed;
- Temperature, flow and relative humidity of the supplied gas were adjusted in order to control the rate of the reaction of moist CO_2 and sodium;
- All the off-gases were collected in a receiver, where all off-gases are accumulated before their discharge into the dedicated ventilation system. Monitoring of the off-gases hydrogen and oxygen content was done at the receiver;
- The off-gases passed through a water trap before their discharge into the ventilation system. The water trap prevents backflow of air from the ventilation system and ensures positive pressure in the reactor system;
- Removal of the sodium residuals is completed when hydrogen release ceases.

II.4.2. Results of hydrocarbonation of the reactor vessel and primary circuits

The sodium residuals removal system was installed, preparation of the equipment was conducted, and start-up activities were successfully carried out by August 2008. Removal of the sodium residuals from the primary circuit by hydrocarbonation then began and was completed in October 2009.

Hydrocarbonation was conducted under various operational modes; it was interrupted for long-term periods of time for a variety of technical reasons. During the whole period of hydrocarbonation 19 325 kg (9832 m^3) of CO_2 was supplied into the primary circuit. According to calculations an estimated 380 kg of residual sodium of the primary circuit was processed. The ratio of processed sodium and supplied CO_2 is therefore $380 kg / 19\,325 kg = 0.0196 kg\ Na/kg\ CO_2$.

Taking into account that the hydrocarbonation process lasted slightly longer than 126 days, the average rate of sodium residuals removal was about 3 kg per day.

After completion of the hydrocarbonation of the primary circuit, a tube bundle was removed from an intermediate heat exchanger to examine the completeness and effectiveness of the carbonation of sodium located on its surface. Figure 25 shows the process of removal of the tube bundle placed in the clean booth from intermediate heat exchanger. During visual examination it was observed that the tubes and casing had no adherent sodium and only slight quantity (traces) of a white substance—presumably, carbonation products. Survey of the internal part (primary circuit side) of the heat exchanger showed residual sodium to be completely carbonized on the internal surfaces of the intermediate heat exchanger vessel (see Fig. 26).

FIG. 25. Removal of the tube bundle.

FIG. 26. Examination of external surface and nozzle of intermediate heat exchanger.

The result of the work conducted supports the inference that all the main internal surfaces of the BN-350 primary circuit pipelines and equipment are completely carbonized. The quantity of metallic sodium remained in the primary circuit (in heels or other areas of thicker sodium) after carbonation is estimated to be 150 kg.

II.4.3. Analysis of further options for removal of sodium residuals from BN-350 primary circuit

As mentioned above, after removal of the primary circuit sodium residual water was to be supplied into the equipment and systems on completion of hydrocarbonation. The intention was that carbonate should dissolve in the water and the remaining sodium residuals, which didn’t react with moist carbonate dioxide, should react directly with the water.

The main disadvantage of this approach is the quantity of liquid radioactive wastes (~500 m^3) generated as a result of water washing of the equipment and systems. The processing of this amount of liquid wastes would require significant additional efforts. Taking into account that the issue of processing existing BN-350 reactor facility liquid radioactive wastes had not been resolved, it was not reasonable to generate new significant volumes of liquid radioactive waste. In addition, the effectiveness of the water washing is under question, since previous experience with washing of BN-350 equipment from residual sodium showed that there is no assurance that the metal sodium will be fully reacted due to the formation of a NaOH barrier. Furthermore, it is difficult to control the process of washing the equipment, resulting in local "micro-explosions" that could result in stressing of the structural materials of the primary circuit and accelerating degradation of its mechanical properties. An additional problem would be the formation of alkali in the primary circuit loops that will result in acceleration of corrosion of the pipelines during the proposed 50-year safe storage of BN-350 reactor facility with a consequent decrease in the safety level of this storage.

The results of sodium residuals carbonation described here support the recommended "*Leave It As It Is*" approach until the vessel and primary circuit loops of the BN-350 reactor facility are dismantled for system washing. This option does not have the disadvantages of generating of large amounts of liquid radioactive waste and creating an alkali, corrosion-active medium, as well as additional stresses in the vessel and the primary circuit loops. At present, the main surfaces of the primary circuit systems and equipment have been completely carbonized and residual metallic sodium in the form of heels being covered with a dry layer of carbonate. This state provides the facility with a high level of safety during 50-year storage. The subsequent dismantling of the BN-350 reactor facility would be carried out by using known and reliable technologies.

II.4.4. Hydro-carbonation of the secondary circuit loops

The estimated quantity of residual sodium in the secondary circuit was 1.93 m^3. The secondary circuit (mainly pipelines) was also successfully carbonized. About 200 kg of sodium was processed into sodium carbonate/bicarbonate. Sodium residuals in the pipelines of the secondary circuit after the first draining and after additional draining and carbonation are shown in Figures 27 and 28. Equipment of the secondary circuit (intermediate heat exchangers, main circulating pumps etc.) is separated from pipelines with plugs and will need additional efforts to be cleaned of their sodium residuals.

FIG. 27. Sodium residues in the pipelines of the secondary circuit after first draining.

FIG. 28. Sodium residues in the pipelines of the secondary circuit after additional draining and carbonation.

II.5. CONCLUSIONS

Passivation of the sodium systems of BN-350 has demonstrated:

- The effectiveness of decontaminating the primary sodium coolant from ^{137}Cs using caesium traps containing reticulated vitreous carbon (RVC) as a capture medium. The original concentration of ^{137}Cs in the BN-350 primary sodium as measured in 2001 following final reactor shutdown (296 MBq/kg) was reduced to 0.37 MBq/kg, a decontamination factor of 800. The key advantage of decontamination at this stage was in reducing potential dose rates to decommissioning personnel for subsequent operations. The seven traps used, containing some 255.3 TBq of radioactivity, were conditioned for long term storage by filling with lead in a process first adopted in Russia;

- The ability to deploy remote drilling equipment deep within the reactor vessel to successfully promote drainage of otherwise non-drainable sodium hold-ups;

- Drainage of a high percentage of the available sodium coolant from the reactor vessel, heat exchanger vessels and piped circuits into storage tanks, where the sodium can be retained in solid form with a nitrogen cover gas;

- Initial operation of the BN-350 Sodium Processing Facility (SPF) to process primary sodium into concentrated NaOH;

- Re-use of secondary sodium for industrial production of special metals;

- Acceptability of geocement stone, as developed in Kazakhstan to make use of indigenous materials, for the eventual immobilisation of radioactively contaminated NaOH from SPF for long term storage;

- In situ processing of residual sodium within the reactor, primary and secondary circuit loops using the carbonisation process to produce a passivated layer of sodium carbonate/bicarbonate. Over 380 kg of primary sodium and about 200 kg of secondary sodium has been converted into carbonate form. Examination following carbonation has shown that sodium on the main surfaces has been fully reacted. The amount of the remaining un-reacted sodium is now believed to be less than originally estimated. The current intention is to leave carbonate residue in situ during the SAFSTOR period.

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ABBREVIATIONS

ANDRA	Agence Nationale pour la Gestion Des Déchets Radioactifs (France)
BN-350	Fast Neutron Reactor
BR-10	Fast Neutron Research Reactor BR-10
CEA	*Commissariat à l'Energie Atomique* (France)
CRCTA	Composite Reactor Components Test Activity
D&D	Deactivation and Decommissioning
DFR	Dounreay Fast Reactor
DOTE	Dummy (false) intermediate heat exchanger
DSRL	Dounreay Site Restoration Limited
EBR-II	Experimental Breeder Reactor II
FFTF	Fast Flux Test Facility
IAEA	International Atomic Energy Agency
INL	Idaho National Laboratory
IPPE	Institute for Physics and Power Engineering
LIPOSO	Area of the junctions between the primary pumps and the core feeding plenum (Superphénix)
NaK	Sodium potassium
PFR	Prototype Fast Reactor
SFEN	French Nuclear Energy Society
SHS	Superheated steam
SPF	Sodium Processing Facility
USA	United States of America
VLLW	Very low level waste
WVN	Wet vapour nitrogen

CONTRIBUTORS TO DRAFTING AND REVIEW

Blynskiy, A.	Nuclear Technology Safety Centre, Kazakhstan
Dooley, K.	Idaho National Laboratory, United States of America
Farabee, A.	U.S. Department of Energy, United States of America
Fletcher, B.	AMEC Engineering Company, United Kingdom
Grandjean, J.	CEA, France
Grenouillet, J	EDF, France
Jones, J.	Idaho National Laboratory, United States of America
Ljubenov, V	International Atomic Energy Agency
Negin, C.	Project Enhancement Corporation, United States of America
Smykov, V.	A.I. Leipunski Institute of Physics and Power Engineering, Russian Federation
O'Sullivan, P.J.	International Atomic Energy Agency
Szilagyi, A.	U.S. Department of Energy, United States of America

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