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Managing Irradiated Graphite Waste

Final Report of a Coordinated Research Project

MANAGING IRRADIATED GRAPHITE WASTE

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MANAGING IRRADIATED GRAPHITE WASTE

FINAL REPORT OF A COORDINATED RESEARCH PROJECT

INTERNATIONAL ATOMIC ENERGY AGENCY
VIENNA, 2024

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FOREWORD

Graphite has been used as a moderator and reflector of neutrons in more than 100 nuclear power plants and in many research and plutonium producing reactors, in quantities ranging from a few kilograms to more than 3000 tonnes depending on the design. In several nuclear reactors graphite is used as fuel sleeve material, leading to the generation of large amounts of less irradiated, but still significantly radioactive, material.

Many of the older reactors have now been shut down, with more approaching the end of their operating lives, and some 250 000 tonnes of radioactive graphite (irradiated graphite, hereinafter referred to as *i*-graphite) have now accumulated worldwide. Progress towards the development of ultimate disposal solutions remains slow, with increasing amounts of *i*-graphite residing in temporary storage facilities awaiting processing for disposal. The pressure to resolve these issues differs widely between Member States depending on the dismantling strategies envisaged by their regulatory authorities for waste management. There is an increasing sense of urgency now to make substantial progress in Member States where it is government policy to commence reactor dismantling in the near future, and this is driving international efforts to further explore the detailed characterization of this waste material (*i*-graphite) as well as potential processing and disposal options.

To support Member States in resolving *i*-graphite management issues up to the industrial implementation of processing technologies, in 2016 the IAEA launched the International Project on Irradiated Graphite Processing Approaches (GRAPA), and held four technical meetings between 2016 and 2019. This publication provides a comprehensive overview of the planned management of *i*-graphite by the members of GRAPA and will serve as a reference for future IAEA publications regarding the disposal of graphite waste.

The IAEA wishes to acknowledge the valuable assistance provided by the contributors listed at the end of the publication. The IAEA officer responsible for this publication was W. Meyer of the Division of Nuclear Fuel Cycle and Waste Technology.

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CONTENTS

1. INTRODUCTION

1.1. BACKGROUND

The IAEA initiated various consultancies and technical meetings regarding the disposal of *i*-graphite (irradiated graphite). The conclusion from the first IAEA 'Technical Meeting on Graphite Moderator Life Cycle Behaviour' at The University of Bath, UK [1], indicated the need for an international database on nuclear graphite properties to preserve knowledge for developing future of graphite-moderated reactors and their subsequent dismantling and disposal. Further consultancies followed [2-5], and publications addressing issues of characterisation, potential treatments and conditioning of *i*-graphite, as well as disposal options intended to assist Member States [3-4]. Between 2006-2021 there were several international initiatives to address the management of *i*-graphite as indicated in Fig. 1.

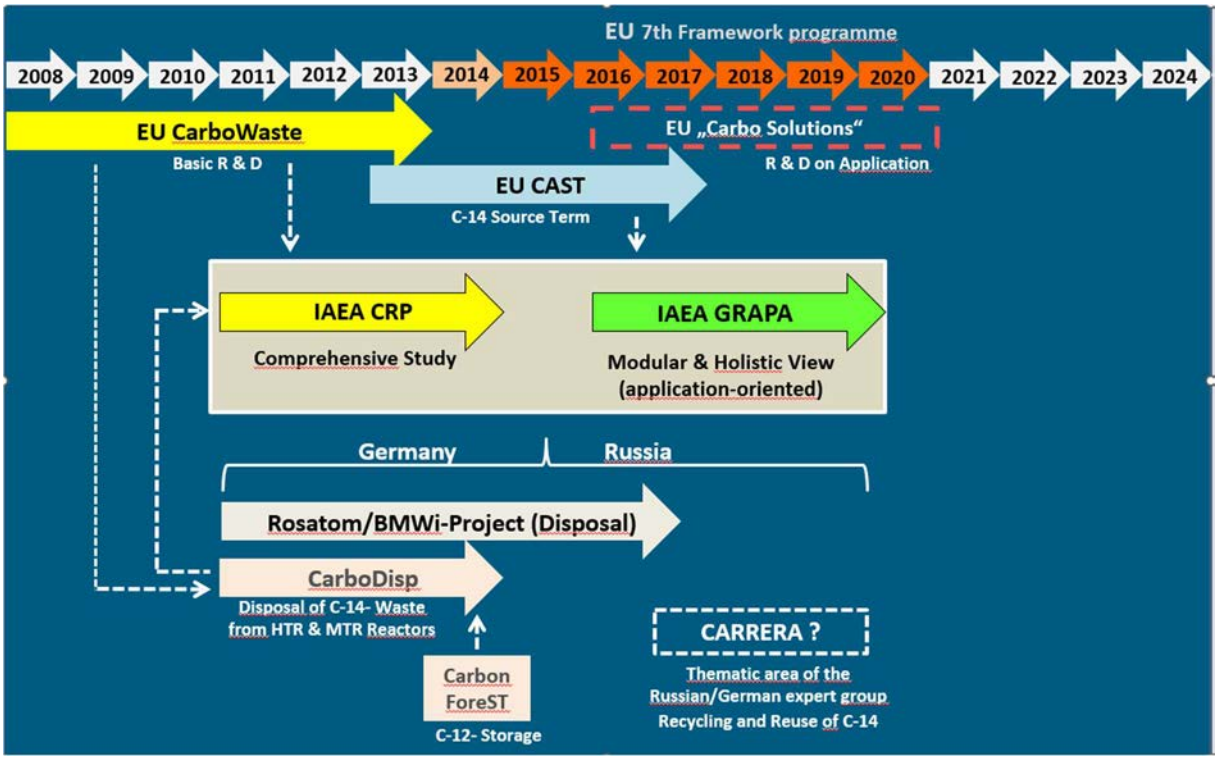

FIG. 1. International initiatives to address the management of i-graphite.

For instance, the Electric Power Research Institute (EPRI, USA) established a 'graphite reactor decommissioning network' and published in a sequence of review papers [6-12]. This information has been studied by Electricité de France (EDF, France) in support of the dismantling of the Natural Uranium Fuelled Graphite-Moderated Reactor (UNGG). Additionally, the Collaborative Research Project on Carbon-Based Nuclear Wastes (CARBOWASTE (2008 – 2013)) was established under the auspices of the 7th Framework Programme of Euratom (EU) with the specific objective 'to develop an integrated waste management concept for treatment and recycling of *i*-graphite'. This programme consisted of 30 participating organisations, from both within and outside the EU. Publication of detailed data from CARBOWASTE studies is not currently available, but it is hoped that publications will become available to complement the presently available summary publications [13-14].

The IAEA organized a coordinated research project (CRP) entitled 'Treatment of Irradiated Graphite to Meet Acceptance Criteria for Waste Disposal (T21026)', thereby expanding the number of participating States Member from the original CARBOWASTE group. The outcome of this CRP was a publication [15] providing information to support decommissioning and dismantling activities on graphite moderated reactors.

Other publications [16-23] cover graphite disposal options, including reflections upon the socio-economic aspects of the issue. Another initiative between 2016-2018 was the EU project on Carbon-14 Source Term (CAST) aimed to develop a better understanding of the generation and release of ^{14}C from radioactive waste under conditions relevant to waste packaging and disposal to geological disposal facilities (with 33 participating organizations) [21].

With many research programs ongoing, the IAEA proposed a network aimed at advancing the practical application of the knowledge gained on *i*-graphite to facilitate current and future dismantling/disposal programmes. The network, International Project on Irradiated Graphite Processing Approaches (GRAPA) was established, and initial contents were the disposal of ^{14}C from the reflectors and fuel of the German reactors 'Arbeitsgemeinschaft Versuchsreaktor' (AVR) and Thorium High-Temperature Reactor (THTR) since it had been realized that the total ^{14}C activity from these sources would exceed the licensed capacity of the KONRAD (former iron-ore mine in Germany) disposal facility. The scope of GRAPA network evolved through several expert Consultancy Meetings and Technical Meetings [24]. Whilst most of the contributed work relates to a single sub-topic, some projects undertaken by the contributing network members cover multiple areas (Fig. 2) [25].

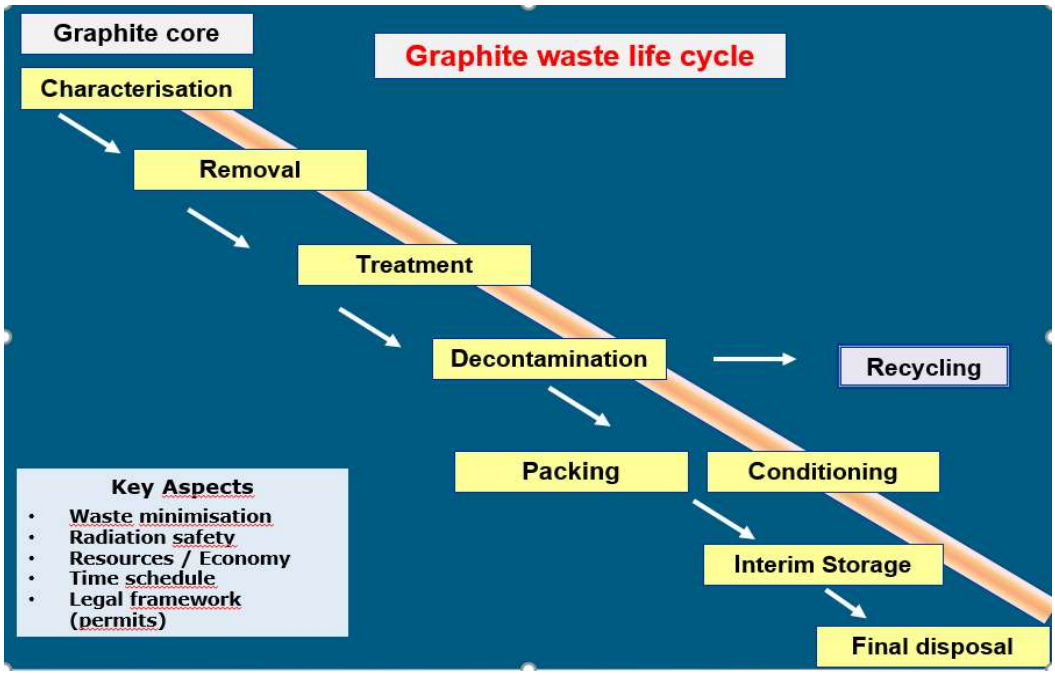

FIG. 2. Projects undertaken by GRAPA to address the management of i-graphite.

In addition, recognizing that research and development activities relate to the practical tasks in dealing with the *i*-graphite in reactors planned or under decommissioning, the following graphite core reactors have been chosen to represent specific reactor types in the network:

— Ignalina Nuclear Power Plant (INPP, Lithuania);

— Chernobyl NPP (Ukraine);

— Latina NPP (Italy), representative of the Magnox design;

— Chinon NPP (France);

— Seversk NPP (Russia), representing production reactors and from which the first graphite has recently been removed;

— Canada India Reactor Utility Services (CIRUS) research reactor at Bhabha Atomic Research Centre for which comprehensive planning for dismantling has been undertaken;

— L-54M research reactor (Politecnico di Milano, Italy);

— WWR-S research reactor (Horia Hulubei National Institute of Physics and Nuclear Engineering (IFIN-HH), Romania).

1.2. OBJECTIVE

The objective of this publication is to provide Member States with relevant information in support of minimizing radioactive waste that could arise during the life cycle of NPPs. This objective will be achieved by reviewing the implementation of the waste management principles into the current practical applications, and taking into consideration the regulatory, technical and financial factors influencing waste minimization practices at NPPs.

1.3. SCOPE

The publication describes and provides guidance on various proven methodologies, practices, and approaches to minimize radioactive waste during the design, operation and decommissioning of NPPs and other nuclear facilities related to management processes of waste arising from NPPs. The information it provides could also facilitate documenting the planning of new facilities development and decommissioning activities. Waste minimization by the use of processing technologies during operation and decommissioning, spent fuel minimization activities, gaseous discharge activities (draft publication to be published available) as well as non-radioactive operational waste arisings (as described in various Agency publications) are excluded from this publication.

1.4. STRUCTURE

The main body of this publication is divided into eight sections including the introduction in Section 1 and the conclusions in Section 8.

Section 2 expands on the factors influencing waste minimization management. Section 3 summarizes the available liquid waste minimization strategies. Section 4 suggests wet solid waste minimization strategies while Section 5 indicate approaches to minimizing dry solid waste. Section 6 provides information regarding the influence of metal composition on waste arisings and Section 7 suggest methodology to monitor, assess and benchmark waste minimization programs.

Finally, the IAEA-TECDOC is complemented by references, annexes providing relevant international case studies and experiences on specific aspects of nuclear waste minimization and abbreviations.

2. CHARACTERIZATION

2.1. INTRODUCTION

The 'Characterization work package' within GRAPA has been coordinated by Dr Grigorijus Duškesas of the Centre for Physical Sciences and Technology (CPST), Vilnius, Lithuania.

The objective of characterization is to capture fully the features of the *i*-graphite which are relevant to subsequent treatment, processing, storage and/or disposal, ideally before any retrieval process has been implemented. Recognizing that the different Member States have very different philosophies regarding *i*-graphite disposal, it may be that the results of the characterization are used to determine a handling/treatment/storage/disposal strategy or that the extent of characterization deemed necessary is determined by a strategy which has already been decided upon. Early reactors were designed, constructed, operated without clear preparation for their eventual shutdown and dismantling. During dismantling, more measurements could have been done to facilitate the characterization of the graphite as a future waste stream. For example, graphite was regularly removed for analysis from the UK Magnox reactors to determine chemical, physical and mechanical-property changes in support of operational safety cases and fault scenarios: many such samples could have been utilized for determining the build-up of radioisotopes, but this work was not undertaken before the samples were disposed of. In addition, for most of these shut-down reactors, the capability to extract new material for characterization ahead of major dismantling activity has been lost because of early disposal of the necessary pile-cap equipment to extract it [26].

The importance of this type of historical omission becomes clear when one understands that no two sources of *i*-graphite will never have the same characteristics, even from reactors of similar design. Operational history is important: for example, in gas-cooled plant where the coolant is circulated directly through steam-raising units, oxidation products from those units may find their way into the graphite pores and subsequently become activated in addition to the initial impurities present, leading to unpredictable variations in activity and in isotopic content. Even the impurities originally presented in the graphite are not uniformly distributed: individual graphite production heats are subject to variations in procedure (time/temperature and the efficiency of the purification process used nominally to control boron content) and there are important differences according to the position of individual graphite blocks within the coke-covered 'stacks' which are employed in the industrial baking and graphitization processes. In practice, one finds significant property variations even within a single block of graphite.

Thus, one may have only some generalized impressions with which to build a dismantling process such as, in the UK case, Advanced Gas-Cooled Reactor (AGR) graphite is much more radioactive than Magnox graphite (it contains a more generous contribution activated to 60Co), and the graphite is a different type (Gilsocarbon-based and quasi-isotropic compared with anisotropic petroleum-coke based extruded material) [26]. There will be better information about the changes in physical and mechanical properties which will contribute to the ability to remove and handle the graphite dimensional change, distortion and potential jamming of components, strength, resilience to impact, and so forth.

On the other hand, uncertainty regarding the decommissioning strategy to be followed may lead in other situations to decisions to undertake more characterization than is necessary for the procedures finally chosen. This can have cost implications and may lead to delays. Wide variations in the degree of 'preparedness' for *i*-graphite disposal in different Member States has led to characterization work continuing to dominate the interests of members of this GRAPA project.

The previous CRP [15] sought to characterize *i*-graphite under four main headings, with the intention to define detailed management strategies for each and thus to better understand the different approaches to dismantling and disposal which were needed. These four categories were:

— Fuel-contaminated graphite, typically classified initially as high-level waste (HLW), that needs immobilization or some form of treatment before being accepted for interim storage or disposal;

— Graphite, that cannot be treated with available processes (due to some technical or non-technical reasons) but can only conditioned for storage or disposal;

— Treatment-expedient graphite where a clear benefit can be seen (reduction in radioisotope content leading to a lower waste category classification) and leading to a management route which may lead to an overall cost saving (allowing reuse of isotopes or recycling of the graphite into carbonaceous products for the continuing nuclear industry;

— Decontamination-expedient graphite, with the aim of reducing the material to free release.

Whilst recognizing now that this classification is over-simplistic, the CRP [15] sought to create a matrix whereby categories of isotopic speciation, structural behaviour including dust generation, propensity to gas release and leaching, Wigner energy content, susceptibility to selective oxidation and so forth were aligned with the four classes. Such activities were intended to support specific elements of the decision process, such as whether there was a need to dismantle a plant under a water blanket or to undertake the process in air. As a result of information from the network, a major decision regarding the need for 36Cl analysis has been reconsidered, illustrating the difficulties faced in aligning the extent of characterization required with handling and disposal plans.

This publication now identifies the characterization work undertaken in GRAPA in each of the eight sub-topics. There has been some deviation from the planned output due to the prioritization of work within the participating organizations but, in general, most planned deliverables have been achieved. In all cases, the network members have provided a detailed technical report on their work which is provided in the Annex I: thus, only short summaries of the most significant points are provided here.

2.2. PUBLICATION REVIEW

Summarized from Annex I, Section 1.

The network members in this sub-topic were ANDRA, Atomic Energy Commission (CEA) and EDF in France, and Radioactive Waste Management Ltd. (RWM) in the UK. The objective was the collection and dissemination of data on *i*-graphite from previous projects [27-30] which are appropriate for planning future activities and as a basis for a comparison of the characteristics of different types of *i*-graphite as well as examining the relevance and utility of developed *i*-graphite processing methods for different graphite sources.

2.2.1. Publications from French organizations

The French organizations (ANDRA, EDF and CEA) have published an internal work document ('Graphite Reference Guide: State of Knowledge', FR.PA.SCM.15.0026/A, 2015) for *i*-graphite management from EDF and CEA facilities that covers the following content:

— Background and content of the publication;

— Graphite and types of reactors in which the graphite is used;

— Graphite production life cycle;

— Radiological inventory of *i*-graphite;

— Multiscale structure of *i*-graphite in reactor;

— Wigner energy;

— ^{36}Cl in *i*-graphite;

— ^{14}C in *i*-graphite.

2.2.2. Publications from European Commission

The European Commission project on Carbon-14 Source Term (CAST) considered all potential sources of ^{14}C in radioactive waste arising from reactor decommissioning, and the consideration of *i*-graphite was covered in CAST work package 5. All CAST publications are freely available [21] including the final publication from the *i*-graphite WP5 at [31]. Twenty international specialists were engaged upon this work package and the reported work involved institutions in France, Lithuania, Italy, Germany, Spain, Romania, Ukraine, and the United Kingdom. It is not appropriate to cite their detailed results here, however, a short summary related to activities carried out in GRAPA are:

— French National Centre for Scientific Research (CNRS) and Institute of Nuclear Physics of Lyon (IPNL) in France conducted ion-irradiation of highly oriented pyrolytic graphite, utilizing 13C implantation as a surrogate for ^{14}C to simulate the displacement of the isotope from its original structural sites in the graphite, with the intention to clarify the understanding of the mobility of ^{14}C and, thus its susceptibility to removal treatment processes;

— The Lithuania Energy Institute provided a detailed theoretical model for 14C in the GR-280 moderator graphite of the INPP;

— Regia Autonoma Tehnologii Pentru Energia Nucleara (RATEN ICN (Romania)) investigated ^{14}C leaching from the graphite of the thermal column in a TRIGA reactor in a cementitious environment and found very low release rates under both aerobic and anaerobic conditions, with organic species released in larger quantity than inorganic;

— EDF and ANDRA provided leaching data from UNGG moderator graphite and assessed that just 30% of the released species were organic;

— National Agency for New Technologies, Energy and Sustainable Economic Development (ENEA (Italy)) investigated ultrasonic exfoliation in both complex organic solution and in aqueous sodium hydroxide;

— Forschungszentrum (FZJ (Germany)), concerned about the limits on disposal of ^{14}C graphite in the KONRAD facility, employed graphite from the Rossendorf research reactor in leaching tests and found that the release of inorganic species was dominant; the extent of release of ^{14}C in a thermal pretreatment was insufficient to justify the costs of so doing for reflector material from AVR and THTR;

— Centro de Investigaciones Energéticas, Medioambientales Tecnológicas (CIEMAT-(Spain)) investigated leaching and thermal treatment on Vandellos graphite and encapsulation with Bentonite and within a glass matrix to minimize such releases;

— Leaching of 14C and 3H was evaluated for the thermal column of the Romanian VVR-S reactor;

— RWM (UK) determined that graphite from the Oldbury Magnox reactor released to solution with only a small proportion of the ^{14}C released appearing in the gas phase: it was found that a substantial fraction of the ^{14}C was non-releasable.

It is of interest to note that differing results on the organic/inorganic species ratio for ^{14}C were found by different organizations with different graphite materials, and this is common to GRAPA work which is described later. The CAST WP5 results were applied by the project to a safety case for a GDF cementitious environment. The WP5 report contains references utilized by the different research groups, unfortunately not directly correlated to the report text. Overall, the result of the CAST work package may be summarized as follows:

— A substantial fraction of the ^{14}C in *i*-graphite is non-releasable;

— ^{14}C release cannot be defined by a single rate constant;

— ^{14}C is released in both gaseous and aqueous phases;

— ^{14}C released to the gas phase may exist in different species (CH4, CO2 and CO);

— Release rates and speciation of the released 14C depends on leaching conditions (pH and presence of oxygen).

2.3. IMPURITY DISTRIBUTION

Summarized from Annex I, Section 2.

Two significant pieces of work have been undertaken under the topic of impurity content of virgin nuclear graphite. Such knowledge enables predictions of eventual isotopic content and activity after irradiation in reactor for at least those sources originally present in the graphite. It cannot, of course, account for any material transferred to the graphite from other origins (drawn into the pore structure by the flow of coolant gas) and then subsequently activated.

2.3.1. Information from University of Manchester

The University of Manchester (UK) has drawn together the known information for the graphite used in the British experimental pile zero (BEPO) research reactor at Harwell, the extensive fleet of Magnox reactors (26 in total), and finally the fourteen AGRs. The situation for the last two is complicated because there were changes in the sourcing of pitch and coke during the manufacturing phase, two different manufacturers were involved for the Magnox fleet (Anglo Great Lakes (AGL) and British Acheson Electrodes Ltd (BAEL) and three for the AGRs (Union Carbide Ltd having taken over BAEL by the time the final four reactors were constructed, and the refining plant for the Gils carbon coke in the USA having had to be rebuilt following a fire). Thus, it is not surprising that there are some important differences in the impurity of the graphite in different reactors, which means that the results of subsequent radio-isotopic surveys cannot easily be applied to other reactors of a similar type even when allowance is made for different total fluence. A major difference between the Magnox graphite and the AGR graphite is found in the cobalt content (as ^{60}Co) which is very much higher in the latter, an unintended

consequence of the application of standards when AGRs were designed which did not exist for the earlier reactors, whose designers took exceptional care to specify the need to minimize impurities as much as possible [26].

2.3.2. Information from Politecnico di Milano

A second important contribution comes from POLIMI (Italy), where virgin USA Graphite Grade (AGOT grade) graphite representative of the L-54M reactor has been investigated. Ground samples were subjected either to acid digestion or combusted to provide residual ash: the samples were then analysed by Inductively-Coupled Plasma Mass Spectrometry (ICP-MS) [32]. It is intended to confirm the results using Prompt Gamma Neutron Activation Analysis (PGNAA) which will provide further information on light and volatile impurities (Li, B, N and Cl) which contribute to the generation of the weak beta-emitting isotopes ^{3}H, ^{14}C and ^{36}Cl. Furthermore, depth profile distribution of N and Cl will be investigated, to attempt the justification of ^{14}C and ^{36}Cl depth distribution by higher surface absorption of their precursors. These results are not here reported as the experiments will be performed after the completion of this publication.

2.4. MODELS OF REACTOR GRAPHITE ACTIVATION

Safety Summarized from Annex I, Section 3.

As explained in the preceding section, data on virgin graphite impurities feed into the modelling of subsequent activation. The reactors which have received attention under the GRAPA project are the 'Reaktor Bolshoy Moshchnosti Kanalnyy' (RBMK) at INPP and L-54M at POLIMI. with handling and disposal plans.

Building upon previous work undertaken by the Lithuanian Energy Institute in support of INPP [33-34], a major study has now been completed by the CPST in support of INPP (and, by inference, other examples of the RBMK-1500). The intention was to provide a comprehensive modelling methodology which, through comparison with the limited experimental data, provided confidence in predictions in other regions of the graphite stacks without the need for comprehensive sampling and measurements.

Identification of the impurity concentration in the virgin RBMK-1500 graphite was previously performed by neutron activation and mass spectrometry methods [35-38]. The results indicated that the concentration of activation products in the GR-280 reactor graphite strongly depends on the activation process (neutron flux during the reactor operation, burnup conditions, and the reactor power history). According to the existing radiological classification the graphite waste is attributed to the long-lived low and intermediate activity waste [38]. To obtain the alpha, beta, and other nuclides content in the graphite waste without the costly and time-consuming experimental measurement procedure, a scaling factor technique is applied [39-40]. Briefly, the scaling factor is based on the perceived dependence between specific activities of nuclides in the investigated sample when the main pollution source is the same:

$$A_i = k_i \cdot A_{key} \qquad (1)$$

where A_i is the specific activity of the difficult-to-measure radionuclide, A_{key} is the specific activity of the easy-to-measure key radionuclide, k_i is a constant called the scaling factor.

The scaling factors of radionuclides, with the specific activity of which can be measured by γ-, α- and β-spectrometric methods, are statistically determined according to the correlation of the investigated radionuclide with the key nuclides. On the other hand, the experimentally validated model of the RBMK graphite can be successfully used saving time and economical resources for the radioactive graphite waste characterization. Models of different complexity could be used depending on the radioactive waste stream, the sorting technique (if applicable) and uncertainty of measured radionuclides. For the scaling factors determination, usually both the model calculations and measurements of some representative samples from the certain radioactive waste are used [41].

2.4.1. Information from Centre for Physical Sciences and Technology

The Centre for Physical Sciences and Technology (CPST) contribution is to analyse the neutron fluence in different parts of the reactor graphite to identify the regions which have the same radiological characteristics of neutron activation. Although the reactor power history is known, changes of the neutron flux intensity and spectrum with the radial and axial position in the reactor graphite moderator cause inhomogeneous activation of impurities. The neutron activation calculation considering variation of irradiation conditions in different parts of the reactor core can influence the limits of application of the scaling factor used for characterization of the same material (graphite). The full 3D reactor core model, ¼ 3D reactor core model and a simplified 3D 4x4 core plateau fragment of the RBMK-1500 reactor have been created using Monte Carlo N-Particle 6 (MCNP6) [42] and Modelling and Simulation Suite for Nuclear Safety Analysis (SCALE 6.1) [43] code packages, respectively. A full scale (MCNP6 model) and 4x4 core 3D (SCALE6.1) approaches for characterization of the RBMK-1500 graphite have been developed in earlier studies [37] and [44-45]. It is not practical to use the full-scale 3D model due to neutron propagation (k_{eff} convergence, entropy of the fission source distribution, takes considerable computer time) difficulties in the large core calculation case. A ¼ core 3D model has been developed which overcomes full-scale model difficulties and which is necessary for representation of periphery zones and the graphite reflectors. The present improved calculation of the neutron energy distribution and fluence variations in the different parts of the reactor core has revealed the optimal separation of the regions with the same radiological characteristics of neutron activation in the core graphite. On the other hand, again, for neutron activation sensitivity studies, the use of the MCNP model takes considerable computer time. The equivalence of the ¼ core model and the 4x4 core fragment model has been validated before applying the simplified approach for sensitivity study analysis when dealing with the reactor plateaux region of the reactor. The activation calculation sensitivity analysis due to neutron power variation in the reactor core plateau region and in the reactor core periphery region for important nuclides has been obtained.

The Lithuania Energy Institute has conducted work under the CAST framework, predicting the ^{14}C in the GR-280 graphite components of the stack [31].

2.4.2. Information from Politecnico di Milano

Modelling of the L-54M reactor included the core with the cooling coils and the fuel solution, the graphite moderator and reflector, the experimental irradiation channels and the heavyweight concrete biological shield, using the MCNP6 code [32]. Figure 3 contains more detail of the Politecnico di Milano L-54M reactor, identifying the graphite components that formed part of the modelling model.

FIG. 3. The L-54M reactor. (Courtesy of POLIMI).

The simulation was run in three stages: simulation of the reactor itself, of the main nuclear reactions, and of the activation of the graphite components. The model was verified using experimental data obtained during the early operational years of the reactor, such as criticality data and normalized flux curves in different positions inside the experimental exposure channels.

Comparisons of the results with the available sample radiological measurements have proved encouraging [46-47.

2.5. RADIOLOGICAL CHARACTERIZATION

Summarized from Annex I, Section 4.

The principal contributions in this area relate to INPP and to the POLIMI L-54M reactor.

INPP and CPST have collaborated upon the acquisition of samples of RBMK-1500 *i*-graphite and upon obtaining radionuclide measurements. By the end of 2019, the sampling campaigns for both RBMK-1500 graphite stacks were completed, and non-destructive measurements of gamma-ray emitting nuclides as well as destructive analysis of selected samples were performed.

It was found that the ^{14}C mass activity in the graphite bulk samples is distributed quite homogeneously, i.e., activity varies in the range of $(1.4 – 3.0) \times 10^5$ Bq/g. The mass activity of alpha-emitting actinides varies in a wide range. For the same nuclide, mass activity differs by up to 2 orders of magnitude in different samples. This is most probably indicating significant heterogeneity of impurities in the samples.

On the L-54M reactor, to corroborate the results of a very preliminary radiological characterization and to test the outcomes of the theoretical modelling mentioned above concerning the generation of radionuclides by the neutron activation process, several sampling points have been identified in the accessible parts of the graphite monolith. Samples (17 samples) were drilled from the external surfaces and along the length of two extractable graphite rods of the reflector. A high purity germanium (HPGe) detector was employed to evaluate the gamma emitting radionuclides present ($^{152,154}Eu$, ^{133}Ba and ^{137}Cs primarily [47]). Moreover, a sequential radiochemical procedure has been developed and optimized through one M.Sc. thesis to allow the simultaneous determination of several hard to measure radionuclides (^{3}H, ^{14}C, ^{36}Cl, ^{41}Ca, ^{55}Fe, ^{59}Ni, ^{63}Ni, ^{90}Sr and ^{129}I) in each *i*-graphite sample.

2.6. LEACHING INFORMATION

Summarized from Annex I, Section 5.

The UK National Nuclear Laboratory (NNL) carried out a small simple experiment to investigate the link between the mobile fraction of 14C observed in leaching studies and 14C rich carbonaceous deposits from pile grade A (PGA) graphite typical of the UK Magnox reactor stacks.

The results showed that the removal of ^{14}C rich carbonaceous deposits did not eliminate the initial rapid release of ^{14}C from the matrix [48].

Regarding *i*-graphite waste, the mechanisms of ^{14}C release during storage conditions (organic, inorganic, dissolved or gaseous species) is crucial in the designing of a storage/disposal facility. At the El Cabril facility, as there is a limitation of 20 TBq on the total amount of ^{14}C that can be disposed of and based on available disposal cells, disposal of *i*-graphite waste will be limited. The first studies investigate the behaviour of the stable impermeable graphite matrix (IGM) without radioactive contaminations under Spanish disposal conditions. Leaching experiments are performed with virgin graphite and the corrosion mechanism is deducted from the mass loss and release of corrosion production into the aqueous phase through chemical analysis of the leachant. When obtaining the results of this first phase demonstrating the applicability of the IGM, a second phase using *i*-graphite from the Vandellós I NPP will be performed to investigate the leaching behaviour of incorporated radionuclides.

2.7. WIGNER ENERGY

Summarized from Annex I, Section 6.

The GRAPA members are aware that the subject of Wigner energy frequently raises concerns among safety authorities, engineers, and scientists unfamiliar with the issue. This is understandable, given the memory of the accident in Windscale Pile No. 1 in 1957 which was very accurately and openly published by Arnold [49]. Indeed, that accident very much concerned the operators of the BGRR on Long Island, USA, since it was operated under very similar conditions to the reactors at Windscale: however, it can be noted that, a safe and successful demolition of the BGRR graphite stack was undertaken by direct mechanical fragmentation, in air and at ambient temperature, with no risk of any Wigner energy release occurring [23].

The Oak Ridge National Laboratory (USA) have published a useful historical record of Wigner energy investigations in support of the development of the High Temperature Reactor [46]. A comprehensive analysis of the issues which need to be considered in dismantling a graphite reactor stack from the viewpoint of Wigner energy release has been conducted as part of GRAPA and includes an analysis of detailed measurements covering Brookhaven Graphite Research Reactor (BGRR), Windscale Piles, Magnox reactors, research reactor at Bhabha Atomic Research Centre (CIRUS (India)) and research reactor of Horia Hulubei National Institute of Physics and Nuclear Engineering (WWR-S (Romania)).

Wigner energy, or ‘stored’ energy in solid graphite, is a result of the displacement of carbon atoms from their original position in the graphite crystal lattice as well as ‘vacancies’ in the lattice. The displaced atoms (and indeed the vacancies) can become mobile and combine in various ways. Classic books on graphite irradiation damage [50-51] discuss the formation of ‘lines’ of atoms and new hexagon-based structures and provide information about their differing mobilities. However, it is now realized that the nature of structures arising from these atomic

displacements is very much more complex [52] and this goes some way towards explaining the observable behaviour of the release of energy when the graphite is heated.

Several basic principles need to be noted:

— The rate of Wigner energy accumulation is inversely related to the operating temperature of the graphite component, and only becomes significant in regions where graphite operating temperature is below ~200°C;

— Wigner energy accumulation is mitigated by thermal annealing during irradiation, and by irradiation-induced annealing (permitting a proportion of atoms to return to 'normal' lattice positions) – this may lead to an effective 'saturation' of Wigner energy when the rate of fast-neutron-induced structural 'damage' matches the rate of annealing;

— A release can be achieved only if the graphite is subsequently heated to around 50 K above its final operational temperature;

— Wigner energy is measured in three ways:

  - By bomb calorimetry or similar methods, which gives a measure of total Wigner energy per unit mass of graphite;
  - By measurement of the fractional change in thermal conductivity (expressed as (k_0/k_{-1})), to which it is directly related for a particular grade of graphite;
  - By differential scanning calorimetry, which measures the rate of release as a function of increasing temperature.

— Only the third of these methods gives a meaningful result in terms of potential release at any stage of the *i*-graphite management process: this parameter has the same units as specific heat capacity;

— A spontaneous release leading to an unexpected temperature increase can only occur if the rate of release exceeds the specific heat capacity (Cp) at the temperature of the graphite and the graphite is in an adiabatic or near-adiabatic environment;

— Finally, Wigner energy cannot be released in any other way, such as handling, impact or even drilling and cutting.

Most of the legacy *i*-graphite presents no significant hazard during dismantling and handling, even where there are measurable accumulations. In Magnox-reactor graphite, for example, the lower regions of the core (temperature range about 190-230°C) contain quantities of stored energy which have saturated and do not approach the nominal operational safety-case limit of 0.8 Cp. As this material will never be exposed to temperatures 50K higher than that operational temperature range except in the most improbable accident scenarios, there is no risk of any significant release, with one specific exception discussed below.

The members of the GRAPA project are dealing with *i*-graphite which has been irradiated under a range of different conditions. One specific item which has engaged particular attention is control-rod displacers from RBMK reactors, which are exposed to very high flux but at low temperature because of their immersion in water-filled tubes [53]. Measurements are reported in more detail in Annex I Section 6 indicating the potential for release if they are exposed to modest heating and, in consequence, a separate strategy must be followed when handling them: small regions of the RBMK cores may have material with a low operating temperature which requires careful evaluation. However, since the average temperature of the RBMK graphite

stack during operation was about 500°C, substantial amounts of Wigner energy are not accumulated in the graphite overall.

For the disposal of *i*-graphite from small reactors, measurements are being performed to confirm the present levels of Wigner energy. As some of those measurements have been based on total stored energy, quite high levels have been noted. However, the value based on high level of total stored energy is not particularly significant, not only because it requires a temperature rise of almost 2 0000C to remove stored energy in its entirety but recognizing that it is the potential rate of release of stored energy which is the important parameter and that this can be effectively managed by designing appropriate handling processes.

A further point of principle is that if the *i*-graphite is to be deliberately subjected to any thermal treatment process, such as those considered in Section 3, then, subject to consideration of manner of any energy release during that treatment (rate of release, avoiding adiabatic conditions), a proportion of the Wigner energy will have been removed post-process and the residual material can be subject to much less rigorous controls from this standpoint.

One point must be kept in mind in relation to storage and is especially important for *i*-graphite which has been irradiated at low temperature and which might reach >50K above that temperature in processing or storage. Thought has been given to potential very slow rates of release at temperatures well below the measurable onset of energy release in differential scanning calorimetry experiments. There is confidence that no significant releases occurred at ambient temperature in the Windscale Piles between the late 1950s and the late 1990s when the material was again sampled. However, on the timescale of a GDF, with potential heating from adjacent radioactive decays and thermal blanketing from grout of concrete, the question has been asked about the consequences of very slow temperature rises (over millennia) leading to eventual triggering of energy release. This has been considered [54-55] and the risk, even for low-temperature-irradiated high-stored-energy material, is thought to be low but cannot, at present, be fully discounted. Therefore, this needs to be thought about when planning the methodology of containment and package stacking in a GDF and is one minor argument in favour of either annealing graphite components from former low temperature facilities under controlled conditions before storage or, indeed, disposing of the *i*-graphite in some other process rather than to disposal in GDF. This issue may need further consideration if a national programme were to consider the co-disposal of irradiated graphite with high heat generating wastes such as spent fuel.

The GRAPA members and their predecessors have considered the risk posed by graphite dusts which contain any Wigner energy in terms of any potential dust explosion. Detailed work on unirradiated material that has been published earlier [27-28] regarding analysis for material containing significant Wigner energy [29], has demonstrated that the presence of the stored energy does not increase the explosibility hazard which is in any case extremely low and can be mitigated by appropriate control of ambient conditions.

2.8. MECHANICAL CHARACTERIZATION

Summarized from Annex I, Section 7.

Società Gestione Impianti Nucleari (SoGIN) has conducted an extremely thorough survey of the mechanical, physical and radiological properties of the PGA moderator graphite in the Latina reactor (Italy) in support of the proposal to use mechanical recovery of intact graphite blocks in the dismantling procedure. Latina, along with Tokai 1 in Japan, are different from the UK Magnox reactors in that the columns of blocks are ‘staggered’ to provide additional stability

in these seismically active areas. A comprehensive report in Annex I includes a review of the oxidation characteristics of the graphite and builds upon analyses of trepanned samples from Latina first conducted at the UK laboratory at Berkeley in Gloucestershire (CEGB, later Nuclear Electric) [51] and draws upon the extensive body of UK monitoring of Magnox-reactor graphite.

This impressive body of work is used to underwrite a detailed programme for design of suitable handling equipment, which is discussed in Section 3.4.

A much smaller but equally important analysis has been undertaken for the AGOT graphite in the Politecnico Milano L-54M reactor.

2.9. BULK CHARACTERIZATION OF REACTOR GRAPHITE

Summarized from Annex I Section 8.

This work is intended to support routine classification of individual *i*-graphite components as they are removed from the reactor, identifying items which are either abnormally high in radioactivity or those which might be handled as a lower level of waste. The principal motivation for this work has come from INPP, seeking to introduce an 'on-line' monitoring process whereby data on strong γ-emitters might be utilized to make judgements about the concentration of other isotopes based on concentration or activity ratios previously derived from modelling work or from limited measurements. The applicability of such a method, based upon the observed high variability of the presence of some isotopes within the graphite from a single location, as well as the inability of modelling to account for mobility of materials which can be trapped in the graphite and subsequently activated is of concern. However, the economic advantages and process utility of such a method are very clear provided that the regulators and radioactive waste management authorities can be satisfied upon the quality of the data.

Work has been undertaken by Sellafield Ltd (UK) on fuel-sleeve graphite from the UK AGRs which is currently stored at Sellafield awaiting a decision on final disposal. CPST has focused strongly on the determination of ^{14}C in the Ignalina graphite and has developed and tested innovative equipment for its rapid determination, shown in Fig. 4.

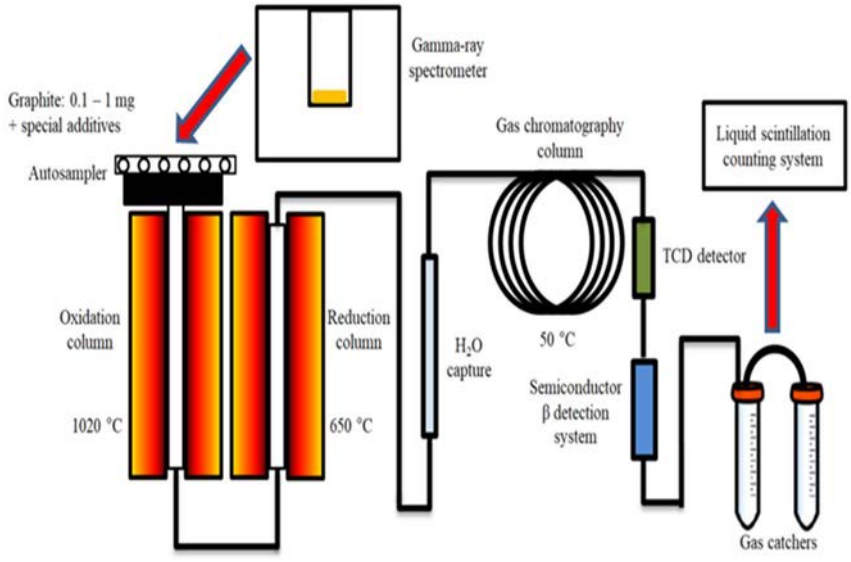

FIG. 4. Rapid system for ^{14}C specific activity determination in the sample. (Courtesy of CPST).

This analysis is based on graphite sample combustion (>1 μg) and measuring the total CO_2 mass released with an elemental analyser and ^{14}C specific activity determination with a semiconductor detector system. These analyses can be performed using a liquid scintillator or a semiconductor system, depending on the target size.

The accuracy of the semiconductor detectors for very small mass samples with low ^{14}C activity was cross-checked using LSC (Liquid Scintillation Counting) measurements and gas catchers (3M NaOH). The linear approximation function obtained from the LSC method and semiconductor detectors could serve as the ^{14}C activity calibration curve for the rapid 14C activity determination in routine measurements. The detection process for samples containing a ^{14}C activity higher than 19 Bq, was taking approximately 10 minutes. However, the estimated efficiency of the semiconductor detectors system is poor (15%), due to the 2π geometry resulting in an uncertainty that is within an acceptable range (10–20%) for radioactive waste characterization purposes.

This fast online radiological characterization has potential for analysis during the dismantling of reactor graphite or for the sorting of graphite waste. The results could be validated using the LSC method as a control measurement (for example, every 20th sample). This method could be used with the aid of a scaling factor for biomedical applications when dealing with the specific activity determination of ^{14}C in a sample.

Attention should be drawn here to an independent study (outside the GRAPA project) on determination of ^{14}C and ^{3}H based upon studies of graphite from the Kurchatov Institute research reactor 'RFT' and on samples from St. Petersburg NPP [56].

2.10. SUMMARY

Characterization has been the largest activity within GRAPA, illustrating the importance which participating organizations attach to the data-gathering processes for *i*-graphite. The network members have discussed the data the general philosophy surrounding characterization, and made the following deductions:

— There has been a lack of foresight during graphite sampling as part of operational procedures but missed opportunities to collect radiological data appropriate to decommissioning and waste management: this has led either to a dearth of necessary information or to expensive and potentially difficult post-decommissioning sampling requirements;

— Sampling requirements need to be carefully managed to minimize costs whilst ensuring that the degree of sampling is statistically adequate to accommodate spatial variations in properties within the *i*-graphite: this has been carefully considered by EDF in the context of UNGG radioisotope characterization [57] using a methodology related to the sampling of heterogeneous and dynamic materials [58]. INPP also paid attention to the operational history in selecting specific locations to take *i*-graphite samples in adequate quantity and quality. Pacific Northwest National Laboratory (PNNL) work relating to sampling similar reactor types for other purposes [59];

— Data acquisition should be matched to, and limited to, information which is clearly needed to define the dismantling and disposal strategy being followed: as an example, if a decision has been to process a particular source of *i*-graphite and it is needed in powder or granular form, it may not be necessary to evaluate the strength of whole components if they are not

to be recovered in this form. Where WAC for the final destiny of the i-graphite are known, this could influence the characterization programme;

— No two sources of *i*-graphite have similar characteristics – not even two nominally identical reactors on the same site, since operational differences can result in significant divergence in certain properties: however, where use can be made of information from other sources, this may save time and cost;

— Prior modelling of radioactive inventories is valuable but, where sample measurements provide results which deviate from modelling predictions, it is the actual data that matter, rather than expending time and effort on changing the modelling which in any event cannot account for adventitious transport of material to the graphite which subsequently becomes activated;

— Ensure that the data are both correct and relevant: an example of the former issue has been the significant downward revision of earlier measured ^{36}Cl inventories, making it a far less serious issue than ^{14}C in terms of eventual potential release from a disposal facility; an example for the relevance issue is to ensure that Wigner energy is assessed in terms of the rate of release per unit temperature rise (which determines the risk) and not as total stored energy which does not, in itself, determine the potential for energy release during processing or storage;

— Seek to avoid delaying decisions on progressing dismantling by requiring more characterization.

While uncertainties remain in WAC, the provision of ultimate disposal facilities and in preferred engineering solutions for dismantling, the relevant specialists seek to engage more directly with the authorities to ensure that continued work adds value and allows movement towards final decisions on the *i*-graphite management philosophy (including interim storage where necessary).

2.11. AGEING MANAGEMENT AND INSPECTION

Storage is temporary and implies that waste packages will eventually be removed from the storage facility, either for further treatment or conditioning or for emplacement in a permanent repository. As such, the storage facility needs to be designed to ensure that the environment promotes package integrity, which may include temperature and humidity control, prevention of salt ingress, deposition, package emplacement and movement procedures which avoid damaging the packages. Additionally, a programme of inspection, maintenance and ageing management is required to ensure that the waste packages and storage facility remains intact for the intended storage period. While the focus of inspection and maintenance is on active components, ageing management focus is on the understanding of stressors that affect the ageing processes, especially of passive components of the storage structure or waste packages. Apart from a robust design of the facility, this will typically involve active maintenance of auxiliary systems, such as lighting, ventilation, security systems, fire detection and suppression systems, radiation monitoring equipment and waste package handling equipment (cranes and/or forklifts). Routine inspections and maintenance of the storage structure is advisable and includes periodic assessments of the condition of the facility itself (inspection for signs of deterioration of structural concrete and steel and filter and gasket replacement). It may require periodic re-assessment of the safety case and/or licence conditions as well as periodic renewal of the operating licence.

3. GRAPHITE RETRIEVAL

The objective of radioactive waste management is the disposal of conditioned radioactive waste and interim storage of waste/waste packages is a key component in achieving this goal. This section contains summaries of important formation needed before selecting or designing storage systems.

3.1. INTRODUCTION

The 'Retrieval' work package within GRAPA has been coordinated by Mr Jon Goodwin of Cyclife, an owned subsidiary of EDF, and Mr Gianluigi Migliore of SoGIN.

Three significant pieces of work have been undertaken in the context of graphite retrieval. The purpose of this part of the GRAPA project was to explore alternatives to the perhaps 'obvious' dismantling philosophy of removing whole blocks piecemeal, while at the same time encouraging appropriate design and development work for such equipment where that is the State policy.

3.2. SIZE REDUCTION OF GRAPHITE BLOCKS IN CORE BEFORE RETRIEVAL

Summarized from Annex II, Section 1.

Nibble and vacuum has been developed by the UK Innovate team (Costain, Tectronics and the University of Manchester) [25] as part of a cradle-to-grave graphite management approach being developed under the 'Thermal Treatment of Irradiated Graphite' project. The work reported here covers the trials undertaken by the Innovate UK team on both the small scale and industrial scale, aimed at developing and underpinning the design considerations for a 'nibble and vacuum' retrieval process for application to graphite moderator blocks to provide a particulate graphite feedstock suitable for downstream gasification approach being developed by Tetronics.

The areas which have been investigated within GRAPA are:

— Particulate size distribution (PSD) of generated particulate ;

— PSD envelope development and matching (Nibble and Vacuum PSD vs thermal treatment requirements);

— Design considerations for optimization of tooling options for pilot scale development.

The nibbling tool with partially removed graphite from graphite blocks can be seen in Fig. 5.

FIG. 5. Nibbling tool and partially removed graphite blocks in an experimental array. (Courtesy of Costain).

scale size reduction trials were undertaken at a UK company facility (Gnat UK) specializing in hydro-demolition and robotic demolition, in 2016. Separate size-reduction trials were undertaken, the key purpose of which were to define the key process parameters (Rotation speed, Pick Number/Spacing, Block Orientation) required to generate a particulate feedstock which is targeted as meeting the requirements for Tetronics thermal gasification process. The trials generated ~30kg of particulate graphite feedstock generated for use by Tetronics in their thermal treatment trials. Comprehensive analysis of the generated particulate has been undertaken, which specific focus on PSD, the particulates aspects ratio and tooling configuration and process speed. The principal conclusions of the test were as follows:

— The initial conservative pilot scale process rate is between 1-4 tonnes per day; however, increasing the tooling size/specification, variation in pick choice and expanded shift allocation will result in a significant increase in envisaged process rate;

— Change in pick type will result in a marked reduction in the dust-sized fraction, thereby increasing particulate proportion with target envelope;

— Increase in platform specification will ensure that the process of size reduction is consistent, thus providing greater control over the process;

— Process output envelope and thermal treatment feedstock envelope have been demonstrated to be perfectly aligned;

— In summary, initial trials have demonstrated ability of the process to achieve the required results; however, this can be further optimized.

Subsequent work as taken place on industrial grinding of nuclear graphite to provide particle sizes suitable as a feedstock for a subsequent heat-treatment process which has been separately investigated on the laboratory scale at the University of Manchester with both virgin and irradiated graphite and on the industrial scale with virgin graphite by Tetronics. The oxidation tests are outlined in Section 4.2. Optimizing the particle-size requirements together with the capabilities of the nibble and vacuum approach leads to two options for optimizing the graphite retrieval:

— In-situ size reduction (i.e., in-reactor size reduction with vacuum retrieval);

— Ex-situ size reduction (i.e., retrieval of graphite as whole intact blocks, or as large pieces, with the application of subsequent size reduction to generate the feedstock for the thermal treatment process).

3.3. DISMANTLING GRAPHITE CORE BY WHOLE BLOCK RETRIEVAL.

Summarized from Annex II, Section 2.

The starting point for examining whole-block removal/retrieval has been the successful dismantling activities at Fort St. Vrain [60] and the Windscale Prototype AGR. For shielding reasons, the first of these was carried out under water and the second in air, making it the benchmark for subsequent 'in-air' operations albeit handling graphite which had experienced a much lower total irradiation than a commercial power reactor or a production reactor. In this latter example, a three-headed drill was used to gain traction on the blocks, which had not experienced significant distortion or degradation in this small core. Sellafield Ltd have reported that designs for dealing with Windscale Pile blocks involved a three-headed expanding lifting device were developed in the late 1990s when significant characterization work was undertaken (involving some members in the current GRAPA project), but dismantling has been deferred and there is no active programme at present.

3.3.1. Dismantling of graphite pile of Latina NPP

Work in this area has been conducted by SoGIN [61] in support of Latina NPP, by INPP, by Rosatom in support of dismantling production reactors and by EDF in the development of the industrial demonstrator at Chinon. Awareness of the consequences of irradiation damage to the crystallites and the consequences for changes in mechanical properties, together with the significant anisotropy arising from the manufacturing process, has meant that careful attention has been paid to the interactions between mechanical handling devices and the individual components. Appropriately, in these cases, careful analysis has been employed to establish both the loadings which the components can safely tolerate, and the potential forces necessarily transferred through the equipment to facilitate component removal. The comprehensive SoGIN study has indicated the need for further work to refine these parameters, which is ongoing.

3.3.2. Dismantling of a RBMK-1500 reactor

INPP has given careful thought to the removal of both intact stack blocks and other graphite components, like sleeves and rings from the fuel-tube channels [62]. A comprehensive design and development programme has been followed, with due regard for the environmental impact: this may include crushing and cutting the latter components, whilst a mock-up assembly for handling intact blocks has been constructed and tested, with due regard for the necessary interim storage facilities and for the requirements of VATESI (Lithuanian Nuclear Regulator).

3.3.3. Dismantling of UGR reactor core

Russian state nuclear energy corporation (Rosatom) has giving attention to the removal of intact components from several Russian developmental reactors and production reactors and has recently successfully commenced the removal of blocks from the ADE-5 reactor. Their analysis covers the development of appropriate tools for dealing with the quite highly distorted *i*-graphite components, envisaging the following operations as being required:

— Capture and vertical lifting of whole graphite blocks;

— Capture and vertical lifting of graphite block fragments;

— Drilling of graphite stacks (full or partial) and removal of generated graphite dust and fragments;

— Transfer of graphite blocks and their fragments into the area suitable for their vertical lifting.

Appropriate tooling has been developed for the penetration of the biological shielding slabs with both a rope saw drive mechanism and plasma cutting. The successful commencement of block removal from the ADE-5 production reactor has been achieved.

EDF is developing their industrial demonstrator facility adjacent to the Chinon site, the purpose of which will be to develop a dedicated mock-up facility to support the preparations for decommissioning. This programme will begin in 2022, with Graphitech (a joint entity established by EDF and Veolia) leading the development and qualification phase using full-scale models to prepare the remote-operation tools for the decommissioning of the Chinon reactor.

3.4. EXPERIENCE IN DISMANTLING THE THERMAL COLUMN OF WWR-S RESEARCH REACTOR AND GRAPHITE REFLECTOR OF CIRUS REACTER

Summarized from Annex II, Section 3.

Given the success on removing the first blocks from ADE-5, it is hoped that fuller details of the procedure and lessons learned will be available in the future under subsequent activities under a successor project to GRAPA. Graphite components have already been removed from additional small reactors, such as WWR-S in Romania

The planning and preliminary execution of the dismantling of the CIRUS research reactor in India provides important experience in this context. This contains both graphite reflector components and a thermal column and the plan is to conduct dismantling in air. The experience to date relates to the thermal column. The comprehensive report on CIRUS which has been provided [Annex II, Section 3] notes issues with unexpectedly high loads being necessary to withdraw graphite plugs (using an existing T-hole on their faces provided precisely for this purpose): these components either rest against the reflector graphite at the commencement of the thermal column or otherwise plug experimental holes. Given the operating temperature of these graphite components (reflector 100-150°C and thermal column <60°C), significant irradiation damage leading to distortions might be expected, and a force more than 250kg was required to pull one of these components free. The required forces were alleviated after ‘cleaning’ with liquid nitrogen and air jets. This experience provides a valuable insight into issues which might be encountered in dismantling plant in other Member States

Remote-handling tools are being designed to facilitate the removal of reflector graphite, as the residual dose even when the reactor vessel has been removed will be significant.

3.5. SUMMARY

Technologies for the removal of *i*-graphite components from reactors (principally moderator and reflector) will be design-specific. In some cases, such as UK Magnox, each successive pair of reactors exhibited significant design differences from the previous ones and such matters as access to the graphite can present different challenges for each site. There are notable differences in the rate of preparation for dismantling in different Member States.

Baseline planning is predicated on the removal of intact components in air, following earlier suggestions in some instances that dismantling under water would offer safety and personnel-dose advantages. However, this option introduces new waste streams and, in some cases,

through a lack of leak-tightness, was found to be impractical except in cases such as Fort St Vrain where the process required simultaneous transfer of fuel compacts and thus demanded significant shielding. Such issues have not been addressed for the developing high-temperature reactor (HTR): being of the pebble-bed variety, this issue will not arise.

Regulators and operators appear reluctant to embrace innovative graphite-retrieval solutions such as ‘nibble and vacuum’ to support the removal of intact components. It is anticipated that alternatives will receive more consideration when it is found that some treatment of graphite is necessary before eventual packaging and disposal, or if projected dates for the development and operation of disposal facilities appropriate for whole components is further delayed.

4. TREATMENT

4.1. INTRODUCTION

The Treatment work package within GRAPA has been coordinated by Professor Abbie Jones of The University of Manchester (UK), and Dr Alexander Pavliuk of PDC UGR (Russia).

This section relates to the potential for treating *i*-graphite, either to reduce its radio-isotopic content (and its waste category) or physical appearance for an alternative disposal strategy to encapsulation and deep geological disposal. Whilst most participating Member States with significant large volumes of *i*-graphite see disposal in a geological facility as their baseline solution, many Member States with smaller quantities of *i*-graphite are considering alternatives in order to enable recategorization of waste to reduce costs for disposal. For those authorities where recoverable near surface disposal options is preferred in anticipation of improved future technological solutions, the activities in this Section are particularly relevant.

It must however be considered that any treatment process may give rise to secondary waste streams and additional contaminated plant which would need to be appropriately managed in addition to the *i*-graphite and whose cost would need to be taken into consideration.

4.2. OXIDATION OF GRAPHITE

Summarized from Annex III, Section 1.

Although nuclear graphite will not burn, it may be incinerated in a fluidized-bed arrangement, especially with an enhanced oxygen concentration at the outset. Early experiments in this regard were conducted by Framatome with a successful pilot plant [62], and the general principle of conducting a process whereby ^{14}C (principally) is released into the atmosphere has been investigated and supported by two key detailed analyses which have investigated both global dispersion and the minimization of local dose effects taking diurnal influences into consideration. [63- 64].

The first of these analyses considered the complete incineration of one Magnox-reactor core per year over a twenty-year period and found that the overall atmospheric content of 14C (global dose) would be raised against the continuing background of generation from 14N by cosmic-ray bombardment by less than one part in 1 000. However, it is the local dose in the region surrounding such a plant that is of more urgent consideration, and the potential take-up of radioactivity by vegetation subsequently consumed by animals and humans. The second reference concurs with the conclusions of the first one but provides much more analysis of the local-dose effects, considering such issues as diurnal variation in throughput to match times at which plant uptake is at a minimum. The papers conclude that the process is viable although there is an obvious conflict with a desire to limit any unnecessary discharge of radioactive material to the environment. This leads both to consideration of suitably remote locations for such an incineration activity, and methods of capturing the ^{14}C (or the $^{14}CO_2$ off-gas): for suitable downstream isotopic blending either via carbon capture and storage or by controlled aerial discharge: this is something that the previously mentioned UK core-to-capture process includes, as discussed in Section 5.5

This latter process does not involve incineration, but controlled oxidation via plasma-heating. Under the GRAPA network both possibilities have been taken forward beyond bench scale demonstrations and progressed up the technology readiness levels. A new pilot incineration plant for *i*-graphite has been established in the Russia Federation (at the pilot and demonstration

centre for decommissioning of uranium-graphite nuclear reactors (PDC UGR) site, which is being used to investigate both total incineration and heat treatments to mobilize specific radioisotopes. Rosatom investigated plasma processing, but the principal work in this area has been conducted by the company Tetronics at plant scale, in association with the University of Manchester (detailed lab-scale evaluation) as part of the UK Innovate project. Overall, this study demonstrates the feasibility of a large-scale thermal treatment as a viable decommissioning route for nuclear graphite. This proposed treatment process could provide an accelerated decommissioning solution and an alternative to the baseline strategy in some national programmes.

4.3. DECONTAMINATION OF GRAPHITE

Summarized from Annex III, Section 2.

The process of thermal decontamination has been pursued with interest by CIEMAT, NNL and Rosatom because it has the capability to reduce the waste category of *i*-graphite, leading to considerable savings in disposal costs and the possibility of facilitating reuse in the nuclear industry via some subsequent processing and reformation of new carbon-based products. It is of potential use for the mobilization of several isotopes, but most research to date has focussed on addressing the important long-lived beta-emitting isotope ^{14}C. This offers the possibility to recover ^{14}C as a useful isotope, mitigating the need for separate commercial production from nitride irradiation.

The original work to investigate the potential mobility of ^{14}C in *i*-graphite was undertaken at the Forschungszentrum Jülich [65] and has subsequently been taken up by numerous organizations seeking to understand the mechanism and to improve on the efficiency of the process. ^{14}C arises from the 1.1% ^{13}C present naturally in the graphite (which may be presumed to be uniformly distributed, at least initially) and with a greater efficiency from any ^{14}N which may be present with the graphite (impurities in the pitch binder used in manufacture being the most likely source).

The developing interest in the topic led, initially, to conflicting views on which production route was dominant, losing sight of the point that the proportion of 14C from the two principal activation routes would vary according to the design of the reactor systems and to its different degree of exposure to external sources of nitrogen. An example of this may be found by comparing two different carbon-dioxide-cooled reactor systems: the French UNGG and the UK Magnox. In the former case, it has at one point been claimed that the entire ^{14}C inventory was generated from the $^{13}C(n, \gamma)^{14}C$ reaction [66], whereas UK research indicated a predominance of the $^{14}N(n, p)^{14}C$ route with additional concentration of ^{14}C being found in carbonaceous deposits on the component surfaces [26]. Idaho State University focussed on the latter reaction and, using inactive nitrogen created an interesting range of surface nitrogen-containing species on graphite surfaces with unique properties [67] suggesting that graphite could act as a ‘getter’ if exposed to coolant gas containing small impurity concentrations of nitrogen, trapping the nitrogen on the graphite surface. In air-cooled systems (production reactors) and in RBMK (helium/nitrogen blanket gas), large proportions of the gas in contact with the graphite is nitrogen and a dominance of the associated generation route for ^{14}C would be anticipated.

Separate investigations have attempted to understand potential mobility of ^{14}C atoms within the graphite while it is still under irradiation. A remarkable property of nuclear graphite is that fast neutron damage over the lifetime of a reactor can cause every carbon atom to be displaced numerous times while yet retaining the physical characteristics of the graphite crystallites. This is expected to lead to a homogenization of the ^{14}C, ^{13}C and ^{12}C isotopic content of the irradiated

material (subject, of course, to new ^{14}C being created at surfaces from external sources of 14N). In addition, the potential for displacement of ^{14}C atoms, through the recoil energies introduced by the slow neutron collisions, leads to the expectation that a significant proportion will move away from the initial lattice position of the ^{12}C or ^{13}C atoms from which they were created [68-69]. It is obvious that there is a great deal about these mechanisms which remains to be clarified.

Most recently, attention has shifted towards improving the efficiency of the mobilization by investigating the optimum conditions (cover gas oxygen content and temperature) to maximise the $^{14}C/^{12}C$ ratio in the released material and obtain a solid *i*-graphite residue suitably depleted in ^{14}C for subsequent disposal, while optimizing the economics of the process. In some cases, a fully inert gas flow (argon) has been utilized, indicating that the release of ^{14}C can occur without direct oxidation. Such work has recently been conducted by Rosatom and the isotopic analysis of the treated graphite is now in progress.

The UK National Nuclear Laboratory has recently confirmed that, in samples from the Oldbury Magnox reactor, the ^{14}C content of carbonaceous deposits is 80 times greater than in the underlying graphite and seeks to rationalize the ^{14}C mass balance in their oxidation process. The University of Manchester has studied the process.

4.4. CHEMICAL TREATMENT OF GRAPHITE

Summarized from Annex III, Section 3.

Moving away from work specifically on ^{14}C, the University of Manchester has developed a molten-salt technology for the general decontamination of *i*-graphite from Magnox reactors using gamma spectrometry. The treatment at 450°C in a LiCl-KCl eutectic included the following procedures: electrochemical cleaning of the salt, initial cyclic voltammetry (CV) of graphite followed by several steps of chronopotentiometry (CP) with a range of currents applied. The release of corrosion and fission products in molten salt media from the irradiated graphite due to electrolysis in a molten salt system was investigated to explore whether this process could be applied to the decontamination of further irradiated graphite and to understand the influence of current value on radioisotope transfer into the salt phase. Results showed that the molten salt treatment can successfully remove ^{60}Co, ^{137}Cs, ^{133}Ba and ^{154}Eu from irradiated PGA graphite and follow-on work continues in these areas as well as consideration of how the process might be economically industrialized for eliminating HLW and ILW-categorized *i*-graphite.

Russian colleagues have successfully achieved electrochemical decontamination using the graphite as electrodes in aqueous solutions of mineral acids, oxidizing salts (potassium permanganate, $KMnO_4$) and hydrogen peroxide: this work is published in Ref. [70].

Attention of the GRAPA members has been drawn to work conducted by National Agency for New Technologies (ENEA (Italy)) under the CAST framework to remove 14C from *i*-graphite by exfoliation in organic solvents. However, the GRAPA members has also noted the conclusion from CAST that, in general, the bulk of the ^{14}C in *i*-graphite is non-releasable.

4.5. NOVEL POTENTIAL TREATMENTS

Summarized from Annex III, Section 4.

Several novel treatment ideas have been briefly considered by the GRAPA members as having potential for further investigation if economically viable. These include microwave heating

(both in-reactor or as a production line on removed material), as proposed by Imperial College Department of Chemical Engineering (UK) but not yet taken up within GRAPA, the use of recovered ^{14}C to produce nuclear batteries (synthetic diamond devices) proposed by The University of Bristol (UK), and decontamination of *i*-graphite using supercritical fluids: however, these activities have not been reported to GRAPA during this programme.

In this last option, work is in progress at the China Institute of Atomic Energy in Beijing using supercritical water, with initial results expected shortly, and work is anticipated at FH Aachen (University of Applied Sciences), Germany, in association with Dustec High-Pressure Technology using supercritical carbon dioxide. In the latter case, this is explicitly related to ^{14}C reduction to assist the disposal of German wastes to the KONRAD facility.

4.6. IMMOBILIZATION OF GRAPPHITE BY HOT PRESSING TECHNOLOGY

Summarized from Annex III, Section 5.

One suggestion for synthesis of a suitably stable compound is to use the technology of hot isostatic pressing in an inert gas (argon). This technology can produce compounds which provide suitable high rheological characteristics including leaching resistance. The PDC UGR (Seversk) and. Frumkin Institute of Physical Chemistry and Electrochemistry (IPCE) (Moscow) teams have finished laboratory scale hot isostatic pressing experiments and are moving forward with pilot-scale hot isostatic pressing experiments.

Separately, Furnaces Nuclear Applications Grenoble (FNAG)(Hanau, Germany) participated during the first year of the GRAPA project to introduce their investigations of glass-impregnated graphite. The encapsulation of different potential waste stream like spent fuel, inorganic ion exchangers or volatile nuclides like ^{137}Cs has been successfully demonstrated by with inactive analogues.

Four promising long-term resistant glass types have been identified for further investigations under conditions more closely representing potential disposal sites.

However, the final improvements must be performed with radioactive samples. Therefore, it is intended to install a hot vacuum pressing facility (HVP) in an isotope lab. This will allow the manufacturing of small samples with irradiated graphite and other radionuclides as tracers. The HVP is already installed at FNAG Grenoble for inactive commissioning. The commissioning in an isotope lab has been proposed as a collaborative project of ENRESA, CIEMAT and FNAG.

4.7. MORTAR AND CONCRETE

Following previous successful incorporation of crushed graphite from Swiss reactor facilities into mortar (Paul Scherrer Institute), the Spanish authorities CIEMAT and ENRESA indicate that research into the possibility of utilizing similar mortar composition at the El Cabril disposal facility for the immobilizing of crushed graphite from reactor core. No additional information form network members were available on this aspect,

4.8. SUMMARY

The focus on *i*-graphite treatment process options during GRAPA has been upon two fronts. The first is thermal treatments, inclusive of both direct oxidation of the graphite and mobilizing

isotopes (specifically ^{14}C preferentially to ^{12}C and ^{13}C) to reduce the waste category of the residual material. There are sufficient inconsistencies in the reported data historically to enable the conclusion that the mobilization process is still not yet fully understood and, if such a process is to be committed to commercially, there first needs to be a very carefully focused and hopefully collaborative programme to optimize the appropriate conditions to maximize efficiency and to produce the required cost savings in subsequent disposal.

The second area is in the use of glasses and mortars, dispersing the graphite as powder in each case and using the product (glass or mortar containing graphite) as an immobilizing medium for other wastes whilst retaining the radioactivity from the graphite itself. The viability of such mortars was established by colleagues from the Paul Scherrer Institute in Switzerland in the preceding CRP [15] and early in the GRAPA network, while the development of the glass matrix continues as a commercial development.

A separate initiative relates to molten-salt treatments to remove isotopes, a process whose development is continuing at the University of Manchester (patent pending) and the use of liquid-chemical methods for contaminated graphite by Rosatom [71-72].

It is felt that these various alternative procedures, while scientifically proven, need to be demonstrated to be commercially viable and to demonstrate clear cost savings (and personnel-dose savings) compared with cementation and disposal to a GDF before onward consideration in national programmes could be considered.

5. PACKAGING, STORAGE AND DISPOSAL

The Packaging, Storage and Disposal work package within GRAPA has been coordinated by Dr Simon Norris of Radioactive Waste Management Ltd, UK.

Several Member States have adopted packaging in drums, grouting and ultimate geological disposal as the intended plan for managing *i*-graphite. In most cases, there is a need for interim storage prior to disposal in geological disposal facilities, recognizing the timescale typically needed to site and develop such infrastructure. Whilst GRAPA has considered a range of methodologies that could potentially be deployed in the management of irradiated graphite in national programmes aside from disposal, it has been important to cover disposal-related topics and experiences, not least because of the imminent needs of INPP. The Russian Federation elected to utilize so-called in-situ disposal for one of its production reactors, whilst the UK Innovate team has considered a 'core-to-capture' treatment philosophy from the viewpoint of minimizing the overall radiation dose exposure from the *i*-graphite and its products.

5.1. PACKAGING AND ON-SITE STORAGE

Summarized from Annex IV, Section 1.

INPP has presented to GRAPA their entire developing strategy for all *i*-graphite materials for discussion and potential support. The first consideration has been the smaller graphite items – sleeves, rings and the protection and control-rod materials – with an environmental impact assessment to facilitate the design and development of the management process. The planning has proceeded with characterization issues, design of facilities for crushing the *i*-graphite, storage in steel 200 litre drums, dosimetry, measurements and packaging modes for the interim storage. A similar planning process has been adopted for retrieval and management of the *i*-graphite items (blocks and rods), including consideration of the possibility of encountering cracked and damaged components of components.

5.2. IN-SITU DISPOSAL

Summarized from Annex IV, Section 2.

The USA first proposed this concept [73-74] in connection with reactors at Savannah River and Hanford and entombs the reactor where it stands with appropriate preparation to immobilize potential radioactivity – thus creating individual 'near-surface' repositories. In the current GRAPA programme, this approach has been applied to the EI-2 reactor in the Russian Federation following work at PDC UGR to establish appropriate conditions for infilling the residual internal regions of the reactor with clay-based material to contain radioactivity after the removal of external equipment. An additional safety barrier is constructed over the top of the reactor – in essence, an artificial hill (Fig. 6 shows the stages of preparation). EI-2, whose 'on-site disposal' was completed in September 2015, is seen as the pilot plant following which a similar method of isolation will be considered for other production reactors.

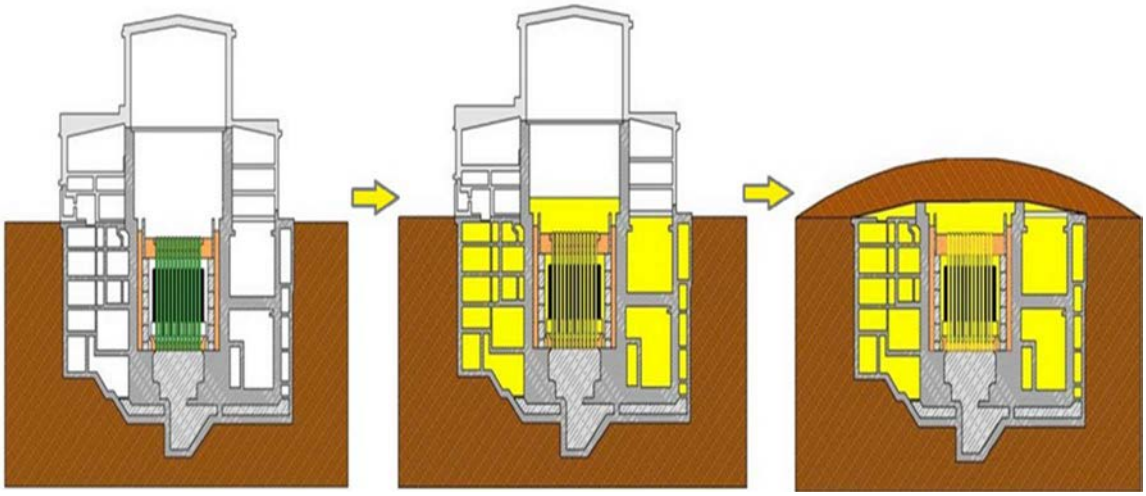

FIG. 6. Disposal stages of the EI-2 reactor. Yellow colour indicates stabilising clay-based material, brown colour represents natural (ground) material. (Courtesy of Rosatom, Russian Federation).

5.3. DISPOSAL

Summarized from Annex IV, Section 3.

RWM (UK) have considered the disposal of *i*-graphite from UK reactors in detail, following separate pathways for England and Wales (GDF, at a depth >200m) and Scotland (near surface disposal close to originating sites), with regard for the potential release of the weak beta-emitters (principally ^{14}C). Although these are the base cases, careful note is being taken of developments in potential treatments as other options for disposal remain available.

The GRAPA members discussed in detail the planning for the management of the much smaller amounts of graphite from the WWR-S reactor in Romania ahead of the availability of a suitable general radioactive waste disposal facility (estimated for 2055) as this provides a useful base case for other small research reactor facilities.

5.4. POTENTIAL REDUCTION OF ^{14}C

Summarized from Annex IV, Section 4.

The UK Innovate team, while noting the formal UK position on *i*-graphite disposal, has noted that (for England and Wales) it is built around the construction of a GDF for which there is currently no identified site and therefore no realistic timescale, that such a construction and disposal route is extremely expensive and carbon intensive and, most particularly, that the principal long-lived isotope ^{14}C is potentially mobile in the very long term in such a facility and potentially capable of re-concentration in various chemical forms. The team has therefore taken a holistic view of the problem, seeking to identify an alternative process which can deal with these issues in a convincing manner, starting with retrieval of the *i*-graphite, processing, and disposal of the carbon (as sequestered carbon dioxide) whilst placing the ^{14}C in an environment where it cannot present any future hazard.

This final aim would be achieved by blending carbon dioxide derived from *i*-graphite (which is rich in ^{14}C) with carbon dioxide derived from fossil fuels (which is lean in ^{14}C and is a by-product of other industrial processes). Such blending is different from environmental dilution, where natural processes can lead to re-concentration as shown in Fig. 7

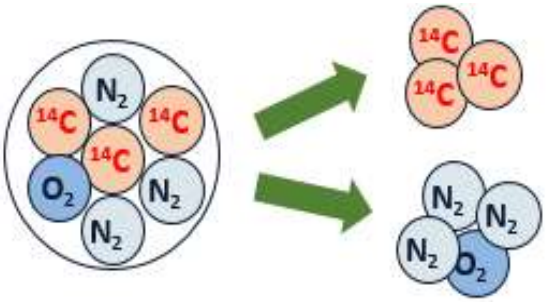

FIG.7. Environmental dilution (discharge to air, either directly or indirectly) leaves the possibility of ultimate 're-concentration' by natural processes. (Courtesy of Costain).

Potential reduction, on the other hand, produces a mixture of ^{14}C and natural carbon. Because these are chemically so similar, there is no natural mechanism for re-concentration as shown in Fig. 8.

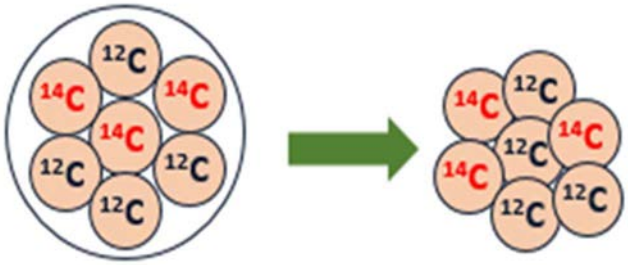

FIG.8. Blending of ^{14}C with natural carbon (^{12}C, ^{13}C in the same chemical form) removes the possibility of future-concentration of the ^{14}C. (Courtesy of Costain).

The process is completed by confining and isolating from the environment the blended carbon dioxide in a carbon sequestration scheme. This is similar in principle to the geological disposal facility concept, but the likelihood and impact of ^{14}C escape is much reduce.

The overall core-to-capture principle is shown in Fig. 9/.

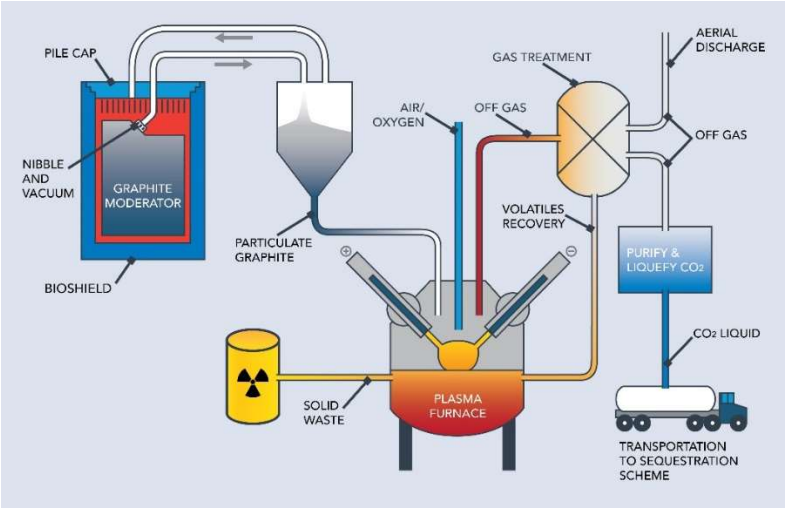

FIG.9. Holistic 'core-to-capture' i-graphite-management scheme. (Courtesy of Costain).

The approach relies on the deployment of Carbon Capture and Storage (CCS) technology to meet its climate change targets the UK power sector needs to be decarbonized by the 2030s. At the same time electricity demands will increase with the increase in electric vehicles. The International Association for the Evaluation of Educational Achievement (IEA), EU and UK

Committee on Climate Change all forecast that the cost of achieving this will be higher if it done without using CCS. CCS technology might be envisaged as utilising depleted oil and gas fields, even to the extent of using existing gas-extraction pipework in reverse: this is a possibility in which the Scottish government has already shown interest. GRAPA members noted that the IAEA has engaged on a study of the economics and practicalities of the geological disposal of CO_2 in this context [75].

Within GRAPA, an engineering design study has been performed to show that it is practical to carry out the gas treatment steps in the process. The gas treatment process is made up of two elements: in the first, non-gaseous radionuclides are removed from the gas stream and in the second carbon dioxide is separated from the gas stream, purified and compressed for storage.

The unit operations are as follows:

— Quench and scrub – where furnace off-gases complete combustion, are cooled by water injection and volatile metals (for example traces of caesium) are removed from the stream;

— Precipitation – where traces of liquid water are removed using an electric charge;

— HEPA filtration – where remaining fine solids are removed.

After HEPA filtration, most of the radioactive gas is removed and the conventional gas-processing-unit operations follow:

— Amine separation – where CO_2 is dissolved in a liquid, allowing air to pass out of the system. The CO_2 is then driven off the liquid giving a pure CO_2 stream. Amine separation is the best available technology for this step, but the project team is aware that technology for carbon dioxide separation is developing rapidly;

— Dehydration – where water is removed using a molecular sieve process;

— Compression – where CO_2 gas is pressurized to make it suitable for injection into a CCS scheme.

The design study [76] has been performed principally using Aspen HYSYS (Process Simulation Software) which is a process simulation tool. The conclusion of the study is that it is practical to engineer the system. Some of the equipment is very small in gas processing terms (for example contact columns would be made from metal pipe rather than being fabricated as vessels), but the operating parameters are within the applicable ranges for the proposed processes and no technical obstacles are foreseen to implementation.

5.5. SUMMARY

Many national waste management organizations are developing or refining existing strategic approaches to the management – including packaging, storage and disposal of irradiated graphite, which, for some national programmes, presents a waste that is significant both in size (volume and mass) and radionuclide inventory (this is particularly the case for 14C and 36Cl, both long-lived radionuclides and shown typically to be prominent concerns in precedent safety cases). Such activities are proceeding outside of GRAPA, and GRAPA has proven to be a useful international forum for knowledge exchange between national programmes, the supply chain, national laboratories and academia.

The approaches taken by respective national programmes to the management of irradiated graphite can vary, influenced by, for example, regulatory criteria and chosen strategy. Hence,

some national programmes have a baseline that assumes irradiated graphite will be removed from reactor, suitably packaged and encapsulated, then disposed to a facility (geological disposal facility, which may be currently available or may be in planning), whereas other national programmes assume waste processing and treatment following removal from reactor will be a necessary or desirable part of the irradiated graphite management route.

There is clearly more than one potentially viable approach to the packaging, storage and disposal of irradiated graphite, and GRAPA has been successful in ensuring such a range is discussed. This can have the beneficial effect of introducing alternative perspectives, new learning being brought to the attention of both national programmes that are participating in this IAEA initiative and those with interests in its deliberations and may help to guide forward thinking on irradiated graphite management strategy based on current expert knowledge and experience.

Interim storage is controversial but is forced upon some operators to make progress with other aspects of dismantling a plant. Thus, the graphite from the Windscale AGR now resides in steel containers in a new building across the road from the original reactor, and one may ask whether this is a safer storage environment than leaving it in the reactor vessel would have been. A question which has previously been raised, but not explored under GRAPA, relates to potential electrochemical action if water gets into steel storage boxes containing graphite, leading to corrosion of the steel and subsequent potential release of radioactivity.

Equally, the decision to leave the reflector graphite from the German AVR inside its reactor vessel but to fill it with concrete, upend it, and place it in a new building raises a number of questions: not only the same ones regarding overall safety and water ingress (it is understood that releases of gaseous radioactivity have been detected) but also of the increased difficulties now to be encountered when the graphite is eventually recovered for a more permanent disposal, such as to KONRAD.

6. CONCLUSIONS

6.1. CHARACTERIZATION

6.1.1. Location of radioisotopes

Earlier work conducted by the CRP [15] on the topics subsequently will not be repeated here, but the most significant content may be briefly summarized as follows:

Under 'Characterization', Section 4 of Ref. [13], the most important discussion relates to the creation and location of radioisotopes in different reactor situations. In the latter context, there is uncertainty regarding the potential mobility of newly-created radioactive atoms formed during recoil reactions (by the departing gamma ray in an n, γ reaction, for example) compared to their bond energies within the graphite structure. This remains relevant particularly to the creation of 14C and 36Cl, for both of which some conflicting observations persist. A related issue which remains open following the GRAPA project is the reconciliation of the predicted and/or observed mobility of isotopes within graphite (and thus their propensity for release) in relation to the known displacement of lattice carbon atoms by continuing fast-neutron irradiation. For instance, irradiation damage is described in terms of displacements per [carbon] atom (which are numerous in the lifetime of a reactor) without clearly explaining either how the graphite retains its fundamental crystal structure and geometrical shape despite significant damage and local re-arrangement of bonds and the creation of vacancies, or how these impacts upon the mobility of isotopes within that structure.

At the end of Section 2, the importance of matching the extent and relevance of characterization data to the intended disposal route for *i*-graphite was emphasized, along with the need to have specialists engage with regulators and radioactive waste management authorities to ensure that, where further data are requested, they are relevant and, where differences in methodologies or interpretations have existed, that internationally agreed standard techniques are employed to allow easy comparison of results. An important example of this relates to the potential leaching of radioisotopes either from the *i*-graphite directly or from grouted or package material. The CAST project (deliverable 5.4) also reviewed methodologies in the interest of harmonizing the acquisition and presentation of leaching data [77].

6.1.2. Leaching studies

For leaching studies in general, two distinct issues need to be borne in:

— Laboratory-based leaching experiments can only provide short-term data (up to three years seems to be typical maximum duration of tests). Such tests have shown significant variations in leaching rates over these timescales – often initially high rates tapering off – which are of value if they suggest that deliberate washing can reduce the content of some isotopes significantly and usefully, or that specific actions need to be taken during such operations as wet grouting to manage potential activity releases. It may be concluded that such tests are relevant only in the so-called operational phase, when the *i*-graphite is being handled and when it is being placed in a storage facility. In these situations, experience has shown that certain isotopes such as ^{3}H, ^{14}C and ^{36}Cl require particular attention in terms of managing operational dose;

— Long-term release rates – on the timescale typically considered in the post-closure safety case for a geological disposal facility (where time periods to 1 million years post GDF

closure may be noted) – cannot be determined experimentally. Projecting far into the future, it appears logical to assume 100% release at some point in (distant) time if the disposal facility containment is breached. The rate-determining steps regarding human and wildlife exposure depends on the transfer rates through the geosphere and biosphere. It is important to review the potential migration of radioactive atoms (particularly long–lived β–emitters) through the geosphere and biosphere. This relates particularly to the chemical form of the active species and signals an extremely important part of the characterization of *i*-graphite since it is important that migration data relevant to the correct chemical form are employed in assessments of the potential movement of the material away from a breached GDF. This is relevant for all radioisotopes, but a particular example about ^{36}Cl migration from a hypothetical disposal facility highlights the importance of understanding the chemical form as well as geosphere and biosphere conditions used in the contaminant transport assessment. Sheppard et al. in Ref. [78] modelled the migration of ^{36}Cl from a spent fuel disposal facility through the geosphere and the biosphere and others have based calculations on transfer factors such as those indicated in the work. They consider that the isotope moves through the geosphere very effectively – at the same speed as the water in which it is presumed to be dispersed. This assumption was based upon a specific disposal facility for Canadian Deuterium/Uranium Reactor Design (CANDU) fuel elements – in which the ^{36}Cl is in the form of chloride ions (Cl-) rather than in the organic chemical form dominant in *i*-graphite - and in which dispersion of isotopes assumes advection within freely-moving water in rock fissures. This would not apply in the case of clay, as an example, in which the transport rate would be much lower. More recently, alternative biosphere transport models (^{36}Cl models), based on empirical transfer factors (IMARC, ERB2A, Aquabios), or on defined specific activities (AquaCl36, SA_36Cl), or on a combination of these methods (MTA_Cl36) has been used for modelling [79]. Results indicated that the long-lived activation product ^{36}Cl will be among the more significant contributors to dose following release to the biosphere from deep or near-surface repositories for radioactive wastes.

Returning to general principles of characterization, the GRAPA members considers that an optimal methodology of radiological characterization of irradiated graphite from small research reactors and big power reactors without substantial spillages of the nuclear fuel has been developed. It combines 3D modelling of graphite activation in a nuclear reactor with experimental analysis of small samples necessary to calibrate the theoretical model.

However, taken in consideration with the investigation of treatment options, it seems apparent that some re-balancing of effort is desirable in the future. A lot of past effort has been devoted to debating the source of some isotopes (especially ^{14}C) whereas the focus needs to be on how they are bonded into the *i*-graphite and the potential for their release, along with understanding the chemical form of such releases (organic or inorganic in the cases of ^{14}C and ^{36}Cl) and how these releases might subsequently behave in the environment. Results, particularly for ^{14}C release, need to be rationalized: various current investigations have reported considerable success in releasing ^{14}C relative to ^{12}C whereas CAST has concluded that the majority of the ^{14}C is non-releasable. In general terms, future characterization programmes need to be planned and undertaken only after addressing the question: What do we need to know?

The members of the GRAPA network have identified under the sub-topic headings of the original project, the following:

— Impurity distribution: isotopic and general impurity content and distribution in German graphite to inform future disposal (KONRAD): this work is currently unresolved;

— Radiological characterization: establish the ^{14}C content and distribution in AGR graphite (NNL, UK): as these reactors approach the end of their operational lives, consideration is being given by EDF-Energy and Cyclife (owned subsidiary of EDF) to obtaining better characterization in general, although the proposed disposal strategy is not yet determined. Ongoing characterization programmes supporting the dismantling of small reactors (L-54M and WWR-S) continue with completion expected shortly;

— Leaching information: important points about this work have already been made. Various leaching studies are planned to continue, again stressing the importance of chemical form of the leached isotopes (gaseous or solution, organic or inorganic) and to establish international agreement on the relative importance of ^{14}C and ^{36}Cl in forward planning: a better understanding of the chemical forms and the mode of creation of these radioisotopes and their movement through the bulk material is desirable;

— Wigner energy: whilst the specialists can agree on the relative importance of stored energy through an understanding of its potential release rates, there seems to be much to be done in educational terms through engaging with regulators and radioactive waste management authorities to reach agreement on true risk associated with this phenomenon. Management plans for difficult sources (RBMK control-rod spacers) are being developed, and measurements on the reflectors and thermal columns from small reactors, especially where low-temperature irradiation zones exist in the graphite, are planned or in progress;

— Mechanical characterization: The only active work in this area at present relates to Latina (Magnox) and the small research reactors. INPP (RBMK-1500) cannot exclude the needs of additional mechanical characterization for next period to support the retrieval/removal/packaging activities. Other utilities whose baseline plans currently assume removal of intact graphite components will commence evaluations of their existing data and performance histories (including significant regions of weight loss through radiolytic oxidation and component cracking in the case of UKs AGRs);

— Isotope ratios and content in bulk components: although significant drawbacks in the utility of mass monitoring of activity have been identified (such as reliance on previously determined or modelled isotope ratios to determine the content of one isotope based on the results for another), such an on-stream procedure has obvious advantages provided the data are sufficiently reliable for regulators and radioactive waste management authorities. INPP is developing such a methodology, and further developments of methods for specific isotopes (CPST work on determination of ^{14}C) are expected;

— Highly contaminated graphite: this is a new topic introduced by PDC UGR (Russian Federation) to address concerns about fuel-contaminated graphite which exists in several production reactors and in the development plant (Beloyarskyia AMB-100 and 200), and to devise methods for determining the extent of the problem and for dealing with the material during dismantling and disposal. The GRAPA members note that this also has relevance for addressing Windscale Pile No 1 (UK) in which distributed fuel remains following the 1957 accident.

6.2. REMOVAL

The state of preparations for removal of *i*-graphite from reactors as intact components or in more innovative methods, ranges in different Member States to minimal current practical activity (UK, despite the historical success of dismantling the Windscale prototype AGR) through to comprehensive and detailed evaluation and planning (INPP). Previous comprehensive studies, such as those of the UK Atomic Energy Authority (UKAEA) in support of the Latina operators and others, were subsequently felt to be over-engineered and hugely expensive, which has in turn been an excuse for delay.

Process planning has, however, reached an advanced state in France for the first UNGG dismantling project and in Ukraine for the undamaged Chernobyl reactors, whilst SoGIN in Italy, following rejection of the previous UKAEA proposals by ENEA, has devoted a large effort (along with the Latina authorities) to development and design of practical dismantling equipment with due regard for minimizing costs while maintaining appropriate levels of safety. Progress is being made at INPP towards the design of equipment for dismantling the moderator stacks. The authorities responsible for small research reactors have begun dismantling such features as thermal columns using locally designed methodologies, and in Russia, where specially designed robotic equipment has successfully commenced the removal of graphite blocks from the production reactor ADE-5. All ongoing and planned graphite-retrieval operations from both test and civil nuclear reactors are to be undertaken in air.

An option of nibble and vacuum has been successfully trialled in the UK at the pilot level and shown to be capable of delivering *i*-graphite feedstock suitable for a subsequent treatment process.

Looking to the future, EDF (France) are in the process of developing an industrial demonstrator pilot facility adjacent to the Chinon site to support the tooling design, development, and qualification using full-scale mock-ups. The Industrial Demonstrator is a facility dedicated to the development of tooling and methodologies for the decommissioning of graphite reactors using realistic full-scale mock-ups. It is being built close to the Chinon nuclear site where the first-of-kind decommissioning project will take place. Trials and development work are due to commence in 2022: its main objective will be to deliver a robust and optimized scenario for the dismantling of Chinon A2 in 2028. Russian colleagues will continue work to support the removal of RMBK *i*-graphite components and for handling damaged or fractured blocks.

An area where future collaboration could prove valuable is in those areas of tooling which are common to more than one reactor system. This might, for example, cover such issues as the design of jacking systems or jaws for manipulating large graphite blocks, or even the creation of an international pilot facility where such devices could be utilized on unirradiated graphite: PDC UCR has already proposed such a cooperation based upon its own facilities.

Work on innovative techniques was planned: CPST proposes work on flotation as a means of moving graphite out of RBMK reactors: this may build upon earlier proposals from a French consultancy relating to moving crumbled or powdered graphite as a slurry.

Finally, there remains a lack of understanding of two related issues across utilities, radioactive waste management authorities and regulators which needs to be addressed in some way to avoid inappropriate decisions and unnecessary costs during *i*-graphite removal from reactors and in subsequent handling. This relates first to the misconception that graphite is a flammable material and therefore presents handling and storage issues – this has been very competently addressed by colleagues from USA national laboratories [80] following extensive work by numerous specialists. The related issue of a misplaced fear of dust explosions has been discussed within this present publication. The second issue is related to all aspects of Wigner

energy accumulation and its potential release – again discussed in detail above – and covers all aspects from choosing the correct parameter for characterization through to the currently unquantified possibility of slow-release rates over periods of geological time in the quasi-adiabatic conditions of grouted graphite in a geological disposal facility.

6.3. TREATMENT

GRAPA members have studied a range of potential predisposal techniques for instance graphite oxidation, thermal and chemical decontamination (molten salt and exfoliation), isotope mobilization and incorporation into glass matrices and mortars. The relevance of treatment as part of a waste management strategy varies between different national programmes; the issue of the creation and management of secondary waste needs to be considered. Some GRAPA members plan to continue work in the following areas:

— Optimization of oxidation parameters (i.e., incineration, plasma oxidation) and further pilot-scale developments with due regard for an overall reduction in collective dose exposure both during the process and in the long term; GRAPA members noted the intention of Korea Atomic Energy Research Institute (KAERI) in Korea to conduct similar work;

— Molten-salt electro-decontamination, and the improvement of decontamination factors, both at the University of Manchester and at PDC UGR, with both organizations also planning to investigate feasibility of scale-up demonstration and the impact of secondary waste streams;

— Supercritical CO_2 extraction techniques.

Such activities are justified against the continuing delays in delivering facilities for dealing with large quantities of intact *i*-graphite in numerous Member States.

It is important to keep in mind other forms of treatment which have been suggested or previously investigated, and whose utility may be worthy of further investigation:

— Microwave heating to mobilize isotopes;

— Electro-disintegration (at present applied only to fuel pebbles but which might be a useful precursor to facilitating incineration under less onerous conditions);

— Biological ‘digestion’ (studied by the former Pebble Bed Modular Reactor Company (PBMR Co), South Africa);

— Recycling of both carbon (to the nuclear industry) and recovery of useful isotopes: the former was shown to be feasible by graphite manufacturers participating in the former CARBOWASTE project but the current low demand for new nuclear graphite meant that the cost of setting up a production facility capable of handling active precursors was not financially viable: the recovery of ^{14}C was not financially viable either, but market conditions may change in the future.

6.4. PACKAGING, STORAGE AND DISPOSAL

As noted in 5.5, there is clearly more than one potentially viable approach to the packaging, storage and disposal of irradiated graphite. GRAPA has been successful in ensuring different approaches being discussed bringing alternative perspectives to the attention of both national

programmes that are participating in this IAEA initiative and those with interests in its deliberations.

Information exchange may help to guide forward thinking on irradiated graphite management strategy in national programmes, based on current expert knowledge and experience. Furthermore, if a national programme is actively considering evolving its strategic position on the management of its irradiated graphite or wishes to enhance the robustness of its current approach, GRAPA provides an opportunity for networking, experiential exchange and a consideration of collaborative working on issues of mutual interest (which could be beneficial to several national programmes were cost reduction and rate of work progress to be positively affected).

Maintaining an IAEA initiative on the management of irradiated graphite beyond the timescale of the GRAPA project is an effective way forward that will allow new learning on irradiated graphite management to be dispersed, ensuring knowledge, skills, best practice relating to the removal of irradiated graphite from reactor, its packaging or processing and storage, especially given the fact that relevant work undertaken in any one national programme is highly likely to be undertaken on a decadal timescale, if not longer.

The interim storage option is forced upon some operators to make progress with other aspects of decommissioning a whole facility. Thus, the graphite from the Windscale AGR now resides in steel containers in a new building across the road from the original reactor. The removal of *i*-graphite from BGRR to the Nevada desert reservations de-risked the reactor site and led to completion of long-term process of maintenance, remediation and site closure activities [23].

In some other cases, one may question whether temporary storage is a safer environment than leaving it in the reactor vessel would have been. Considering a lack of progress of *i*-graphite treatment and long-lived radioactive waste disposal construction during the last 10 years, the interim *i*-graphite storage option, as a predisposal routine operation, may provide the practical pathway prior final (deep geological or/and disposal at intermediate dept) disposal facility establishing relevant WAC for all radioactive waste generated during decommissioning. A question which has previously been raised, but not explored under GRAPA, relates to potential electrochemical action if water gets into storage packages containing graphite, leading to corrosion of the steel and subsequent potential release of radioactivity.

Equally, the decision to leave the reflector graphite from the German AVR inside its reactor vessel but to fill it with concrete, upend it, and place it in a new building raises a number of questions: not only the same ones regarding overall safety and water ingress (it is understood that releases of radioactivity have been detected) but also of the increased difficulties now to be encountered when the graphite is eventually recovered for a more permanent disposal, such as to KONRAD.

In-situ disposal (or entombment) is not a recognized strategy for decommissioning within the IAEA Safety Standards in case of planned permanent shutdown of a nuclear facility. According to para. 5.17 of IAEA Safety Standards Series No. SSG-47, Decommissioning of Nuclear Power Plants, Research Reactors and Other Nuclear Fuel Cycle Facilities [81], entombment, in which all or part of the facility is encased in a structurally long-lived material, should not be considered as an acceptable strategy for core waste disposal. It might be considered only in ‘exceptional circumstances’ (e.g., for managing facilities that have been damaged in an accident).

6.5. PRACTICAL APPLICATIONS

The original intention of GRAPA, defined at the initial consultancy meeting, was to seek practical advances in the management of *i*-graphite. Suggestions made at the outset included the establishment of an international pilot-plant facility, and a potential location in the Russian Federation was offered by PDC UGR. The perceived difficulty in providing a modular test facility to refine all stages of the *i*-graphite management process was the obvious differences in the needs of the operator for each source of *i*-graphite, starting with the block design, their environment, different irradiation conditions, different accesses to dismantle, and so forth.

It quickly became apparent that the interests of the initial GRAPA members instead leaned heavily towards characterization and towards the development of innovative management processes as alternatives to the general Member States intentions to deliver *i*-graphite to a geological disposal facility, via interim storage. Some specialists are of the opinion that interim storage is not an appropriate route to follow since it involves additional handling and transport with potential consequences for personal dose exposure. However, in some situations, with some reactor designs, especially reactors containing very large amounts of graphite, this may be unavoidable.

Another aspect of *i*-graphite management which has become apparent during the project is that there are situations where more sampling of the *i*-graphite is desirable, but facilities have already been decommissioned. In such situations there is a reliance on modelling which cannot, however, reproduce all the potential source terms for activated materials except those present as impurities in the original graphite, and data on those is frequently incomplete and inaccurate, not least because of the high variability in the graphite, even within individual components.

Nonetheless, during the project, it has become clear that ongoing activities at nominated representative plants identified in the original GRAPA work-breakdown structure, have indeed demonstrated practical advances during the period of the programme, all related to the individual sub-topics studied. At INPP, extensive planning and preparations for dismantling are in progress and all graphite-related activities have been fully discussed within GRAPA: the same is true for the French pilot-dismantling planning at Chinon. Ukraine is developing its programme for characterization of *i*-graphite from the Chernobyl reactors along with design of facilities and equipment for graphite retrieval and handling based upon the experience of colleagues in the GRAPA project.

Development of handling equipment for removal of the core graphite blocks is well in hand at Latina, while significant progress has been made at two Russian production reactors (one by in-situ disposal and the other undergoing dismantling of the core). Active work is being undertaken at the Indian CIRUS plant and at small research reactors in Romania and Italy. Sellafield Ltd in the UK are characterizing the activity of the accumulating AGR fuel-sleeve graphite with the intention of identifying a proportion of the material as being suitable for disposal as LLW. Likewise, the Horia Hulubei National Institute of Physics and Nuclear Engineering (IFIN HH) intention is to dispose the entire quantity of the Romanian *i*-graphite coming from the decommissioning of the WWR-R research reactor inside its National Radioactive Waste Repository (NRWR) in Baita (Bihor County), based on the preliminary characterization results obtained under the CAST project.

Noted that one of the major successes of GRAPA has been to advise and inform those new to *i*-graphite management (groups dealing with small research reactors especially) of the depth of knowledge and experience of irradiated-graphite properties and behaviour, which has proven to be of great value in assisting the development of their dismantling programmes, and it is desirable that such international collaboration in this field is encouraged to continue.

6.6. IRRADIATED GRAPHITE MANAGEMENT SAFETY CASES

A safety case, in the context of radioactive waste management, is a suite of documents that assesses the safety and environmental implications of a proposed concept for waste handling, including its potential disposal. A safety case could cover the extraction of waste from an NPP under decommissioning, the packaging of waste, its transportation to. a geological disposal facility, the construction, operation and closure of this facility, and the evolution of emplaced waste post facility closure, to potentially a very long future timescale. A safety case demonstrates how waste can be safely managed and disposed in a manner that is compliant with national regulations often being informed by advice provided by the IAEA.

A safety case is therefore central to the iterative development of plans for radioactive waste management. Safety case documents include the specification, design, safety assessments and strongly depend upon the underpinning knowledge base which is itself derived from projects such as GRAPA. The purpose of safety case is to:

— Demonstrate that the organisation leading work on radioactive waste management is confident that the waste can be safely managed, potentially including safe disposal;

— Invite and support discussions with regulators and other interesting parties, such as waste producers;

— Maintain a basis on which the organisation can provide advice to waste producers on waste management options, including packaging for storage or disposal and assessing the disposability of waste packages;

— Support the waste management process by providing information to potentially affected communities;

— Inform the organisation’s forward plan by identifying research and development needs;

— Justify the siting of a facility to host waste;

— Provide a source of information for the development of site-specific designs and safety cases;

— Consider radiological safety in several phases of the system life cycle, potentially to disposal (if that were the end point);

— Transport the waste from the site of arising to the disposal facility or waste treatment and/or storage facility; this may involve more than one transport campaign;

— Construct, operate and potentially decommission of the facility to host the waste;

— To assess the long-term period, e.g., once waste has been emplaced, or once waste treatment has been completed.

The safety case includes conventional safety in the operational phase, socioeconomic assessment, health impact assessment and non-radiological environmental and sustainability assessments during the design development stages of the siting process for the waste storage facility. A site-specific environment impact assessment may be needed. Respective guidance is provided in IAEA Safety Standards Series No. GSG-10, Prospective Radiological Environmental Impact Assessment for Facilities and Activities [82].

Depending on how an organization chooses to operate, its research and development programme may be needs-driven, with R&D requirements identified primarily from the iterative development of a safety case. Such an approach lends justification to the prioritization

of R&D activities, ensuring work that will enhance and build confidence in the robustness of the safety case is undertaken in preference, with less important activities – in the context of the benefit they would bring to the safety case – deprioritized. It is important to recall here that a safety case necessarily needs to consider waste from the point of arising, through intermediate stages, to the proposed procedural stage where further active management is not needed, disposal to a suitable facility-needs-driven R&D is to consider this whole process and prioritize further R&D accordingly, rather than allowing undue focus on unimportant or nugatory matters – in the context of the safety case to be R&D prioritization drivers.

A needs-driven R&D programme, leading to the enhancement of an organisation's knowledge base, could be a fundamental component of the organization's business model, ensuring the delivery of the required research and technical development in the optimum cost-effective manner, whilst mitigating risks of future delays to the programme.

6.7. NEW INITIATIVES AND RECOMMENDATIONS

Consideration has been given in this TECDOC to reporting related activities that are being, or have been, undertaken in support decommissioning and radioactive waste management national programmes, by implementing organisations, by academia and by national laboratories, in relation to irradiated graphite. It is important to note that this publication along with Ref [13] are not in themselves intended to present a safety case for irradiated graphite management; rather, they provide research-derived information pertaining to this waste that could add to the associated knowledge base and could be used to enhance the robustness of a safety case in any national programme.

The GRAPA members considers that, in addition to the further work identified in Section 6.5 which is already planned to be conducted by the contributing organizations:

— The GRAPA members notes that, significant advances have been made towards graphite-reactor decommissioning (in test and commercial reactors) and there is evidence of an intent to reduce decommissioning timescales. It is therefore proposed that the network of irradiated-graphite specialists be maintained under the auspices of the IAEA's International Predisposal Network and the International Decommissioning Network, noting the need to be aware of but not to duplicate other international activities;

— The development of integrated waste management strategies is desirable, incorporating the results of specific investigations but taking note of potential interactions and economic consequences: as an example, a particular treatment option may appear desirable to reduce the activity of the *i*-graphite waste but needs to be assessed in terms of cost-benefit ratio given the cost of introducing new plant and the production of secondary waste streams. This may, for example, provide a basis for a decision to recommend intermediate level waste disposal rather than deep geological disposal in a dedicated *i*-graphite facility;

— Within the existing GRAPA work-package definitions, the following issues are highlighted for further development:

- Characterization:
    - Knowledge gaps exist regarding baked carbon, non-graphitized carbonaceous materials and graphite with high weight loss, where it is conceivable that permeability and porosity may affect isotope release rates;

- Investigation of the value of identifying *i*-graphite as a unique waste form rather than including it under the general ILW or LLW categories. It has very specific chemical, physical and mechanical properties which may lend themselves to categorizing it independently with specific rules and conditions applied. This reflects a general view that the risk associated with *i*-graphite (in terms of activity release) is lower than for other materials with the same current waste category;
- In addition to the Wigner energy issues noted in Section 6.1, there are two scenarios in which further assurance is needed. These relate to the possibility of energy release during grouting process which involve a heat cycle: this is unlikely to be a concern but needs to be assessed in individual cases where significant Wigner energy exists in the *i*-graphite. The second scenario is the creation of semi-adiabatic environments in storage facilities which lead to slow temperature rises (assisted by decay heat from other adjacent wastes) and release rates which are far slower than can be measured by conventional apparatus but which, over very long time periods, could result in significant temperature rises in the stored material and its containers which might increase beyond the range for which the containment has been evaluated.

- Removal/Retrieval:
  - It has become clear that significant damage could occur to graphite components during the operational period of the reactor. Methodologies and equipment needs to be developed to address the retrieval of fragmented *i*-graphite in these cases.
- Treatment:
  - The scientific development of graphite treatment options to support the desire for accelerated decommissioning;
  - Supporting the development of treatment and conditioning options for graphite would enable reclassification of some graphite material from ILW to LLW whilst still maintaining confidence in graphite stability,14C removal and long-term leach rates;
  - An improved understanding of chemical speciation is desirable to enable development of selective treatments targeted at long-lived radionuclides such as 36Cl;
  - Treatment options are required to deal with problematic waste forms (fuel-contaminated graphite, special fuels containing enriched uranium as admixture with graphite, waste streams as identified by Member States);
  - It is important to demonstrate that treatment processes can be up scaled and there is an adequate business case to do so.
- Packaging
  - It is noted that in certain Member States there are no packaging containers specifically developed for *i*-graphite. There is a potential for large cost savings if the unique properties of *i*-graphite are taken into consideration in the design of suitable packaging, additionally with savings in engineering complexity;

- It is considered that encapsulation of *i*-graphite could be improved and simplified compared with current practice when the unique nature of *i*-graphite is taken into consideration along with the development of alternative encapsulation materials.

There is a clear need to take a holistic view of the waste management process i.e., a whole life-cycle approach to *i*-graphite: with the lessons learnt from past reactors, the combined understanding can be used to enable high-temperature reactor (HTR) and molten-salt reactor (MSR) communities to minimize waste forms and reuse graphite for next generation use. This is particularly important for HTR fuel elements which have a very high and potentially reusable carbon content, and for future lithium fluoride and beryllium fluoride contaminated graphite (FLiBe-graphite), with and without integrated fuel from proposed MSR.

An example of the holistic approach is provided in Fig. 10. Whilst this needs some adaptation to be specific for *i*-graphite, the interactions are clear and appropriate procedures could be developed as part of future projects. This has a particular value when sections of the cycle changes and its impact upon the overall system determined.

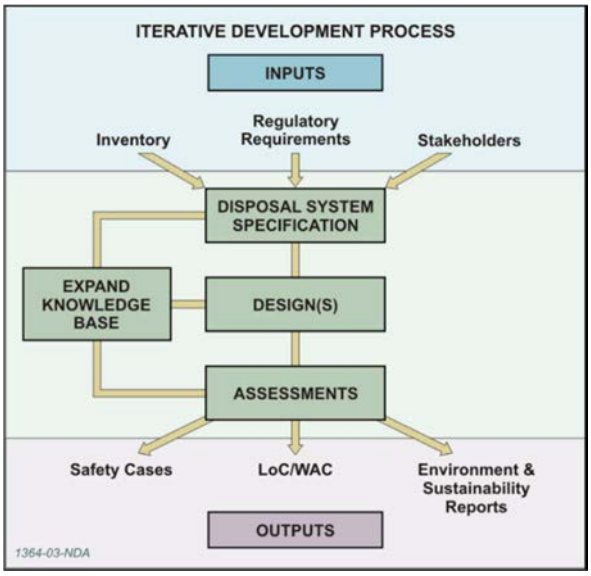

FIG. 10: Example of Iterative Disposal System Development: this can be applied to irradiated graphite, as well as other wastes. (Courtesy of NDA).

In Fig. 10, 'Letter of Compliance' (LOC), now referred to in the UK as the Disposability Assessment process: waste packagers liaise with RWM – the UK implementer for the Geological Disposal Facility – to ensure that plans for waste packaging will result in a packaged waste form that is compliant with the GDF safety cases and WAC.

A number of legal changes have occurred in Member States which may change the future management strategies for some categories of *i*-graphite including planning for disposal of the material. This is a clear example where knowledge sharing from other utilities and Member States will facilitate progress.

More specifically, methodologies are being developed for categorising individual graphite blocks, noting the wide variations in overall and local radiation dose which are being noted as reactor dismantling continues in the Russian Federation. This relates to a more general concern about the need to understand better the impact of radiation fields and the impact on personnel dose as well as the potential cost benefits in separation of the waste into independent streams of differing activity.

GRAPA members recommends that additional collaborative work in *i*-graphite processing will support those Member States in the process of managing irradiated graphite wastes or Member States developing or considering future new-build of graphite-moderated reactors and the *i*-graphite waste that will arise.

REFERENCES

[1] INTERNATIONAL ATOMIC ENERGY AGENCY, Graphite Moderator life cycle Behaviour, Proceedings of a Specialists Meeting held in Bath, United Kingdom, 24-27 September 1995, IAEA-TECDOC-901, IAEA, Vienna (1996).

[2] INTERNATIONAL ATOMIC ENERGY AGENCY, Nuclear Graphite Waste Management: Technical Committee Meeting on Nuclear Graphite Waste Management, Manchester UK, 18-20 October 1999, CD-ROM 01-00120, IAEA, Vienna (2001).

[3] INTERNATIONAL ATOMIC ENERGY AGENCY, Progress in Radioactive Graphite Waste Management; IAEA-TECDOC-1647, IAEA Vienna (2010).

[4] INTERNATIONAL ATOMIC ENERGY AGENCY, Characterisation, Treatment and Conditioning of Radioactive Graphite from Decommissioning of Nuclear Reactors, IAEA-TECDOC-1521, IAEA Vienna (2006).

[5] WHITE I.F., SMITH G.M., SAUNDERS L.J., KAYE C.J., MARTIN T.J., CLARKE G.H. and WAKERLEY M.W., Assessment of Management Modes for Graphite from Reactor Decommissioning, Commission of the European Communities, Report EUR-9232 (1984).

[6] BRADBURY D. and WICKHAM A.J., Graphite Decommissioning, EPRI, Palo Alto, CA: 1013091 (2006).

[7] BRADBURY D. and WICKHAM A.J., Graphite Dust Deflagration, EPRI, Palo Alto, CA: 1014797 and supplement 1015460 (2007).

[8] BRADBURY D. and WICKHAM A.J., Graphite Leaching, EPRI, Palo Alto, CA: 1016772 (2008).

[9] BRADBURY D. and MASON B., Program on Technology Innovation: Graphite Waste Separation, EPRI, Palo Alto, CA: 1016098 (2008)

[10] WICKHAM A.J., Carbon-14 in Irradiated Graphite Waste, EPRI, Palo Alto, CA: 1021109 (2010).

[11] BRADBURY D. and GOODWIN J., Innovative Graphite Removal Technology for Graphite Moderated Reactor Decommissioning: Nibble and Vacuum, EPRI, Palo Alto, CA: 1021110 (2010).

[12] WICKHAM A.J., Behaviour of Cl-36 and Tritium in Irradiated Graphite Wastes, EPRI, Palo Alto, CA: 1025312 (2012).

[13] WAREING A., ABRAHAMSEN L., BANFORD A., METCALFE M. and VON LENSA W., Deliverable D-0.3.12: Final Publishable CARBOWASTE Report, European Union (2013).

[14] GRAMBOW B., NORRIS S., PETIT L., BLIN V., COMTE J. and DE VISSER-TYNOVA E., Disposal Behaviour of Irradiated Graphite and Carbonaceous Wastes – Final Report; CARBOWASTE Work package 6 document 6.0.0, European Union (2013).

[15] INTERNATIONAL ATOMIC ENERGY AGENCY, Processing of Radioactive Graphite to Meet Waste-Acceptance Criteria for Waste Disposal; IAEA-TECDOC-1790, IAEA, Vienna, (2016).

[16] MARSDEN B.J. and WICKHAM A.J., Graphite Disposal Options – a Comparison of the Approaches proposed by UK and Russian Reactor Operators, Proc. International Conference on Nuclear Decommissioning, Nr. 9, I. Mech. E., London (1998) 145-153.

[17] NEIGHBOUR G.B., WICKHAM A.J. and HACKER P.J., Determining the Future for Irradiated Graphite Disposal, Nuclear Energy, **39** (2000) 179-186.

[18] OJOVAN M.I. and WICKHAM A.J., Treatment of Irradiated Graphite to Meet Acceptance Criteria for Waste Disposal: Problem and Solutions, Mat. Res. Soc. Symp. Proc., Nr.1665 (2014).

[19] OJOVAN M.I. and WICKHAM A.J., Radiation Effects in Graphite. Proc., Joint ICTP-IAEA Workshop on Radiation Effects in Nuclear Waste Forms and their Consequences for Storage and Disposal. International Centre for Theoretical Physics, Trieste, Italy, (2016) http://indico.ictp.it/event/7633/

[20] METCALFE M.P. and WAREING A., Deliverable D-1.7.2: Collation of Data/Information from other Work packages. Report of the CARBOWASTE project, European Union (2013).

[21] EU PROJECT ON CARBON-14 SOURCE TERM (CAST), Cast project, European Union (2018).

https://www.projectcast.eu/publications/reports

[22] WICKHAM A. J., personal communication (2020).

[23] KIRBY W., Brookhaven Graphite Research Reactor (BGRR) D and D Project – 11243; Waste Management Symposium 2011, Phoenix AZ (2011).

[24] WICKHAM A.J., STEINMETZ H.-J., O'SULLIVAN P. and OJOVAN M.I., Updating Irradiated Graphite Disposal: Project GRAPA and the International Predisposal Network, J. Environmental Radioactivity, **171 (**2017).

[25] GOODWIN J.D., BRADBURY D., BLACK S., TOMLINSON T., LIVESEY B., ROBINSON J., LINDBERG M., NEWTON C., JONES A. and WICKHAM A.J., From Core to Capture: Graphite management by gasification and carbon capture (2014).

https://inis.iaea.org/collection/nclcollectionstore/_public/47/090/47090149.pdf

[26] METCALFE M.P. and MILLS R.W., Radiocarbon Mass Balance for a Magnox Nuclear Power Station, Annals of Nuclear Energy, **75** (2015).

[27] WICKHAM A.J. and RAHMANI L., Graphite Dust Explosibility in: A Demonstration of Minimal Risk; supplementary material to IAEA, Progress in Radioactive Graphite Waste Management, IAEA-TECDOC-1647, IAEA, Vienna (2010).

[28] PHYLAKTOU H., ANDREWS G.E., MKPADI M., WILLACY S. and BOREHAM B., Explosion Hazards of Graphite Dust in Windscale Pile 1: Final Report on Phase 1, Leeds University Report, UK (2004).

[29] PHYLAKTOU H., ANDREWS G.E., MKPADI M., WILLACY S. and BOREHAM B., Explosion Hazards of Graphite Dust in Windscale Pile 1: Report on Phase 2 (Part 1) More Data/Analysis on Test Reproducibility, Graphite Ageing and Contamination Effects, Leeds University Report , UK (2004).

[30] MINSHALL P.C., The Contribution of Wigner Energy to Graphite Deflagration; J. Nuclear Materials, **492** (2017).

[31] CAST PROJECT, Modelling of C-14 migration from RBMK-1500 reactor graphite disposed of in a potential geological repository in crystalline rocks in Lithuania, (2018). https://www.projectcast.eu/publications

[32] PARMA G., ROSSI F.M., MOSSINI E., GIOLA M., MACERATA E., PADOVANI E., CAMMI A. and MARIANI M., MCNP Model of L-54 M Nuclear Research Reactor: Development and Preliminary Verification; J. Radioanal. Nucl. Chem., **318** (2018).

[33] ALMENAS K., KALIATKA A. and UŠPURAS E., Ignalina RBMK-1500: A Source Book., Lithuania Energy Institute, ISBN 9986492351, 9789986492351, Lithuania (1998).

[34] NARKUNAS E., SMAIZYS A., POSKAS P. and KILDA R., Assessment of Different Mechanisms of ^{14}C Production in Irradiated Graphite of RBMK-1500 Reactors, Kerntechnik, **75**, (2010) 185 – 194.

[35] BYLKIN B.K., DAVYDOVA G.B., ZVERKOV Y.A., KRAYUSHKIN A.V., NERETIN Y.A., NOSOVSKY A.V., SEYDA V.A. and SHORT S.M., Induced Radioactivity and Waste Classification of Reactor Zone Components of the Chernobyl Nuclear Power Plant Unit 1 after Final Shutdown, Nuclear Technology, **136** (2001)

[36] PUZAS A., REMEIKIS V., EŽERINSKIS Ž., SERAPINAS P., PLUSKIS A., and DUŠKESAS G., Mass-Spectrometric Determination of Impurities in Reactor Core Graphite for Radioactive Waste Composition Modelling; Lith. J. Phys., **50** (4) (2010).

[37] ANCIUS D., RIDIKAS D., REMEIKIS V., PLUSKIS A., PLUKIENĖ R. and COMETTO M.; Evaluation of the Activity of Irradiated Graphite in the Ignalina Nuclear Power Plant RBMK-1500 Reactor; Nukleonika, **50** (3) (2005).

[38] MACIEKA E., REMEIKIS V., ANCIUS D. and RIDIKAS D., Evaluation of the Radiological Consequences of ^{14}C due to Contaminated Ignalina NPP Graphite Incineration; Lith. J. Phys., **45** (5) (2005).

[39] INTERNATIONAL ATOMIC ENERGY AGENCY, Determination and Use of Scaling Factors for Waste Characterization in Nuclear Power Plants, IAEA Nuclear Energy Series No. NW-T-1.18, IAEA, Vienna (2009).

[40] LUKAUSKAS D., PLUKIENĖ R., PLUKIS A., GUDELIS A., DUŠKESAS G., JUODIS L., DRUTEIKIENĖ R., LUJANIENĖ G., LUKŠIENĖ B. and REMEIKIS V.;

Method of Determining the Nuclide Inventory for Low-Activity Waste of the RBMK-1500 reactor; Lith. J. Phys., **46** (4) (2006).

[41] REMEIKIS V., PLUKIS A., JUODIS L., GUDELIS A., LUKAUSKAS D., DRUTEIKIENĖ R., LUJANIENĖ G., LUKŠIENĖ B., PLUKIENĖ R. and DUŠKESAS G.; Study of the Nuclide Inventory of Operational Radioactive Waste for the RBMK-1500 Reactor; Nucl. Eng. Des., **239** (2009).

[42] PELOWITZ D. B., MCNP6 User‘s Manual, Report LA-CP-13-00634, Los Alamos National Laboratory, New Mexico (2013).

[43] SCALE: A Modular Code System for Performing Standardized Computer, Analyses for Licensing Evaluation, ORNL/TM-2005/39, Version 6,. **I–III** (2009).

[44] PLUKIENĖ R., PLUKIS A., PUZAS A., REMEIKIS V., DUŠKESAS G. and GERMANAS d.; Modelling of Impurity Activation in the RBMK Reactor Graphite Using MCNP, Progr. Nucl. Sc. Techn., **2** (2011).

[45] PLUKIENĖ R., PLUKIS A., BARKAUSKAS V., GUDELIS A., GVOZDAITĖ R. DUŠKESAS G. and REMEIKIS V.; Actinides in Irradiated Graphite of RBMK-1500 Reactor, Nucl. En. Des., **277** (2014).

[46] MOSSINI E., PARMA G., ROSSI F. M., GIOLA M., CAMMI A., MACERATA E., PADOVANI E. and MARIANI M.; Monte Carlo Integrated Approach to Radiological Characterization for Nuclear Facilities Decommissioning. Radiation Effects and Defects in Solids, **173** (2018)..

[47] MOSSINI E., CODISPOTI L., PARMA G., ROSSI F.M., MACERATA E., PORTA A., CAMPI F. and MARIANI M; MCNP Model of L-54M Nuclear Research Reactor: Validation by Preliminary Graphite Radiological Characterization; J. Radioanal. Nucl. Chem., **322** (2019).

[48] METCALFE P., TZELEPI, A., and COPELAND, G., The Release of Carbon-14 from Irradiated PGA Graphite by Thermal Treatment in Air, Annals of Nuclear Energy, **133** (110) (2019).

[49] ARNOLD L., Windscale 1957: Anatomy of a Nuclear Accident, Publisher. Gill & Macmillan Ltd, · ISBN-10. 0717119297, UK (1992).

[50] GALLEGO N.C. and BURCHELL T.D., A Review of Stored-Energy Release of Irradiated Graphite, Oak Ridge National Laboratory Report ORNL/TM-2011/378 (2011).

[51] KELLY B.T., Physics of Graphite; UKAEA Springfields Nuclear Power Development Labs., Springer ISBN 0 85334 960 6, London (1981).

[52] TELLING R.H. and HEGGIE M.I., Radiation Damage in Graphite, Philosophical Magazine, **87** (2007) 4796-4846.

[53] DOSTOV, APAVLIUK A.O., KOTLYAREVSKY S.G., BESPALA E.V. and NOVOSELOV, I.Y., Dynamics of Temperature Fields during Wigner Energy Release in Bulk Graphite Irradiated at Low Temperature; J. Nuclear Materials, **515 (**2019).

[54] MINSHALL P.C. and WICKHAM A.J., Wigner Energy and the Behaviour of Graphite from the Windscale Piles, BNFL Magnox Generation Report M/TE/GEN/REP/0037/99 (1999).

[55] WICKHAM A.J., BOWDEN E.A.T. and DE SIMONI L., Assessment of Graphite-Monitoring Data for Latina Power Station, CEGB Report (Berkeley Nuclear Laboratories) RD/B/6003/R88 (1988).

[56] SIMIRSSKII I., STEPANOV A., SEMIN I. and VOLKOVICH A., Determination of C-14 and Tritium in Irradiated Reactor Graphite; RAD Conference Proceedings, **3** (2018)

[57] PONCET B. and PETIT L., Method to Assess the Radionuclide Inventory of Irradiated Graphite Waste from Gas-Cooled Reactors, J. Radioanalytical Chem, **298** (2013).

[58] GY P., Sampling of Heterogeneous and Dynamic Material Systems – Theories of Heterogeneity, Sampling and Homogenising, Elsevier, Amsterdam (1992).

[59] HEASLER P.G. and JENSEN L., Statistical Evaluations of Current Sampling Procedures and Incomplete Core Recovery, Pacific Northwest Laboratories Report PNL-9408 (1994).

[60] FISHER, M., Fort St. Vrain Decommissioning Project, in Technologies for Gas-Cooled-Reactor Decommissioning, Fuel Storage and Waste Disposal, Proc. Technical Committee Meeting, Jülich, Germany, September 1997, IAEA-TECDOC-1043, IAEA, Vienna (1998).

[61] CANZONE,G., LO FRANO, R., SUMINI, M. and TROIANI, F., Dismantling of the graphite pile of Latina NPP: Characterization and handling/removal equipment for single brick or multi-bricks, Progress in Nuclear Energy, **93** (2016).

[62] GUIROY J.J., Graphite Waste Incineration in a Fluidised Bed; Proc. Specialists Meeting on Graphite Moderator Life Cycle Behaviour, Bath UK, September 1995, IAEA-TECDOC-901,IAEA, Vienna (1996).

[63] NAIR S., A Model for Global Dispersion of ^{14}C Released to the Atmosphere as CO_2; J. Soc. Radiological Protection, **3** (1983)

[64] MACEIKA E., REMEIKIS V. ANCIUS D. and RIDIKAS D., Evaluation of the Radiological Consequences of C-14 due to Contaminated Ignalina NPP Graphite Incineration; Lithuanian J. Physics, **45** (2005).

[65] PODRUZHINA T., Graphite as Radioactive Waste: Corrosion Behaviour under Final Repository Conditions and Thermal Treatment, Ph.D. Thesis (D 82), University of Aachen, and issued as Berichte des Forschungszentrums Jülich Report Jül-4166, Institut für Sicherheitsforschung und Reaktortechnik, ISSN 0944-2952 (2004).

[66] PONCET B., and PETIT L., EDF Analysis on the Origins of ^{14}C in UNGG Cores; 8th EPRI International Decommissioning and Radioactive Waste Workshop, Hamburg, Germany (2008).

[67] DUNZIK-GOUGAR, M.L., CLEAVER, J., LABRIER, D., NELSON, K. and SMITH, T., Chemical Characterization and Removal of Carbon-14 from Irradiated Graphite – III, Proceedings of Waste Management Symposium 2014, Phoenix, AZ (2014).

[68] NABBI R., PROBST H. and VON LENSA W., Molecular Dynamic Simulation of the Transport Behaviour of ^{14}C in Irradiated Nuclear Graphite, Irradiation Damage and ^{14}C Formation in Nuclear Graphite, Joint IAEA-CARBOWASTE Workshop, Visaginas, Lithuania (2013).

[69] WICKHAM A.J., Analysing Recoil Energy Effects to Determine Radioisotope Speciation and Behaviour in Irradiated Graphite; 13th International Nuclear Graphite Specialists Meeting, Meitingen Germany (2012).

[70] BESPALA E.V., ANTONENLO M.V., CHUBREEV D.O., LEONOV A.V., NOVOSELOV I.Y., PAVLENKO A.P. and KOTOV V.N., Electrochemical Treatment of Irradiated Nuclear Graphite; J. Nuclear Materials, **526** (2019).

[71] VOLKOVA A, ZAKHAROVA E, RODYGINA N, PAVLYUK A, SHIRYAEV A.; Radionuclides in Irradiated Graphite of Uranium–Graphite Reactors: Decontamination by Thermochemical Methods, Radiochemistry, **60**(6) (2018).

[72] VOLKOVA A, ZAKHAROVA E, PAVLYUK A, SHIRYAEV A.; Radionuclides in Irradiated Graphite of Uranium Graphite Reactors: Decontamination of Sleeves Using Liquid Reagents, Radiochemistry, **60**(5) (2018).

[73] NEGIN C.A., SZILAGYI A.P., COLLAZO Y.T., LEE P., GLADDEN J. SEITZ R. and WILSON M., In Situ Decommissioning Moves Ahead., Waste Management Symposium 2010, Phoenix, AZ, (2010).

[74] LANGTON C.A., SERRATO M.G., BLANKENSHIP J.K. and GRIFFIN W.B., Savannah River Site Reactor Disassembly Basin *In-Situ* Decommissioning, Waste Management Symposium 2010, Phoenix, AZ (2010).

[75] INTERNATIONAL ATOMIC ENERGY AGENCY, Techno-Economic Comparison of Geological Disposal of Carbon Dioxide and Radioactive Waste, IAEA-TECDOC-1758, IAEA, Vienna (2014).

[76] HESPE, E.D., Leach Testing of Immobilised Radioactive Waste Solids: A Proposal for a Standard Method, Atomic Energy Reviews, **9** (1971).

[77] PETROVA E., SHCHERBINA N., WILLIAMS S. and PIÑA; Definition of a Recommended Scientific Scope of Leaching Experiments and Harmonised Leaching Parameters, CAST Project deliverable D5.4 (2015).

[78] SHEPPARD S.C., JOHNSON L.H., GOODWIN B.W., TAIT J.C., WUSCHKE D.M. and DAVISON C.C., Chlorine-36 in Nuclear Waste Disposal – 1: Assessment Results for Used Fuel with Comparison to ^{129}I and ^{14}C, Waste Management, **16** (7) (1996) 604-614.

[79] BYTWERK D., LIMER L., ALBRECHT A., MARANG I., SMITH G. and THORNE M., Sources and Significance of the Variation in the Dose Estimates of ^{36}Cl Biosphere Transfer Models: A Model Intercomparison Study, Journal of Radiological Protection, **31** (2011).

[80] KANE J.J., CONTESCU C.I., SMITH R.E., STRYDOM G. and WINDES W.E., Understanding the Reaction of Nuclear Graphite with Molecular Oxygen: Kinetics, Transport, and Structural Evolution; J. Nuclear Materials, **493** (2017).

[81] INTERNATIONAL ATOMIC ENERGY AGENCY, Decommissioning of Nuclear Power Plants, Research Reactors and Other Nuclear Fuel Cycle Facilities, IAEA Safety Standards Series No. SSG-47, IAEA, Vienna (2018).

[82] INTERNATIONAL ATOMIC ENERGY AGENCY, Prospective Radiological Environmental Impact Assessment for Facilities and Activities, IAEA Safety Standards Series No. GSG-10, IAEA, Vienna (2018).

CONTENTS OF THE ANNEXES

Annexes contains supplemental electronic files on each of the four Work-Package topics with detailed reports provided by the GRAPA members. This information does not necessarily reflect best practice available, and no judgement is made on the situations described. The information presented is not intended to be exhaustive and can be found on the publication's individual web page at www.iaea.org/publications. The following annexes are available as part of this publication:

Annex I: Characterization;

Annex II: Graphite retrieval/removal;

Annex III: Graphite treatment and conditioning;

Annex IV: Packaging, storage and disposal.

LIST OF ABBREVIATIONS

AGL	Anglo Great Lakes (former graphite manufacturer, UK)
AGOT	USA Graphite Grade
AGR	Advanced Gas-Cooled Reactor (UK)
ANDRA	French National Radioactive Waste Management Agency
AVR	Arbeitsgemeinschaft Versuchsreaktor Jülich, Germany
BAEL	British Acheson Electrodes Ltd (former graphite manufacturer, UK)
BEPO	British Experimental Pile Zero (Harwell, UK)
BGRR	Brookhaven Graphite Research Reactor (USA)
CARBOWASTE	EU Collaborative Research Project on Carbon-Based Nuclear Wastes
CANDU	Canadian Deuterium/Uranium Reactor Design
CAST	Carbon-14 Source Terms (EU Collaborative Project)
CCS	Carbon Capture and Storage
CEA	Atomic Energy Commission (France)
CIEMAT	Centro de Investigaciones Energéticas, Medioambientales Tecnológicas (Spain)
CIRUS	Research Reactor at Bhabha Atomic Research Centre, India
CPST	Centre for Physical Science and Technology, Vilnius, Lithuania
CRP	Coordinated Research Project (IAEA)
EDF	Electricité de France
ENEA	National Agency for New Technologies, Energy and Sustainable Economic Development, Italy
ENRESA	Empresa Nacional de Residuos Radiactivos, S.A., Spain
EPRI	Electric Power Research Institute (USA)
FNAG	Furnaces Nuclear Applications Grenoble, now part of ALD France S.A.
GDF	Geological Disposal Facility
GRAPA	Irradiated Graphite Processing Approaches
HPGe	High Purity Germanium Detector
HTR (HGTR)	High Temperature Gas-Cooled Reactor
HVP	High Vacuum Pressing
ICP-MS	Inductively-Coupled Plasma Mass Spectrometry
IMMONET	IAEA Platform for Archiving Project Working Materials
KAERI	Korea Atomic Energy Research Institute
KONRAD	Former Iron-Ore Mine in Germany proposed as Waste Storage
LLW	Low Level Waste
LoC	Letter of Comfort

LSC	Liquid Scintillation Counting
MSR	Molten Salt Reactor
NNL	National Nuclear Laboratory (UK)
NPP	Nuclear Power Plant
PBMR	Pebble Bed Modular Reactor Co. Pty (Rep. South Africa)
PDC UGR	Pilot & Demonstration Centre for Decommissioning of Uranium-Graphite Nuclear Reactors (Seversk, Russian Federation)
PGNAA	Prompt Gamma Neutron Activation Analysis
PNNL	Pacific Northwest National Laboratory (USA)
POLIMI	Politecnico di Milano (Italy)
RBMK	Reaktor Bolshoy Moshchnosti Kanalnyy, 'High Power Channel-type Reactor' (Soviet Design)
RWM	Radioactive Waste Management Ltd (UK)
SoGIN	Società Gestione Impianti Nucleari, Italy: State Company responsible for Decommissioning
THTR	Thorium High-Temperature Reactor (Germany)
UNGG	Natural Uranium Fuelled Graphite-Moderated Reactor (France)
WWR-S	Nuclear Research Reactor of Horia Hulubei National Institute of Physics and Nuclear Engineering at Magurele-Bucharest
VATESI	Nuclear Regulatory Body, Lithuania

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