Treatment of Irradiated Graphite to meet Acceptance Criteria for Waste Disposal: A New IAEA Collaborative Research Program - 12443

A.J. Wickham[1] and Z. Drace[2]
[1]Nuclear Technology Consultancy, PO Box 50, Builth Wells, Powys LD2 3XA, United Kingdom
[2]Waste Technology Section, Division of Nuclear Fuel Cycle and Waste Technology, International Atomic Energy Agency, Wagramerstraße 5, PO Box 100, A-1400, Vienna, Austria

ABSTRACT

World-wide, more than 250,000 tonnes of irradiated graphite have arisen through commercial nuclear-power operations and from military production reactors. Whilst most nations responsible for the generation of this material have in mind repository disposal alongside other radwaste, the lack of progress in this regard has led in some cases to difficulties where, for example, the site of an existing graphite-moderated reactor is required for re-utilisation. In any case, graphite as a radwaste stream has unique chemical and physical properties which may lend itself to more radical and innovative treatment and disposal options, including the recovery of useful isotopes and also recycling within the nuclear industry. Such aspects are important in making the case for future graphite-moderated reactor options (*for example*, High-Temperature Reactors planned for simultaneous power production and high-grade heat sources for such applications as hydrogen production for road fuel). A number of initiatives have taken place since the mid 1990s aimed at exploring such alternative strategies and, more recently, improving technology offers new options at all stages of the dismantling and disposal process. A new IAEA Collaborative Research Program aims to build upon the work already done and the knowledge achieved, in order to identify the risks and uncertainties associated with alternative options for graphite disposal, along with cost comparisons, thus enabling individual Member States to have the best-available information at their disposal to configure their own programs.

INTRODUCTION

From the present population of research, production and commercial power reactors moderated by graphite, along with a smaller contribution from weapons-programme waste, some 250,000 tonnes of radioactive graphite will arise for disposal during decommissioning activities. The conventional wisdom is that this material, mostly intermediate-level waste (ILW), would be encapsulated to immobilise the radioactive content during handling and storage, and disposed of in a number of deep repositories, along with other waste including suitably-treated high-level wastes. The primary need for encapsulation and deep burial of the graphite, as presently perceived by the industry, is the presence of the radioactive isotope Cl-36, which has a half-life of 301,000 years, and of certain other highly mobile radioisotopes, because of their propensity to enter the food chain upon leaching from the repository. The reality is that no such repository exists after several decades of good intentions, and only France has recently devoted attention to the design of a separate repository specifically intended for graphite waste together with radium-bearing waste.

THE NEED FOR PROGRESS

The global need for CO_2-free energy production in the 21st century, and a growing acceptance in a number of Member States that nuclear generation should form part of a balanced mix of diverse sources of supply, means that a re-vitalisation of nuclear electricity generation is likely, and utilities are looking to existing licensed sites to install new generating capacity. There is therefore increasing pressure to dismantle retired graphite-moderated plant in order to release site availability, two specific and urgent examples being Ignalina (Lithuania) and Tokai (Japan).

The capability of mechanical dismantling of graphite stacks has already been demonstrated (Brookhaven and Fort St. Vrain, USA and Windscale AGR, UK), but this has resulted in the undesirable outcome of the graphite residing in a 'temporary' surface storage where it can be argued that leaving it inside the original pressure vessel (where they exist) until a final disposal route became available would be a more suitable option from the perspective of radiological safety. Only in the case of the UK very-low-energy reactor GLEEP (which was effectively dismantled by hand because of its low radioactivity burden) has the graphite been treated to bring its isotope content below the limits for 'free release' of the material: this was done by calcinations, to mobilise the C-14 and H-3 after first 'crumbling' the graphite.

The key question therefore is to identify the ideal graphite-disposal route in each example and to demonstrate this to the international community. For some Member States of the IAEA, problems have arisen with planned graphite-disposal methodologies and/or timescales and a review of alternatives is now seen as an urgent priority: in other cases, a long-term disposal plan exists into which the clearance of graphite has been integrated, but there remains an 'open mind' about other options and a willingness to consider them.

INNOVATIVE APPROACHES

Alternative disposal options for irradiated graphite have been discussed for at least 30 years, with some, such as fluidised-bed incineration, being developed to a pilot-plant stage [1]. In the mid 1990s, incineration of graphite was being taken quite seriously, and a UK analysis showed that combusting graphite at the rate of one 'Magnox' core per year for twenty years would raise the global C-14 burden by less than one part of in 1000, suggesting that this activity would be virtually unnoticed against the high background production of C-14 in the upper atmosphere by the reaction $^{14}N(n,p)^{14}C$ [2]. However, concerns about the *local* dose effects of discharging C-14 from a commercial facility, and the obligation to regard artificial isotopes as somehow different from their naturally-occurring equivalents, have led to political indifference and opposition to such alternatives. Other respected authorities have published contrary opinions about the dangers posed by graphite wastes (*e.g.* the comprehensive analysis in [3]), and it is desirable to place the true objective risk into a proper context alongside the natural background of radioactivity which surrounds us all [4].

It is now being recognised internationally that the business of graphite stack dismantling and disposal must receive renewed and innovative thought. The European Union has funded a major project under the Seventh Framework Initiative, *CARBOWASTE*, in which more than 20 European and international organisations and partners are studying the viability of alternative strategies, and also using improved modern techniques for characterization of the isotopic content of irradiated graphites, including their distribution within different features of the graphite matrix (coke/filler particles, binder phase, impurity inclusions, pore surfaces). CARBOWASTE is currently negotiating an extension to its program schedule with the EU in order to follow sup some of the more promising lines of investigation.

The US body EPRI ('Electric Power Research Institute') has also instigated a comprehensive review of all features of dismantling graphite-moderated reactors and disposing of the material: the work has included comprehensive investigations of investigations of graphite recovery, dust explosibility (the potential for which is frequently overstated [5]), leaching behaviors, the creation and subsequent behaviour of specific long-lived beta emitters (C-14, Cl-36 and H-3), and isotope recovery [6-12].

THE ROLE OF THE IAEA

Recognising the unique nature of the irradiated-graphite waste stream, and the quantities of irradiated graphite to be disposed, IAEA aims to build on the initiatives mentioned earlier and to assist Member States in refining their national strategies for disposal of such wastes. Already, three highly significant Technical Discussions on this subject have taken place, resulting in the publication of three valuable compilations of technical papers [13-15] and a separate technical review [16]. The present Collaborative Research Program will be a new initiative, undertaking by examination and by practical demonstration the potentially available technologies for treatment, handling or disposal, which may offer any or all of the following advantages:

- a significant reduction in waste volume or packaging requirements;
- pre-treatments which either reduce significantly the radioisotope content of the material, or facilitate other processing options, or both;
- a significant cost saving, either through operational efficiencies or by allowing a useful product or products to be recovered from the material;
- a favourable timescale (earlier option to achieve some disposal result which would otherwise be delayed).

The new IAEA Collaborative Research Programme (CRP) is intended to complement CARBOWASTE program and, under the direction of the present authors (IAEA Scientific Secretary, *ZD* and Chief Scientific Investigator, *AJW*), it will address the following issues:

1. Direct chemical or physical treatment of graphite leading to its disposal in an alternative form to solid graphite, with economic and long-term radiological benefits;
2. Pre-treatment of graphite ahead of other disposal or innovative treatment, usually in order to reduce the radio-isotope content and to facilitate the economics and radiological safety of the following process operations;
3. Treatment of the products of innovative process to improve radiological safety or for economic improvement (such as separation and recycling of useful isotopes for the nuclear and/or medical industries).

It is considered that investment in these practical evaluations offers a real potential economic value in assisting the effective disposal or recycling of graphite materials from the nuclear programme. An initial wide-ranging discussion amongst the members of the CRP has identified the following more specific issues:

- Methods for 'cleaning' of graphite to be evaluated (reduction of radio-isotopic content and/or, where relevant, other toxins such as heavy metals);
- Isotope separation techniques, to include thermal diffusion, pressure-swing absorption, centrifuging and cryogenic methods, for all relevant isotopes;
- The philosophy of 'dilute and contain' for treatment of specific isotopes such as C-14;
- Novel handling methodologies which facilitate alternative treatments (*e.g.* graphite removal as crumbled and vacuumed particulate, transfer in liquid foams, etc.).

…identifying, for all options:

- Feasibility (by practical demonstration); *and*
- Cost effectiveness

The CRP presently has participation from more than 20 organisations in 11 IAEA Member States.

It is important that the breadth of innovation that is being considered here is understood. A number of the submissions to the Collaborative Research Program are in carefully coordinated groups; one draws together the activities of ANDRA, CEA and EdF CIDEN in France along with the specialised knowledge of radioisotope generation, characterization and mobilisation of the Institute of Nuclear Physics of Lyon in France; a group of UK organisations (Bradtec Ltd, Hyder Consulting, Studsvik UK and Costain Ltd) has put together a program which investigates in its entirety an innovative retrieval technology methodology, isotope recovery stage, gasification and ultimate disposal technology in gaseous form alongside sequestered flue gas from conventional power plant into depleted oil or gas reservoirs where the sequestered gas could have a lower specific radioactivity content than natural background (making use of the Suess effect whereby fossil fuels are naturally depleted in C-14 [17]. Other organisations are investigating methods specifically designed to deal with graphite which is highly contaminated by fuel debris: these methods also offer potential benefits to other groups (for example, a possible application to the graphite from Windscale Pile 1 in the UK where fire-damaged fuel remains distributed in regions of the stack [18]).

It is also proposed to include evaluation of the possibilities for, as examples, the transport of graphite waste as small particulate in aqueous foams, and direct removal of graphite from pressure vessels, both by chemical and innovative mechanical means, with appropriate ‘treatment’ as necessary.

It is acknowledged that there remains within the international waste community a body of opinion which strongly adheres to the waste routes which were identified more than thirty years ago. However, these have manifestly failed to ‘deliver’ on a reasonable timescale, and the high confinement targets set for these routes have resulted in projected costs mounting to hundreds of millions of dollars, which have contributed to the general public perception until recently that nuclear generation is both radiologically unsafe and extremely expensive, notably in regard to the waste.

The Collaborative Research Program held its first ‘Research Coordination Meeting’ in November 2011 in which the principal purpose was to draw up a timescale for completion of the suggested work, to define deliverables and milestones along the way. The Program expects to produce its final report as an IAEA TECDOC in 2015, although interim proposals and recommendations may well appear earlier. It is *not* the intention to provide a substantive IAEA recommendation to Member States, but rather to evaluate all known options and alternatives for handling of this considerable waste stream using objective risk analysis alongside a cost-benefit analysis, in order that individual Member States may use the results to inform their own programs for graphite waste disposal.

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