Disposal Solution for Graphite Waste from the Decommissioning of Chernobyl NPP*

Borys Zlobenko, Mykola Proskura
Chornobyl Nuclear Power Plant

At present the reactors of Chornobyl NPP, power units 1, 2, and 3 are shut down (1996, 1991, 2000) and are at the stage of being removed from service. Chornobyl power units 4 was destroyed in the 1986 after accident.
Graphite management is one a keys issue when it comes to the decommissioning of 1-3 units Chernobyl NPP with reactor type RBMK-1000. Graphite is used in RMBK reactors as a moderator and reflector. Fission products and activation of impurities in the graphite contaminate this graphite during reactor operation.
Decommissioning of Chornobyl NPP

The problem of shutdown reactor decommissioning in the part of nuclear graphite is studied in order to developing principles on safe handling with irradiated graphite and technologies.

Some of these are:

- definition of radioactivity, its distribution;
- investigation of graphite properties;
- technology of graphite bricks dismounting;
- technologies of chemical and physical influence on radioactive graphite (breaking, milling, cutting of layers with fuel, impregnation by conservants, burning of graphite etc.);
- technologies of storage.
Recently developed National Strategy and Concept for State Programme for Radioactive Waste Management in Ukraine (Part 1), including a Strategy for NNEGC Energoatom Radwaste Management (Part 2.) Based on identified deficiencies the strategy proposed to:

- to improve the classification system by categorizing the waste in four categories based on four disposal options, including the category of very low level waste,

- to develop an integrated radwaste management system at NPPs, including reduction of radioactive waste generation, optimization of solid and liquid waste treatment, radiation protection during waste treatment, technologies for decontamination of large components and systems for control and accounting of waste at all stages.
Last years several of normative documents have been developed by SNRCU in compliance with international practice:


- Basic sanitary rules for radiation safety of Ukraine (ospu-2005), approved by the Ministry of Health of Ukraine No. 54 of 2 February 2005 and registered in the Ministry of Justice of 20 May 2005

- Procedure for issuing certificates on approval of package designs and radioactive materials, special conditions and some shipments (NP 306.5.06-2003), registered in the Ministry of Justice of 23 May 2003
The classification of radwaste and exemption levels are prescribed in a national General Sanitary Rules (OSPU-2005). The general protection rules are set by Norms of Radiation Safety -97 (NRBU-97) and Supplement D/2000 (NRBU-97/D2000). These documents are based on ICRP 60 publication (establishing annual dose limit for any member of the public - 1 mSv and occupational dose limit 20mSv committed on average per year during any 5 years but not greater 50 mSv by one year).
<table>
<thead>
<tr>
<th>Waste group</th>
<th>Solid radwaste</th>
<th>Exemption level, kBq/kg</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Transuranium alpha bearing radionuclides</td>
<td>0.1</td>
</tr>
<tr>
<td>2</td>
<td>Alpha bearing radionuclides (with the exception of transuranium nuclides)</td>
<td>1</td>
</tr>
<tr>
<td>3</td>
<td>Beta-gamma bearing (with the exception of 4 group)</td>
<td>10</td>
</tr>
<tr>
<td>4</td>
<td>H-3, C-14, Cl-36, Cf-45, Mn-53, Fe-55, Ni-59, Ni-63, Nb-93m, Tc-99, Cd-109, Cs-135, Pm-147, Sm-151, Tm-171, Tl-204</td>
<td>1000</td>
</tr>
</tbody>
</table>

According to the Ukrainian standard the radioactive residual material is considered to be radioactive waste if activity concentration exceeds the following exemption levels.
On the basis of activity concentration, radwaste are classified on low-, intermediate and high level waste categories.
The radioactive waste classification based on acceptability of disposal mode

| Radwaste type                        | The potential exposure after 300 years | Possible release mode from regulatory control beyond a period of 300 years | Acceptable mode of disposal |
|--------------------------------------|----------------------------------------|****************************************************************************|
| Short-lived                          | Less than 1 mSv/year                   | Full, restricted release                                                  | Near surface or shallow ground repositories |
| Determined by reconciliation with regulatory body | Grater than 1 mSv/year and smaller than 50 mSv/ year | Restricted release is allowed                                               | Determined by reconciliation with regulatory body |
| Long-lived                           | Grater than 50 mSv/ year               | Unconsidered                                                               | Geological repositories         |
Radioactive waste management

Radioactive graphite require special attention if it are classified as radioactive waste.

Graphite as a radioactive waste presents a unique set of technical issues, including both surface contamination and bulk activation, accumulation of energy (Wigner energy), and difficulties in volume reduction and processing into a chemically stable form.

Currently, experience in the conditioning of graphite from large scale facilities is not yet available, even though a number of techniques for the treatment of radioactive graphite have been developed.
Radioactive waste management

- In compliance with the requirements of the regulatory documents, the composition and activities of radionuclides accumulated in structural materials and structures during the operation of the NPP power unit must be evaluated before its removal from service.
The information on the wastes to be disposed of in the repository needed for a safety assessment would be as follows:

- radionuclide inventory;
- physical characteristics of waste;
- chemical characteristics of waste;
- form of waste packaging.
Technological channels with graphite elements RBMK

The graphite bricks are manufactured from GR-280 graphite. The size of a brick is 600 (500,300,200) mm high by 250 mm x 250 mm square, with a central hole 114 mm diameter. The split graphite rings are manufactured from a better quality graphite GRP2-125 with a higher density.
<table>
<thead>
<tr>
<th>Isotope</th>
<th>Outer surface Contents, Ku/probe</th>
<th>Inner surface Contents, Ku/probe</th>
</tr>
</thead>
<tbody>
<tr>
<td>Co-60</td>
<td>3.1 $10^{-7}$</td>
<td>Co-60</td>
</tr>
<tr>
<td>Cs -134</td>
<td>1.2 $10^{-8}$</td>
<td>Cs- 134</td>
</tr>
<tr>
<td>Cs-137</td>
<td>2.0 $10^{-8}$</td>
<td>Mn-54</td>
</tr>
<tr>
<td>Total $\gamma$</td>
<td>3.4 $10^{-7}$</td>
<td>Total $\gamma$</td>
</tr>
</tbody>
</table>

Maximal neutron fluence on the inner surface of graphite bricks was about $1 \cdot 10^{22}$ neutron/cm$^2$ (En > 0.18 MeV). The graphite temperature was estimated as 500–600°C.

(P.A. PLATONOV, at all., 1999)
Split graphite rings

- The graphite rings were taken from different places along the reactor core (RC) height (mainly from the RC center and the region with the maximum neutron fluence).

- After the termination of irradiation (1991), the graphite rings were kept in water in the cooling pond at a temperature of 40–60°C until they were taken out in 2002.
Split graphite rings

The GRP–2–125 graphite porosity measurement showed that the total porosity of the sample was practically identical for irradiated and nonirradiated graphite, $P_{\text{irr}} = 0.145 \pm 0.029$ and $P_{\text{nonirr}} = 0.153 \pm 0.018$ as measured by the scanning electron microscopy, and $P_{\text{irr}} = 0.149 \pm 0.012$ and $P_{\text{nonirr}} = 0.164 \pm 0.012$ as follows from the density measurement by the gravimetric method. With these values, open porosity amounts to 90% of total porosity.
Table 1. Isotope content of the GRP-2-125 graphite rings

(M. D. Bondar’kov, 2009)

<table>
<thead>
<tr>
<th>Sample</th>
<th>Specific concentration, Bq/g</th>
<th>Total activity, Bq g⁻¹</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>^3H</td>
<td>^14C</td>
</tr>
<tr>
<td>B 427 1062</td>
<td>490</td>
<td>8520</td>
</tr>
<tr>
<td>B 427 603</td>
<td>690</td>
<td>4080</td>
</tr>
<tr>
<td>B 427 962</td>
<td>1230</td>
<td>12100</td>
</tr>
<tr>
<td>B 427 1145</td>
<td>1050</td>
<td>28000</td>
</tr>
<tr>
<td>G 75 962</td>
<td>610</td>
<td>4000</td>
</tr>
<tr>
<td>G 75 1062</td>
<td>1740</td>
<td>13280</td>
</tr>
<tr>
<td>G 75 1145</td>
<td>605</td>
<td>5600</td>
</tr>
<tr>
<td>G 75 603</td>
<td>620</td>
<td>10840</td>
</tr>
<tr>
<td>G 209 1062</td>
<td>820</td>
<td>14600</td>
</tr>
<tr>
<td>G 209 962</td>
<td>520</td>
<td>7240</td>
</tr>
<tr>
<td>G 209 603</td>
<td>680</td>
<td>14400</td>
</tr>
<tr>
<td>G 209 1145</td>
<td>750</td>
<td>10720</td>
</tr>
<tr>
<td>B 299 1145</td>
<td>980</td>
<td>9080</td>
</tr>
<tr>
<td>B 299 1062</td>
<td>650</td>
<td>5360</td>
</tr>
<tr>
<td>B 299 962</td>
<td>450</td>
<td>10600</td>
</tr>
<tr>
<td>B 299 603</td>
<td>710</td>
<td>8520</td>
</tr>
<tr>
<td>B 358 1145</td>
<td>420</td>
<td>8960</td>
</tr>
<tr>
<td>B 358 962</td>
<td>780</td>
<td>6320</td>
</tr>
<tr>
<td>B 358 1062</td>
<td>890</td>
<td>10400</td>
</tr>
<tr>
<td>B 358 603</td>
<td>1290</td>
<td>18000</td>
</tr>
<tr>
<td>Average isotope activity</td>
<td>799</td>
<td>10531</td>
</tr>
<tr>
<td>Isotope percentage</td>
<td>4.22</td>
<td>55.65</td>
</tr>
</tbody>
</table>
Activity of the irradiated GRP–2–125 graphite is determined first of all by the $^{14}$C content and also by the content of impurity and technogenic radionuclides ($^3$H, $^{36}$Cl, $^{55}$Fe, $^{59}$Ni, $^{60}$Co, $^{63}$Ni, $^{93}$m$^{108}$m$^{133}$Ba, $^{154}$Eu, $^{155}$Eu). Contamination of the GRP–2–125 graphite by fuel fission products results from lost of tightness by the channels in the course of reactor operation.
Problems with disposal of graphite

- There are two principal graphite aging processes:
  - irradiation damage from fast neutrons creates lattice defects leading to dimensional changes;
  - changes in properties including coefficient of thermal expansion, thermal conductivity, modulus and strength.

- Exposure gamma irradiation results in radiolytic graphite oxidation.

- Irradiated graphite contains relatively high concentrations of C-14 and Cl-36.

- Irradiated graphite contains a number of active species including 3-H, 55-Fe, 60-Co and 63-Ni.

- A part of the graphite waste will be contaminated with fission products and actinides as a consequence of fuel element failures.
In accordance with the Ukrainian regulatory requirements, such waste must be immobilised to a stable physical and chemical form. Graphite waste contaminated (accident on power unit 2) by fuel leakage is classified as high-level waste, containing fissile materials.

The waste inventory of irradiated graphite waste - near 6000 t. in ChNPP Units 1-3 and 0,8t in Shelter). Since the graphite blocks may have various activity levels and different contaminations, it is necessary to define a strategy: treatment-reduction of volume and conditioning-storage.
The radionuclide inventory of irradiated graphite is unusual in comparison with other nuclear wastes. The principal isotopes of short term importance are cobalt-60 and tritium; in the longer term carbon-14 and chlorine-36 are dominant.

The three main options for disposal of graphite are oxidation to the gas phase and release as carbon dioxide, direct burial or recycling into new products for the nuclear industry. In each case opportunities exist for pre-processing to concentrate or remove radionuclides and thereby enhance the safety of the chosen option.
A significant difficulty for the treatment of graphite stacking elements with fragments of nuclear fuel. It should be noted that graphite reactor of Unit-2 stacking was contaminated Chernobyl fission products and transuranic elements as a result of the accident in 1982, with the technological gap channel and ingress of nuclear fuel in a stacking vessel.
Waste volume reduction technologies

Graphitic waste → Pre-treatment, e.g. pulverisation → Thermal oxidisation → Ash → ILW Repository

Gaseous release or ILW 3H, 36Cl & 14C solids

Treated Graphite → LLW repository or free release/reuse
Radwaste- solid waste immobilised with cement grout in concrete containers KTZ-3.0 as received after sorting and processing (fragmentation, incineration, compaction) of solid radwaste from ChNPP interim storages (operational and accident waste). Radwaste resulting from sorting and processing constitutes ash briquettes after incineration and compaction of combustible waste; briquettes of compacted waste; metal fragments etc.
Characteristics of disposal containers and their contents

- **Metal drum KT-0,2 (type I package)**
  - material – carbon steel;
  - outer diameter, mm – 600;
  - height, mm – 843;
  - wall thickness, mm – 1,5
  - inner volume – 0.2 m³
  - drum weight – 37 kg

- **Concrete container KTZ-3.0 (type II package)**
  - material – reinforced concrete;
  - outer dimensions – 1940×1940×1660 mm;
  - inner volume – 3,0 m³;
  - total volume – 6,25 m³;
  - wall thickness – 150 mm;
  - sorption, permeability – data not available
The characteristics of packages should comply with the disposal acceptance criteria established by the “Vector” Operator, which include requirements on permissible values of the dose rate on package surface, absence of aggressive chemicals, explosives, combustible materials inside the packages etc. There are also requirements on the waste radionuclide composition, specific and total activity radwaste and permissible content of individual radionuclides, including long-lived alpha- and beta-emitting radionuclides.
Standard waste drums (200 litres) may be suitable for used the small quantities of graphite rings produced from the reactor dismantling graphite programme which will packed and may be storage at Vector site.

Storage boxes with a concrete lining to provide the necessary shielding are deemed to be more suitable for transporting such material to a repository. The shielding requirement is dependent upon the elapsed time since reactor shutdown.
Alternative disposal methods to deep geological repository


- The feasibility study was carried out to identify sites for hosting a radioactive waste repository in a disused mine similar to potential repository sites in Germany (e.g. Konrad).
Objectives of the Project

- The objective of this generic study was to look into possibility of disposing of long-lived non-heat-generating waste in the near future by emplacing such waste in suitable dry geological formations.
- In order to be cost-effective, already existing mines (where access shafts and preferably transport equipment are present) could be used for this purpose.
- Cost (macro-economic), safety and environmental assessment of options for managing graphite.
After an appropriate ranking process and consideration of the availability of sufficient assessment data, the Saksagan iron ore mine at Krivoy Rog was selected as the lost appropriate to host a repository for long-lived non-heat-generating low- and intermediate-level radioactive wastes.

Such wastes may origin in the decommission of nuclear power plants or from the remediation of contaminated areas after the Chernobyl accident.

Since most of the information needed as a safety assessment basis such as radionuclide inventory, physical and chemical characteristics of the waste and the form of waste packaging was not available within the scope of the project a number of assumptions were made.
Thus this feasibility study has shown that it could be safe to use the Saksagan mine as a repository for low- and intermediate-level wastes. It complies with the current Ukrainian law on radioactive waste management.

The Saksagan mine provides particular favourable conditions for hosting a radioactive waste repository since it not only operated for mining iron ore but also for mining granite at considerable. These latter areas have been selected for the emplacement tunnels.
Open questions in development of disposal for long-lived waste are following:

- Uncertainties with long-lived waste inventory
- Poor knowledge about waste properties
- Necessity of improvement of current RAW classification
LLW-LL Disposal Facility

- Time required for containment requires safety options comparable to deep disposal
- Requirement for performance not as stringent as for deep disposal

François Besnus – IRSN, 2008
Thank you for attention