International Conference on the Safety of Radioactive Waste Management Conference ID: 50807
International Atomic Energy Agency Vienna, Austria 21-25 November, 2016

PROVEN TECHNOLOGIES FOR THE SOLIDIFICATION OF COMPLEX LIQUID RADIOACTIVE WASTE (LRW): GLOBAL CASE STUDIES OF APPLICATIONS AND DISPOSAL OPTIONS

Dennis Kelley
Pacific Nuclear Solutions, USA
Dr. Artem Gelis
Nuclear Engineering Division, Argonne National Laboratory, USA

Legacy radioactive waste streams from the Cold War still exist and newly generated waste streams from nuclear power plants and research institutes go untreated and expose environmental hazards at many nuclear sites. The nature of the waste is extremely diverse, depending upon the source or the process from which it originated. The most problematic waste streams include complex liquids such as organic (tri-butyl-phosphate TBP) solutions contaminated with Pu and U isotopes, mixed sludge types, high acid radioactive waste, tritium contaminated organic and aqueous streams, etc. Environmental and economic challenges exist for the treatment and disposal of such waste streams.

A proven technology that has been applied to LRW on a global basis provides a low cost solution to legacy streams and small volume, highly complex LRW frequently found during decommissioning. The engineered polymer technology from Nochar, USA, is capable of solidifying standard and highly complex LLW and ILW waste streams for interim or final storage, or for incineration. This paper examines various case studies involving the use of the polymer at several global nuclear sites.

Case Study No. 1: United Kingdom – Harwell and Sellafield Sites

In 2014, the Harwell Remote Handled-ILW Storage Tubes Water Recovery project commenced in three facilities known as the “tube stores” (Fig. 1). The legacy stores (7 meters in depth) contain contaminated ILW water which requires immobilization to meet the Geological Disposal Facilities, Letter of Compliance. Under Magnox management, a team was created to assess the problem and propose solutions. Nochar’s N960 polymer was selected to solidify the ILW water through perforated bags, then an encapsulation process with cement (Fig. 2). Final disposal is in 500 liter drums.

Fig. 1 Tube Stores

Fig. 2 Cement/Polymer Solidification

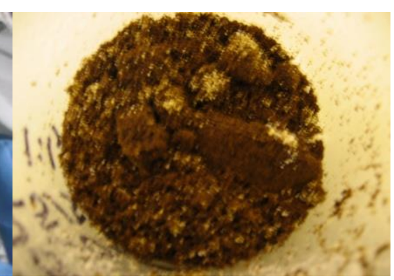

Fig. 3 Oil Solidification

Sellafield Decommissioning Characterization & Clearance group commenced an oil immobilization test program in 2006 with more than 90 oil sludge types in the oil waste storage facility. All of the oil sludge forms were uncharacterized and the experiments were undertaken on a small scale using 200 mL of oil.

Polymers N910 and N960 were applied to the oil and sludge forms at a 1.5 (liquid) to 1 (polymer) ratio (Fig. 3). Following solidification, a test program was conducted to encapsulate the waste form into an acceptable cement grout matrix for final disposal.

Case Study No. 2: France – Cadarache LOR (Liquides Organiques Radioactifs) Waste Streams

STMI-AREVA carried out a study to pre-treat waste streams in order to obtain an agreement with the French regulator, ANDRA, for final disposal. Two radioactive scintillation cocktails were legacy streams produced at Cadarache and stored in the CEREES cell and held in the ICPE 312 facility. The two streams were composed of organic compounds including xylene and TBP. Water content was about 80%. Radiological contaminants included: Pu-238, Pu-239, Pu-240, Am-241, Cs-137 and H-3.

Polymers N910 and 960 were combined with the streams to form a solidified matrix. The results indicate that the solidification matrix appears suitable for a final acceptance at ANDRA storage site. Additional test programs are currently underway by STMI-AREVA to validate the findings in this extensive research project.

Fig. 4 Aqueous / Organic -TBP

Fig. 5 Solidified Solution

Case Study No. 3: Romania – Cernavoda Nuclear Power Plant

Since 2008, MATE-FIN Co., based in Bucharest, has been providing a variety of technical services to Cernavoda NPP including the treatment and packaging of LRW. A series of organic waste streams have been generated by the CANDU 6 reactors that require treatment and disposal. These streams include spent oil with tritium, spent solvents, scintillation cocktails (LSC), flammable solids (solid-organic liquid mixture) and sludge. Polymer N910 is used to permanently solidify the hydrocarbon based liquids. N960 is used to immobilize any aqueous liquids including H-3. The solidification ratio is 2.5 : 1.

Large scale solidification production is conducted in steel drums, combining the LRW with the polymers, then using an electric mixer, for a slow, short-term mixing procedure (Fig. 6 & 7). Over 30 metric tons of oil have been solidified and shipped to Studsvik, Sweden for incineration. The N910 polymer is also applied to flammable solids (textiles, clothes, plastic materials) that are contaminated with tritiated oil. The solids are layered with the polymer for removal of the flammable liquids (Fig. 8).

Fig. 6 Oil with Tritium

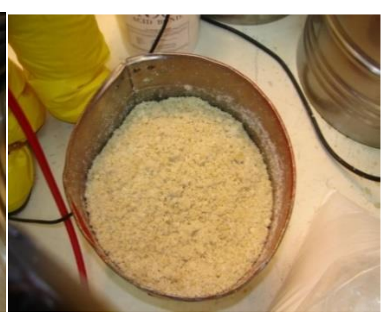

Fig. 7 Solidification Completed

Fig. 8 Treatment of Solids

Case Study No. 4: Russia – KRI Gatchyna and Mining Chemical Combine-Krasnoyarsk

The Khlopin Radium Institute, St. Petersburg, and the U.S. Department of Energy partnered under a three-year program to conduct experimental work with Nochar polymers. Tests included radiation and thermal stability using a Cobalt 60 gamma source irradiator, evaporation utilizing heat to evaluate the polymer's absorption capacity, gas generation on ILW waste forms, fire and safety, and cement encapsulation for final disposal. Over 250 real waste streams including a variety of organic and aqueous solutions were solidified with a particular focus on sodium nitrate solutions and high salt content solutions.

Irradiation tests (Cobalt 60 gamma source) proved the polymers stability with one test duration of 103 days for a total exposure of 27 million gray (Fig. 11). Gas generation tests verified that the polymers are not gas generators. In addition, experiments proved that the polymers maintain stability when combined with sodium nitrate solutions. In real applications, ILW waste was solidified at Gatchyna and packaged in a thick polyethylene bag, then encapsulated in cement (Fig. 9). The cement form was then packaged in polyethylene drums for final disposal at the RADON Sosnovy Bor site (Fig. 10).

Nochar polymer technology is the first foreign absorbent technology to be formally certified by ROSATOM, for use at Russian sites.

An extensive test program at the Mining Chemical Combine (MCC) near Krasnoyarsk was sponsored by the U.S. Department of Energy in 2009-2010. This experimental program evaluated the application of the polymers with a solution of TBP in hexachlorobutadiene (HCBD), a highly toxic waste stream. At a 5:1 ratio (N910), the solution was solidified and was capped by paraffin wax to prohibit gas emission.

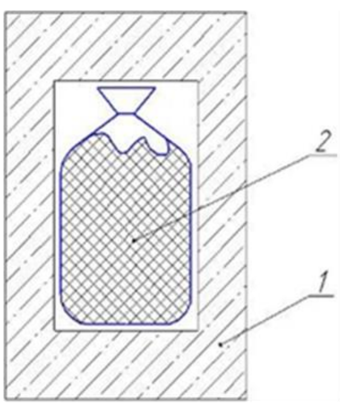

Fig. 9 Packaging Design

Fig. 10 Waste for Final Storage

Fig. 11 Irradiation at 770,000 Gray at 30 day's exposure

Case Study No. 5: United States – DOE Rocky Flats

Rocky Flats Technology Site was one of the first DOE nuclear weapons complex to undergo full decommissioning, with a final closure date of 2006. Critical to meeting the closure timeline was the treatment and removal of "orphan" liquid waste streams. These streams had no existing approved pathway for treatment or final disposal.

In 2000, the OASIS treatment system (cementation) failed to meet DOE criteria for transport and final disposal. Nochar's polymers were selected for application to the several thousand liters of transuranic (TRU) waste forms which included concentrations of plutonium. Three waste streams were treated:

- Methanol based solution with organic contaminants such as cyclohexane
- Mixed organic waste consisting of Freon, carbon tetrachloride and trichloroethylene
- Spent pump oil

All solidified waste was packaged in carbon steel 55 gallon drums and delivered to the Waste Isolation Pilot Plant (WIPP) for final storage. Cost savings to DOE for this project exceeded $ 10 million.

Case Study No. 6: Kazakhstan – BN-350 Fast Breeder Reactor, Aktau

The Mangystau Atomic Energy Combine (MAEC-Kazatomprom) located along the Caspian Sea closed operations in 1999. The BN-350 reactor was decommissioned with completion in 2010. Remaining on site are 3,217 cubic meters of liquid waste. Total activity of Cs-137 nuclide is 9,618 Ci. A test program to validate the effectiveness of the polymers was undertaken in 2014. Four waste streams were tested: machine oil, sodium hydroxide, and two saline solutions with high salt content. Solidification on all four waste streams was achieved by using Nochar's N910 polymer for oil and N960 polymer for the other streams. The solidification ratios were between 2:1 and 3:1 (liquid : polymer by weight). (Fig. 13, 14)

In addition to the solidification program, DOE and Kazakh scientists created an inexpensive method to dispose of the solid mass by encapsulating the machine oil / N910 into an inorganic filler and molecular sulfur material. After heating the entire mass in a short time, the mixture hardens and a solid waste form is generated. The organic waste content in the final form can reach up to 60 wt. % at a compressive strength of about 100 kg/cm² (Fig.12). If a harder form is required, then the waste content can be decreased to 25-30%, and the compressive strength will increase to 300 kg/cm².

Fig. 12 Samples of Sulfur Composite Waste Form Volume of Oil Used for Each Form – 30 ml

Fig. 13 Solidification

Fig. 14 Sodium hydroxide & N960 at 3:1 ratio

Conclusion

Nochar polymers have been used effectively in the global nuclear sector for over 17 years. The polymers are applied to all liquids including all hydrocarbon based products, all aqueous products including acid and alkaline, and light alcohols including ethanol and methanol. The solidified waste forms may be loaded directly into carbon or stainless steel drums, or encapsulated in cement for interim or final storage. The waste may also be incinerated.

Technical papers describing all of the above described case studies are available upon request.