ABSTRACT

AREVA is responsible for the treatment of about 10,000 m³ of legacy radioactive sludge (equivalent to 3,400 m³ of dry extract) produced by co-precipitation of radioactive liquid waste and currently stored at the La Hague site, France. This sludge is to be retrieved and packaged for permanent disposal in a deep repository. For many years, the baseline solution for packaging this waste has been to immobilize it in bitumen. Cementation has also been considered as a technically feasible alternative. However, the French safety authorities have determined that the risk of radiolysis in bitumen-immobilized sludge waste can no longer be accepted. Furthermore, even if the more stringent safety requirements that have emerged can be complied with, the poor loading rates achievable with these processes significantly increase the final waste volume, and thus the cost for disposal in a deep repository.

In response to the expectations of regulators and the inefficiencies inherent in current approaches to sludge packaging, a new baseline plan has been established. Therefore, the sludge is to be retrieved from existing storage silos in the form of slurry and collected in two large staging tanks. The slurry is to be homogenized to ensure consistency in the physical, chemical, and radiochemical characteristics of the waste batch, dried under prescribed process conditions, compressed into pellets, and finally transferred into qualified waste packages along with sand to fill void spaces.

Design requirements for this approach were challenging with a radioactivity level in the dried sludge expected to correspond to intermediate level waste [i.e. 60 GBq/kg of βγ activity and 3.2 GB/kg of α activity], requiring remotely operating and maintaining process equipment.

The design of the main process equipment for sludge drying and compacting is derived from commercial designs proven in chemical processing and pharmaceutical industry applications. Selection criteria also considered the feasibility and costs to “nuclearize” commercial equipment – that is, modify it as necessary to ensure dust confinement, mitigate potential for clogging, and provide for remote operation and maintenance.
The sludge drying technology selected for development consists of a vertical thin-film evaporator operating under vacuum conditions and inert atmosphere. The dried extract is fed to a pelletizing mill which then compacts the waste,

An extensive testing program using representative non-radioactive waste simulants has been performed to verify performance of planned unit operations under the range of process conditions expected to be encountered. Pilot-scale testing was successfully completed in June 2013 at the AREVA “Hall de Recherche de Beaumont” (HRB), while non-radioactive testing on a full-scale dryer is scheduled for December 2015, prior to deploying the technology in the nuclear facility.

Technologies developed for conditioning and packaging this legacy sludge are adaptable to other radioactive wastes. Lessons learned from this project demonstrate that pilot-scale tests can identify the key operating parameters necessary for reliable sludge drying and compacting operations.

INTRODUCTION

Located at La Hague, UP2-400 (see Fig. 1) was AREVA’s first commercial used fuel recycling plant. Between 1966 and 1998, UP2-400 processed nearly 5,000 tons of natural uranium used fuel from graphite-gas power reactors, 4,500 tons of fuel from pressurized water reactors, and a few tons from fast breeder reactors, as well as research reactors. UP2-400 was shut down in late 2003 and replaced with two new plants: UP2-800 and UP3.

The decommissioning project was launched in 2009 and will be conducted over a 25-year period. This is a large D&D project with significant legacy radioactive material to retrieve (hulls, end pieces, spent ion exchange resins, sludge, etc.) prior to starting the actual dismantling operations. There is about 32,000 m³ of waste to remove and process. Among them, AREVA is responsible for the retrieval, treatment, and packaging of approximately 10,000 m³ of radioactive sludge (representing 3,400 metric tons of dry salts) from storage tanks in the STE2 Facility. STE2 was the effluent treatment facility of UP2-400 where the liquid waste was decontaminated using a chemical co-precipitation process. At the beginning of the operation of UP2-400, before the adoption of an on-line bituminization process to immobilize the
radioactive co-precipitation sludge, the sludge was stored in several concrete-wall silos. This sludge is to be retrieved and packaged for permanent disposal in a deep repository.

**CHALLENGES**

The baseline solution used for packaging this waste has been to immobilize it in bitumen. Cementation has also been considered as a technically feasible alternative. However, the French safety authorities have determined that the risk of radiolysis in bitumen-immobilized sludge waste can no longer be accepted. Furthermore, even if the more stringent safety requirements that have emerged can be complied with, the poor loading rates achievable with these processes significantly increase the waste volume, and thus the cost for disposal in a deep repository.

In response to the expectations of regulators and the inefficiencies inherent in current approaches to sludge packaging, AREVA engineers and scientists re-examined requirements and technical alternatives for legacy sludge conditioning. These studies provided the basis for establishing a new baseline plan – condition the sludge by drying, pelletizing the dried extract, and filling packages with waste pellets surrounded by a sand matrix. A waste volume reduction factor of 2 to 3 is expected with this process.

Design requirements for this approach were also challenging:

- radioactivity in the dried sludge is expected to correspond to intermediate level waste [i.e. 60 GBq/kg of $\beta\gamma$ activity and 3.2 GB/kg of $\alpha$ activity], this requires remotely operating and maintaining the process equipment;
- drying and compacting operations must not alter the chemical characteristics of the sludge;
- residual water in the package should be as low as possible to minimize hydrogen gas that may be produced by radiolysis;
- safety considerations warrant minimizing the hold-up of waste within processing equipment and confinement of dust resulting from conditioning operations; and,
- the number and size of waste packages required to dispose of the sludge, the costs of construction, operation and D&D for conditioning and packaging it, and the volume of secondary waste produced should all be minimized while ensuring full compliance with requirements for safety and environmental protection.

**DESCRIPTION OF THE PROCESS**

In order to ensure performance objectives are met, the design of the main process equipment for sludge drying and compacting were derived from commercial designs proven in chemical processing and pharmaceutical industry applications. Selection criteria also considered the feasibility and costs to “nuclearize” commercial equipment – that is, modify it as necessary to contain dust, mitigate potential for clogging, and provide for remote operation and maintenance.

The sludge drying technology selected for development consists of a vertical thin-film evaporator operating under vacuum conditions and inert atmosphere. The dried extract is fed to a pelletizing mill which then compacts the waste, delivering a final form that can be managed with simple mechanical handling equipment.
The Process Diagram is shown in Figure 2. The sludge at an approximately 190 g/L total salts concentration is fed to each dryer at a rate of 250 L/h and the dryer rotor provides a spiral path for the fed material in an annular space as the sludge proceeds by gravity to the output buffer vessel. The outside of the dryer consists of a heating jacket (steam) to dry the sludge at a temperature lower than 200°C and under vacuum. Vapors are extracted by a vacuum pump through a candle filter, a condenser, a demister and filters. The candle filter traps particles entrained with vapors and periodical unclogging is performed to allow the entrained powder to flow back to the dryer bottom through a dedicated pipe. Dry powder is pressed into pellets, which are received into a stainless steel drum called C5. Sand is added into the drum to fill the voids, and then the drum cover is welded.

All equipment implemented in this process is designed for installation in hot cells, and subject to remote operation and maintenance. All equipment is designed to have a reliability/availability consistent with a Total Operating Efficiency target of 70%.

Fig. 2. La Hague Sludge Drying and Pressing Process Diagram

**Drying process**

The drying process is based upon the vertical thin-film drying technology described in Fig. 3.
Fig. 3. Drying Process Diagram

To support the project, this drying technology to be applied for STE2 sludge processing has been extensively tested at the AREVA Beaumont Testing Facility (HRB) at La Hague. There, a 30 L/h nominal flow rate pilot-scale system (see Figures 4 and 5) has been - and is currently - operated in support of the design studies.

Fig. 4. Pilot-Scale Dryer at HRB (top view)
AREVA has performed extensive tests on this engineering-scale pilot dryer (1/10th of the planned throughput) implementing the vertical thin-film drying technology. All tests performed in the Beaumont Testing Facility (HRB) pilot system use non-radioactive simulated sludge, representative of STE2 sludge. Along the history of AREVA La Hague UP2 Plant, STE2 sludge has been produced by several co-precipitation decontamination processes of liquid waste.

The non-radioactive simulated sludge covers a range of:

- Chemical compositions (two sludge families are tested: calcium carbonate based sludge and barium sulfate based sludge – called “baseline” slurries),
- Total salts concentration (120 to 200 g/L) as well as soluble salts fraction (2% to 28% of total salts),
- Aging (freshly prepared sludge and 20 year-old sludge produced during cold start-up tests of the STE3 Effluent Treatment Facility in UP3 Plant, which implements a similar co-precipitation process).

The HRB pilot system has also supported the investigation of a wide range of operating conditions for the dryer, such as variations in the rotor rotation speed; heating medium temperature; and sludge feed flow rate.

This pilot-scale system has achieved the following goals:

- Determine initial water evaporation capability,
- Determine nominal operating conditions for two kinds of non-radioactive simulated sludge: (“baseline” slurries),
- Determine the heat transfer coefficient between stator and evaporating sludge; and perform thermal balance,
• Implement and test devices to develop an appropriate powder discharge system; an appropriate rinsing procedure; an appropriate unclogging procedure; and an appropriate off-gas treatment system (candle filter),
• Evaluate the influence of sludge characteristics variations on drying efficiency in nominal operating conditions; and investigate potential adjustments of operating conditions to maintain drying efficiency,
• Validate the increase in total salts concentration in the sludge feed,
• Perform 100-hour continuous runs for “baseline” slurries,
• Investigate means for powder dryness measurement, and
• Investigate wear issues and potential solutions.

This scaled testing demonstrated that more than 95% of the liquid in sludge can be removed with this technology. Pilot-scale testing was successfully completed in June 2013. This program will be followed by non-radioactive testing with a full-scale dryer planned at HRB from December 2015, prior to deploying the technology in the nuclear facility.

Dried sludge pressing process (Pelletization)

The dried sludge pressing process uses mechanical presses with eccentric gear. To support the project, this pressing technology to be applied for STE2 sludge processing has been tested by a Vendor which exhibits extensive experience in Research and Development (R&D) of pharmaceutical presses. Tests enabled to determine pressing parameters and pellets characteristics for the two STE2 sludge simulants (“baseline” slurries) previously dried in the HRB pilot-scale system.

Figure 6 shows the supplier’s R&D press, which was used for the above-mentioned tests.
CONCLUSION

The Detail Design phase has been completed and the project is currently in early procurement specification phase.

This new sludge drying and packaging process will be established in the La Hague’s STE3 process building. This building currently houses the now-obsolete bituminization process, so construction of the new system will start once the existing process is dismantled. Initial preparations within the host facility began in late 2013, while construction of the new process will start in 2014 for a commissioning scheduled in 2017. The sludge drying system is expected to be fed with radioactive material by 2018.

Technologies developed for conditioning and packaging this legacy sludge are adaptable to other radioactive wastes. Lessons learned from this project show pilot-scale tests can identify the key operating parameters necessary for reliable sludge drying and compacting operations.

For example, this process could be favorably used to treat the contact-handled transuranic (CH-TRU) waste stored in eleven tanks at the Hanford B and T Tank Farms to produce a waste form that meets the Waste Isolation Pilot Plant (WIPP) waste acceptance criteria. Indeed, lab-scale testing performed by AREVA at HRB using a non-radioactive Hanford CH-TRU waste simulant showed very promising results.