### Radioactive Spent Ion-Exchange Resins Conditioning by the Hot Supercompaction Process at Tihange NPP – 10043

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# ABSTRACT

Besides safety precautions, the immobilization of spent ion-exchange resins requires special treatment and conditioning techniques to meet the acceptance criteria for disposal. Waste acceptance criteria define, among others, the quality of waste forms for disposal, and therefore will sometimes orient the choice of a specific process. In Belgium, for economical reasons, the Volume Reduction Factor is a key criterion.

After Tractebel Engineering performed a technical and economical comparison of the industrially available systems, Tihange NPP decided to install a spent ion-exchange resins hot supercompaction unit. Tractebel Engineering is in charge of the project management and takes an active part in the process optimization.

The treatment and conditioning unit processes the spent ion-exchange resins through the following steps: dewatering of the resins, drying the resins under deep vacuum with a binding additive, discharging the dried mixture of resins and additive into compactable drums, supercompacting the drums to generate pellets, grouting the pellets into standard 400 litres waste drums (overpacks) licensed for final disposal in the near-surface repository in Belgium.

In order to avoid cracks on the compacted drum, and external surface contamination from resin leaks, some improvement in additive selection and drum design has been made. Powdered cationic ion-exchange resins (as used for BWR condensate polishing) were found to minimize springback as well as the risk of cracking the drum wall better than common polymer powders.

Placing the compactable drum inside a second, slightly larger drum, also guarantees clean and reproducible pellets with a powder additive proportion limited to 25% of the total waste volume. Without this safety wrapping, the additive proportion must be raised to at least 30%.

With this process, the final matrix inside the pellets is a water free solid, but somewhat brittle, block. Volume Reduction Factors between 1.8 and 1.9 are achieved. Special care must be taken about the mortar quality if pellets are to be grouted.

## **INTRODUCTION**

Ion exchange is one of the most common and effective treatment methods for liquid radioactive waste. Spent ion-exchange resins are considered to be problematic waste that, in many cases, requires special approaches and precautions during their conditioning to meet the acceptance criteria for disposal.

With the evolution of disposal facility acceptance criteria, it is now required that spent ion-exchange resins meet specific quality requirements prior to disposal. Where final disposal facilities exist, waste acceptance criteria define, among others, the quality of waste forms for disposal, and therefore will sometimes define appropriate treatment options. For example, disposal facilities normally define acceptable levels of free liquids and requirements for waste form stability as part of their waste acceptance criteria.

The selection of treatment options for spent ion-exchange resins must consider their physical, chemical and radiological characteristics. Basically, two of the main methods for the treatment of spent organic ion-exchange resins, following pre-treatment methods like dewatering, grinding, foaming or decontamination by activity stripping are [1]:

- Direct immobilization, producing a stable end product by using cement, bitumen, polymer or high integrity containers;
- The complete removal of the resin inner structural water by a thermal process followed by the supercompaction of the hot dried resins.

Part one of this paper will describe the principle of a reference Resins Hot Compaction process. Part two of the paper will introduce the application of this process at Tihange NPP (Belgium) operating PWRs.

# **REFERENCE RESINS HOT COMPACTION PROCESS**

### Type of spent resins to be conditioned

Hot compaction has been developed initially for a reference NPP operating two types of reactors and producing two types of spent resins:

- One Boiling Water Reactor (BWR) producing mostly spent powdered resins (condensate polishing);
- One Pressurized Water Reactor (PWR) generating bead resins.

The waste to process in the reference plant is a mixture of spent powdered resins (75–90%) and bead resins (10-25%).

#### Selection of the conditioning process

In an environment without final disposal options and limited interim storage capacity, volume reduction is an essential criterion, leading to the choice of hot compaction.

#### **Treatment process**

Spent ion-exchange resins are first dewatered by a centrifuge (separator and decanter) system and filled in intermediate drums. After dewatering, the residual internal structural water inventory is about 50-60% of the resin total weight.

Intermediate drums will be transferred to a thermal-oil heated, vacuum drying vessel where resins are poured by means of a special docking device.

This drying vessel can also be used for mixing bead and powdered resins. In the drying and mixing unit, resins are heated to the necessary process temperature. After meeting the drying criteria, resins are discharged into special compactable steel drums, on which a lid is automatically placed. After these operations, the drums are immediately transferred to a high force compactor. Resulting pellets (compacted drums) are routed to a measuring unit, where the dose rate, height and weight are automatically measured and recorded.

For the operation of the waste conditioning processes, two operators are needed. Additionally one operator is needed for the necessary crane activities during transport to the interim drum storage area as well as filling the overpack with the resulting pellets.

The dried hot resin supercompaction unit (see Fig. 1) is remotely operated by a PLC (Program Logic Controller) system. There is thus no operator radiological exposure resulting from the unit operation.



Fig. 1: Resins hot supercompaction installation for Tihange NPP.

# **Process performances**

The Volume Reduction Factor (VRF) of the Hot Compaction process is very good:

$$VRF = \frac{\text{Dewatered resins volume}}{\text{Volume of produced pellets}} \approx \frac{2.5}{2.5}$$
 (Eq. 1)

The overall VRF depends on the overpack type.

Some 3800 satisfactory pellets have been produced by the reference Hot Compaction plant [2].

The advantage of the process is that products suitable for final disposal will be generated and, at the same time, an important volume reduction for interim storage will be achieved. Moreover, with respect to the retrievability, the pellets can be very easily retrieved from the overpacks, or the overpacks themselves can be placed in larger final disposal packages.

# SPENT RESIN HOT COMPACTION PROCESS AT TIHANGE NPP (BELGIUM)

# Spent resins quantity and type

Tihange NPP, located in Belgium on the banks of the Meuse River, operates 3 PWRs with a power of about 1000 MWe each. Spent ion-exchange bead resins are 640  $\mu$ m (mean wet particle size) and the average production amounts to some 8 m<sup>3</sup>/year.

## Selection of the conditioning process

Until 2005, these ion-exchange resins were immobilized in an organic matrix (styrol, epoxy based materials) by use of a mobile unit provided by an external service supply company. The resins are immobilized in ONDRAF/NIRAS<sup>1</sup> licensed overpacks (standard 400 litres drums), on the basis of one campaign (30-36 m<sup>3</sup> of resins) every four years. The process overall VRF was found to lay in the range:

$$VRF = \frac{\text{Dewatered resins volume}}{\text{Volume of conditioned 400 litres drums}} \approx \underline{0,5}.$$
 (Eq. 2)

Due to concerns linked to the high cost of the process, and to fire hazards, Tihange NPP requested from Tractebel Engineering in 2005 a complete survey and reassessment of the currently available and industrially proven spent ion-exchange resins conditioning processes with, as final aim, the recommendation of the best suited process taking into account the internal/external constraints prevailing at the plant.

Each surveyed process was assessed in accordance with the 6 following criteria:

- Overall cost including, as appropriate: investment, consumables, operation/maintenance, secondary waste management, process qualification, management of the conditioned waste packages (transportation, interim storage, final disposal), dismantling (in case of a new fixed installation);
- Autonomy (fixed installation) versus dependence (mobile installation operated by external service companies);
- Manpower requested qualification for the process implementation;
- Qualification of the process, i.e. compliance of the end product with the ONDRAF/NIRAS Waste Acceptance Criteria (WAC);
- Nuclear and industrial safety;
- Industrial references.

For each process, each criterion was ranked from 1 (lowest) to 6 (best). A weighing factor was then attributed to each criterion, enabling to end up with an optimized proposal. Sensitivity analysis, including variations of the weighing factor numerical values, enabled the robustness of each process to be assessed against uncertainties.

This multi-criteria analysis result recommended the installation of a fixed dried resin Hot Compaction unit, i.e. the by far best ranked process, under the constraints prevailing at Tihange NPP.

# **Process modifications**

Tractebel Engineering and contractor Hansa Projekt Anlagentechnik (HPA) contributed both to set up and optimize the design of the new plant.

With respect to the reference process, several evolutions have been developed in order to meet the constraints and standards at Tihange NPP. Solutions for concerns about springback, dust releases and grouting are described hereunder.

The most significant difference between Tihange NPP and the reference plant is the absence of powdered resins in the Tihange waste.

<sup>&</sup>lt;sup>1</sup> ONDRAF/NIRAS: Belgian waste management agency, in charge of the collection, the conditioning and the final disposal of the radioactive waste produced in Belgium.

A major impact of this absence is the excessive springback of the produced pellets. Full scale trials with inactive resin beads only led to following observations:

- Endothermic decomposition of anion exchange functional groups is responsible for increasing the heating time and limiting the product temperature at 120 130°C;
- At these temperatures the resins chemical matrix remains basically stable: the material remains elastic within the range of deformation caused by compaction forces;
- Consequently deformed spherical beads tend to recover their initial shape, creating an important springback of the pellets exiting the supercompactor;
- Springback exceeding 20% of the pellet height is responsible for serious damages: cracks in the metal wall, detached lid, surface contamination (see Fig. 2).

However, hot compaction of mixed powder and bead resins in the reference plant does not give rise to detrimental springback, indicating that powdered resins play a key role in the process. A powdered additive is required to fill the gaps between beads, prevent excessive deformation and elastic springback, and bind both products together into a cohesive matrix.

Several thermoplastic powders were tested (polypropylene, polyethylene, polystyrene), in proportions between 15 and 25% of the wet bead resin volume, which was the estimated void ratio between the beads. Unfortunately the natural elasticity of these plastics was found to be too high to get a sufficient reduction of the springback effect after compaction. The produced pellets were also featured by random cracks and surface contamination. Moreover these polymers used as conditioning additive gave rise to concerns about radiolytic gas releases (H<sub>2</sub>).



(A)

(B)

Fig. 2: Springback effect after supercompaction of bead resins alone (A) or mixed with thermoplastics (B).

In order to decrease the springback and to ensure the production of stable, reproducible, contamination free pellets, two options were successfully investigated (see Fig. 3):

• Additive option: the same product as in the reference process was tested. Powdered ion-exchange resins (polystyrene - divinylbenzene based) in a proportion of 25-30% of the total mixed volume, provides a much lower springback, and finally satisfactory pellets. An interpretation of this successful difference is that ion exchanging functional groups seem to make polymers less compressible. Cationic powdered resins only are used because of their better thermal stability. The final matrix inside the pellets is a water free solid, but somewhat brittle, block;

• Double drum option: placing the compactable drum inside a second, slightly larger drum, appears to provide clean, reproducible pellets, with a powdered additive proportion limited to 25% of the total waste volume. Without this safety wrapping, the additive proportion must be raised to at least 30% (lower VRF).



(A)

(B)

Fig. 3: (A) Double drum: the external drum is smaller in height, slightly larger in diameter, and ensures an efficient protection of the internal drum - (B) The addition of powdered ion-exchange resins and the use of a double drum design reduce the pellet springback when the piston of the supercompactor moves up.

It must be pointed out that powdered resins, once dried, become a very thin dust (particle size  $\sim 50 \ \mu m$ ) which can be somewhat radioactive after blending with spent beads inside the dryer. Hence, the following minimum precautions are mandatory to ensure the system operation is safe:

- Good docking of the compactable drum below the conical dryer when the dried mixture is discharged;
- Confinement of the discharge (drumming) and capping area, with ventilation;
- Assessment of dust explosion risks and subsequent preventive & protective measures.

According to waste acceptance criteria related to heterogeneous radioactive waste in Belgium, pellets containing dried, hot compacted resins must be inserted into standard 400 litres drums (licensed overpack – see Fig. 4), cooled down and grouted.

Grouting tests have shown that the mortar recipe has a great influence on the compliance with waste acceptance criteria. Indeed, traditional fresh liquid mortar is often featured by the segregation of its components, and the short term existence of a watery phase is likely to infiltrate pellets, to create free liquid pockets and to generate risks of resin swelling. A specific, ready made, premixed mortar (as used in sealing applications) was found to perform good grouting without seeping into the dried resin pellets.

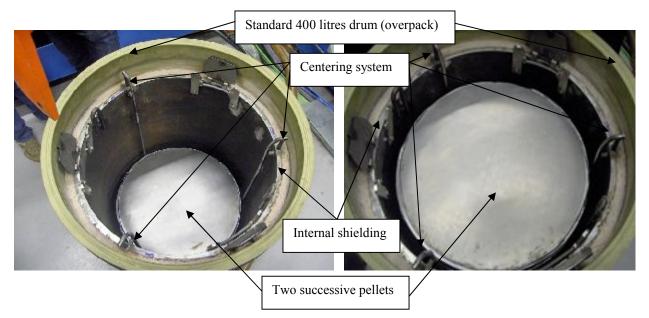


Fig. 4: Two pellets, with outer diameter of 55 cm and average height of 40 cm each, are inserted inside the standard 400 litres drum overpack with internal shielding and centering system. The pellets must still be immobilized by grouting before interim storage.

### **Equipment modifications**

The main component used in the application of the Hot Compaction Process for Tihange NPP is a 450 litre vacuum conical dryer with mixing screw and thermal oil heating, as well as a 2 000 tons supercompactor like in the reference plant. The following equipment modifications have been implemented at Tihange NPP:

## Resin feeding and dewatering

Tihange NPP spent resins are stored in tanks and will not be dewatered nor filled in drums prior to treatment. Actually, resin storage tanks are located above the conical dryer, so resins can be routed by gravity into the dryer. Dewatering is performed by a special device directly inside the drying vessel, prior to starting the drying operation.

#### Additive dosing device

Based on test trials for the process, a dosing device has been added to the system for dosing the required volume of additive (powdered cationic ion-exchange resins) needed into the drying vessel for mixing with waste resin beads before heating and drying the mixture.

#### Optimized overpack filling system

The pellets resulting from the Resins Hot Compaction Process are regular in terms of height and weight. In order to guarantee an optimized filling grade of the final overpacks, a selection turntable for pellet buffer storage has been added to the system.

#### Confinement housings

Confinement housings under negative pressure, with decontamination facilities, are provided around the dry resin discharge and lid capping area, and around the compaction area.

# Selection of supercompactor

The overall duration of the resin conditioning campaigns and installation maintenance activities will be shorter than 2 months per year. Therefore, in addition to resin conditioning, the new compaction system can process other solid waste streams. A 2000 tons supercompactor was selected for this reason.

# **Process Performances**

Dried resins are discharged into compactable drums (capacity 190 litres) which, after receiving a lid, are supercompacted. The Volume Reduction Factor (VRF) of the hot compaction process is given by:

	$VRF = $ Dewatered resin volume $\approx 1.9$	. (Eq. 3)
ľ	Volume of produced pellets $\sim \frac{1.9}{1.9}$	

Due to the need to add a limited quantity of powdered resins, which is not part of the primary waste, this value is lower than that achieved in the reference plant.

Pellets are then piled up into the standard 400 litres drums licensed for final disposal. The void volume between the pellets and the drum inner walls is filled with grouting mortar. The overall Volume Reduction Factor (VRF) is given by:

$$VRF = \frac{\text{Dewatered resin volume}}{\text{Volume of conditioned 400 litres drums}} \approx 0.95$$
 (Eq. 4)

This value is much larger than that obtained with immobilization in cement or organic matrixes.

Tihange NPP treatment and conditioning system can process 380 litres of spent bead resins per day with annual campaigns lasting about 22 days. The layout requirements of the entire treatment and conditioning system are:

- Ground floor surface area : 13 m x 7.5 m;
- Elevation (max): 6.2 m.

# CONCLUSIONS

More than 3 800 drums of mixed powdered and bead resins have been processed by the reference Hot Compaction process, achieving a Volume Reduction Factor (VRF) of 2.5. The equipment has been proven to be a reliable technology with low operation and maintenance costs.

Tractebel Engineering has initiated and is managing the construction of a new application of this process in Belgium at Tihange NPP. Several developments were required to adapt the reference process and equipment to PWR spent ion-exchange bead resins and Belgian radioactive waste acceptance criteria.

In an environment of very limited space for interim storage and in the absence of an operating final repository site, or in the case of high final disposal costs, the process exhibits the following key advantages:

- Achieving a Volume Reduction Factor (VRF) close to 1 (overpack included) for the interim storage instead of increased volumes observed with other currently available processes;
- Achieving a water free end product;
- Creating a flexible waste product for interim storage (pellet), which can be retrieved and routed into alternative types of package later, if not initially grouted;

- Using well proven standard technologies like drying and compaction;
- Flexible use of the system components for the supercompaction of other operational solid waste streams when not conducting resins conditioning campaigns.

### REFERENCES

- [1] E. Grundke, H.-D. Harass, "Conditioning Radioactive Waste at the Point of Origin", KONTEC'93 Conference Transcript, Hamburg (18-19 March 1993).
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