#### MDS: A PROVEN AND VERSATILE SOLVENT MINERALIZATION PROCESS

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### ABSTRACT

The Solvent Mineralization facility (MDS) at the La Hague site in France has been designed to condition spent solvent distillation residues as a solid waste. Active start-up of the facility is described and the results of the first solvent mineralization campaigns are presented.

Since entering service in early 1998, the MDS facility has demonstrated the feasibility of operating a facility to condition reprocessing solvent residues in a stabilized form approved by waste disposal authorities, while generating only low releases with negligible radioactivity. Application of the MDS process to the disposal of solvent backlogs at nuclear production or research centers is discussed.

### INTRODUCTION

Cogema operates the world largest nuclear fuel reprocessing center at La Hague, France. The center's two reprocessing plants, UP3 and UP2-800, hold the world record for reprocessing LWR fuels. The plants are also committed to managing all their waste, especially by volume minimization and conditioning. The bulk of fission products is now confined by vitrification, leaving the focus on less active waste such as the spent solvent which was not successfully managed in most previous plants. This issue has now been resolved at the La Hague site, thanks to the MDS facility built by SGN for Cogema. The MDS is the newest facility in operation on the site.

The MDS facility, in conjunction with upstream solvent treatment units, produces solid waste suitable for near-surface disposal, in accordance with Cogema's policy of reducing solid waste ready for deep underground disposal. Cogema also has a waste processing inline policy.

The OWT (Organic Waste Treatment by distillation) facilities for UP3 and UP2-800 have dramatically decreased the amount of spent solvent produced per ton of fuel processed, as well as its radionuclides content, compared with other plants using more conventional solvent management systems [1] [2]. The distillation process produces a small flow of residue which used to build up in interim storage tanks. Thanks to the MDS facility, this spent solvent stockpile will now decrease and ultimately disappear.

The process development and related R&D have already been described [3]. This paper briefly describes the process and focuses on the active start-up of the facility, initial operating results and application of the process to spent solvent in various facilities.

# PROCESS DESCRIPTION

The process is illustrated in Figure 1.



Figure 1 - MDS process diagram.

# **Feed Preparation**

The first step is the preparation of the feed. Initially, the distillation residue (typical composition TBP > 90%;  $\beta\gamma$  emitters: 7 MBq/l;  $\alpha$  emitters: 74 kBq/l) is transferred to an adjustment tank, where it is mixed with oil or diluent if there is any to process. The final

grade must be more than 60% TBP up to about pure TBP. In another stirred vessel, the magnesia is mixed with the emulsifier and the spent solvent. The emulsion is then transferred to the feed tank. The vessels are dimensioned to perform a preparation only once a week.

#### **Pyrolysis Reactor**

The emulsion is fed at a design flowrate of 3 kg/h of TBP to a stirred pebble ball pyrolysis reactor (Figure 2) through two nozzles. The reactor bed is kept at high temperature by heaters on the walls of the reactor. The balls are made of carbon steel. To prevent any reaction with air, the reactor is filled with nitrogen.

Contact of the emulsion with the heated balls causes the water and the diluent to evaporate, and the TBP to react with magnesium hydroxide, yielding butene, some butanol and various magnesium phosphates. Ultimately, the feed is split into a gaseous stream (steam, nitrogen, butene, butanol and dodecane vapors) and a solid stream (ashes which are a mixture of magnesium oxide and phosphates). The ashes fall into the funnel, then are admitted into a hopper. In order to prevent any air intake into the funnel, the ashes are transferred to the hopper through an airlock. The hopper is cooled, to bring the ashes to room temperature.

When enough cooled ashes are accumulated in the hopper, they are transferred into a mixer and mixed with concrete and batch grouting is performed. The resulting grout is poured into drums.



Figure 2 - Pyrolysis reactor - Overall height: 3.5 m; Diameter: 1.1 m.

# Gas Treatment

The pyrolysis vapors are routed through high efficiency filters to separate the off-gases from the fine dust of magnesium phosphates and magnesia. This filtration also prevents the small amount of radionuclides in the spent solvent from being removed with the off-gases. The filtration unit is operated at high temperature to prevent hydrocarbons from condensing and clogging the filters, which are arranged as a cluster of candle filters. Countercurrent declogging is performed periodically using nitrogen blasts. After filtration, the vapors are sent to a combustion chamber. A diluent (or fuel) burner provides a pilot flame and permits the safe incineration of the vapors. The combustion off-gases are scrubbed, filtered and released to the stack.

### Layout

The MDS facility was built near the STE 3 building to benefit from the proximity of the spent solvent tanks and from the utility networks, in an existing building.

# PLANT START-UP

### Cold Testing

Cold tests were performed on the facility from the end of 1995 up to May 1997, with the following objectives:

- fine-tuning the components;
- adjusting the procedures;
- preparing the facility to be integrated in an industrial nuclear site with special constraints.

The tests were performed initially in parts and, at the end, by integrated testing which quickly demonstrated a high degree of reliability. Some time was devoted to testing the effects of the quality of reagents.

The tests confirmed the R&D results, which had indicated that selection of reagents with specific physical and chemical characteristics was crucial for proper operation of the facility.

### Active Operation

The MDS facility was commissioned on January 13, 1998. Active start-up was scheduled to begin at half the maximum throughput (3 kg/h of emulsion) for a certain time, to test performance in active conditions, and to collect operating experience before increasing the feed-rate in steps up to the design capacity. The first campaign lasted approximately 40 days, i. e., 950 hours. The initial load was prepared by using distillation residues of the OWT facility of UP3. Results of the first campaign were very satisfying.

At the end of the first campaign, an inspection was performed by opening the reactor with contact maintenance techniques planned for the facility, including the use of vinyl airlocks, with the cooperation of the health physics department. The results confirmed that contact maintenance is valid because of the extremely low level of irradiation. The inspection showed that the reactor was in excellent condition.

The facility was prepared for service and re-started at the beginning of April 1998, with the objectives of increasing throughput to reach the design capacity and to qualify certain magnesia selected for their high specified quality, in order to diversify the supply of reliable industrial-grade reagents.

In 1999, the objectives were to sustain the design feedrate for long periods of time. This required careful elimination of minor sources of shutdown (regulation defects, sensor failures, and so on). In the first half of 1999, long runs were performed with an emulsion feed flowrate of 6 kg/hour. The target has been met and the facility has since been operating normally.

### OUTLOOK

Thanks to the MDS facility, spent solvent is now fully processed at the La Hague site to produce grouted solid waste suitable for near-surface disposal in compliance with Cogema's policy of in-line processing of all the waste produced by spent fuel reprocessing. The years of R&D performed on the mineralization process to make it suitable for nuclear applications, and the months of tests performed on the site, have been rewarding. The facility is operating smoothly and has been integrated into the rest of the complex at La Hague.

The MDS process has been selected by Belgoprocess and adapted to contaminated solvent with a lower TBP content than the La Hague distillation residues. Belgoprocess has completed inactive testing of its facility using the process.

Because it enables conditioning of waste in concrete or other matrixes for permanent disposal, the MDS process is an attractive solution for all plants that previously lacked a proven technique for eliminating contaminated solvent stockpiles. The contact maintenance concept contributes to MDS cost effectiveness and should be maintained if the process is applied to significantly contaminated solvent. Cogema's experience in chemical decontamination of solvents containing radioactive fission products indicates that prior decontamination is the best way to ensure the cost effectiveness of the operation.

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