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ENVIROBOND™/ENVIROBRIC™ CHEMICAL STABILIZATION PROCESS TREATMENT OF SILO 3 URANIUM-THORIUM CALCINED WASTE AT THE DOE FERNALD

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ABSTRACT

Rocky Mountain Remediation Services, L.L.C. (RMRS), was selected by Fluor Daniel Fernald (FDF) to stabilize 5,100 cubic yards of uranium-thorium calcined waste material with high concentrations of heavy metals currently stored in Silo 3 at the Department of Energy's Fernald Site. The calcining process that generated the waste produced a finely powdered metal oxide that is difficult to stabilize using traditional cementation or vitrification methods.

RMRS will use a vacuum system to retrieve the material from the silo, and the company's proprietary Envirobond™/Envirobric™ process to stabilize and volume reduce the waste to pass TCLP limits while obtaining a 40 percent volume reduction. The Envirobond™ process is an enhanced chemical stabilization process that allows for small volumes of reagents to bind and stabilize heavy metals. Envirobond™ does not bulk the treated material as much as traditional stabilization techniques, but rather acts as a binder so the material can be compressed into a brick form called Envirobric™. This reduces the treated material's volume, thus reducing overall life cycle costs.

This paper describes the applicability of the Envirobond™/Envirobric™ process to the Silo 3 material by presenting laboratory treatability results and relevant portions of the conceptual design for the project. The Envirobond™/Envirobric™ process will produce a waste form that meets the Fernald criteria for stabilization of heavy metals, no free liquids in packaged waste, and absence of fine particulate matter.

INTRODUCTION

Silo 3 waste at the Department of Energy's (DOE) Fernald Site contains approximately 5,100 cubic yards of uranium-thorium contaminated waste that accumulated during uranium extraction operations in the 1950's. The predominant radionuclide within Silo 3 is Thorium-230. Silo 3 material includes the RCRA regulated metals, arsenic, cadmium, chromium, and selenium. Although Silo 3 material is classified from the former Atomic Energy Commission designation as 11 (e) (2) material and is therefore not regulated by RCRA, the material is considered sufficiently similar to hazardous waste under CERCLA guidelines, and some RCRA requirements are relevant and appropriate.

RMRS will use a vacuum system to retrieve the material from the silo, and the Envirobond™/Envirobric™ process to stabilize the waste. The treated waste is compacted into a shippable brick that passes RCRA Toxic Characteristic Leaching Procedure (TCLP) limits and reduces the volume of the waste by more than 40 percent volume. The process uses a non-hazardous chemical binder called Envirobond™ that chemically binds with metal contaminants, preventing leaching under the most stringent conditions. This chemical stabilization forms a new compound with extremely low solubility, that incorporates the heavy metals into the chemical structure.

The effectiveness of the Envirobond™ process has been previously proven with mining waste and mill tailings. In addition to meeting the Environmental Protection Agency (EPA) standards for TCLP, the results of the TCLP testing have typically met the more stringent Universal Treatment Standards (UTS). Less than 4 wt. %, Envirobond™ reagents and additives are added to the waste using equipment that is readily available from commercial vendors. Costs are typically lower than cementation, vitrification, and other solidification methods. With cementation, the additional bulking of the waste can easily add a 50% volume increase, and the solidified waste can be difficult to package and transport. Often, the cost of materials is higher and the mixing requirements are more difficult.

The following two sections discuss the results from a recent treatability study that was done using samples of Silo 3 waste and present the proposed design information for treating Silo 3 material.

LABORATORY TREATABILITY RESULTS

The objective of the laboratory treatability study was to show that Silo 3 material treated with the Envirobond™ process will meet or exceed all Fernald Silo 3 waste acceptance criteria (WAC). These three criteria include the stabilization of heavy metals, no presence of free liquid, and absence of fine particulate matter. The attainment of these minimum standards was demonstrated during the treatability testing.

- Toxic Characteristic Leach Procedure (TCLP) results for all eight RCRA metals, (arsenic, barium, cadmium, chromium, lead, mercury, selenium, silver), were found to be below the RCRA standard after treatment with the Envirobond™ process. Nickel, which is also present at high levels in the waste, was reduced to 1 ppm, below the Universal Treatment Standard (UTS) for nickel of 11 ppm. In addition to the RCRA standards, all metals also passed the much more stringent UTS standard.

- The waste was formed into a solid monolith, with no free liquid either on the surface or in the interior of the brick. The paint filter test (SW 846 – Method 9045) was passed. The bricks begin to harden immediately, and less than one hour after fabrication, they were found to have greater than 1000 p.s.i. compressive strength. Additional curing raises the compressive strength to 4700 p.s.i. after fourteen days.

- Finally, screening of the waste resulted in no discernable particulate matter less than 200 µm and less than 1% less than 10 µm in size. This meets the acceptance criteria of less than 15% of fines less than 200 µm, and less than 1% of fines less than 10 µm.

In addition to the Silo 3 WAC, the Envirobond™ process will be designed to treat waste to meet DOT shipping requirements and the Waste Acceptance Criteria for the Nevada Test Site or commercial disposal facilities. Several key process parameters were also studied. They included variability of moisture, the density, and waste loading. The effect of Envirobond™ on radon emanation was also included to ensure that disposal criteria would be met. The four heavy metals, which were found to be of potential concern, (arsenic, cadmium, chromium, and selenium), were studied by increasing the concentration by three or more times. With all four metals, elevated concentrations were successfully treated with Envirobond™ to levels below the Waste Acceptance Criteria (WAC) for shipping Silo 3 waste. In addition, waste samples that had previous been below the RCRA standard, (before optimizing the formulation), were successfully reprocessed to levels far below the Silo 3 WAC. Table I shows the results of treating a sample of Silo 3 waste with Envirobond™.

Table I. Testing to Meet Universal Treatment Standards

RCRA Metals					Universal Treatment Standards
Arsenic	1	6.26	0.53	2.6	5.0
Cadmium	0.0024	0.06	0.0024	0.02	0.11
Chromium	0.85	0.24	0.57	0.55	0.65
Selenium	0.76	0.9	1.4	1	5.7
Nickel	N/A	N/A	1	1	11

The moisture content was known to affect the quality of the bricks. To determine whether adequate bricks could be made, the moisture content was raised to a maximum of 40%. Envirobond™ was added with additional absorbent, and acceptable bricks were produced. No effect was seen on TCLP results. It was concluded that dry to saturated material can be processed using Envirobond™.

One of the key advantages of the Envirobond™ process is its ability to treat the heavy metals without adding to the volume of waste. In fact, by adding a compaction step to the process, the volume was reduced by more than 40%. The density of the product was 90 lbs/ft^3, which is acceptable for packaging and disposal. Physical tests were also conducted to determine the effect of Envirobond™ on radon emanation. The use of Envirobond™ reduced the radon flux by 26% using the laboratory scale bricks, (3.5" x 2.5" x ¾ "). Extrapolating to full-size bricks, the radon flux is reduced by up to 40%. The final waste product meets the disposal criteria of less than 20 pCi/m^2/ sec.

The treatability testing showed that the Envirobond™ process successfully treats Silo 3 waste by stabilizing the heavy metals in an insoluble metal complex. The waste form easily passed all physical criterion, including no free liquids, sufficient strength, no respirable fines, and radon emanation standards. TCLP testing of the waste form has shown that heavy metals are treated to levels not only below the Silo 3 Waste Acceptance Criteria for shipping material offsite, WAC, but also to levels below the Universal Treatment Standards (UTS) standards. The process works well with the Silo 3 particulate matter to form a monolithic brick that is easy to handle, ship, and dispose.

PROCESS CONTROL PARAMETER EVALUATION

The parameters studied for incorporation into process control are listed in Table II below:

Table II. Silo 3 Waste Process Parameters Based on Treatability Testing

Treatment Formula Specification:	Absorbent: 10 to 20 wt. % Envirobond™: 5 to 9 wt. %
Moisture Content range to make stable brick product	Minimum tested 3 wt%[1] Maximum tested 40 wt%[2]
Reprocessing parameters for products that fail to meet TCLP LDR standards.	Absorbent: minimum 15% Envirobond™: minimum 8% Rough grinding (1 to 3 mm) sufficient size reduction for reprocessing
Upper range of total metals content tested and found to meet TCLP LDR (Surrogates have been tested at higher concentrations for some metals)	As 4180 ppm Ba 650 ppm Cd 56 ppm Cr 570 ppm Pb 628 ppm Hg 0.05 ppm Se 200 ppm Ag 0.09 ppm

[1] The raw Silo 3 waste contained 3 % moisture. Additional moisture was added to raise the water content to the ideal 10 to 15 % range for brick making.

[2] to simulate high moisture conditions, water was added to the raw waste to raise the moisture content to 40%. Absorbent was added to reduce the water content to the 10 to 15% range

CONCEPTUAL DESIGN FOR RETRIEVING AND PROCESSING WASTE

Stabilization/Solidification Process Approach

The stabilization/solidification approach is in two discrete stages: Retrieval and Stabilization. The retrieval and stabilization process is described in detail in the following sections.

Waste Retrieval

The contamination levels and dispersability associated with the Silo 3 waste material requires that the retrieval process does not affect the integrity of the silo structure and maintains operator dose As Low As Reasonably Achievable (ALARA).

A support gantry will be built over the top of the silo and this structure will be used to support the retrieval equipment and the enclosures in which the retrieval equipment is housed. The purpose of the gantry is to ensure the load limitations on the silo are not exceeded. No loads will be placed within the center 20-ft section of the silo dome; no loads in excess of 700 lbs. will be placed outside the center 20-ft section of the silo dome. A remotely operated vacuum system will be deployed within the silo using a simple manipulator. This will be deployed remotely from the silo gantry on the silo top and locked into position. The vacuum will retrieve primarily from the center of the silo via the existing manway. Additional sections of vacuum pipework will be added as required using the support gantry as a working/service platform.

Additional retrieval coverage of the silo is obtained, if necessary, by articulating the manipulator. To provide the capability to retrieve waste with different physical characteristics, multiple end effectors can be attached to the vacuum unit including an auger for the break-up of compacted material.

The waste is assumed to be in three physical forms within the Silo:

- Loose dry powder/crystal waste in the top portion of the silo
- Compact dry powder/crystal waste in the middle portion of the silo
- Moist/sludge waste in the lower portion of the silo

RMRS does not propose to deploy the manipulator in any other access point other than in the center of the silo. However RMRS anticipates the deployment of lights and cameras at other access ports on top of the silo.

The sequence of retrieval operations is as follows:

Stage 1 - The manipulator is deployed into the silo.

Stage 2 - The manipulator will deploy the vacuum suction head within the silo. It may be necessary to break up compacted waste using the manipulator.

Stage 3 - With the compacted waste broken, remove waste using the vacuum system. If this is not found to be effective, then the retrieval head will be fitted with a delumping device to break up the waste to allow it to be transferred.

Stage 4 - After the compacted waste has been removed, break up moist/sludge material using a scarifier similar to the operations carried out in Stage 3.

Stage 5 – Remove any solid debris wastes which may be in the solo such as PPE, wood, etc., using manually operated long handled tools and the manipulator. This approach has been used successfully by BNFL at Sellafield to retrieve debris waste from fuel storage ponds.

Stage 6 - After removal of all of the Silo wastes, the inside will be visually inspected to ensure it has been completely emptied.

At no time during the retrieval operations will water be used to assist retrieval. The retrieval strategy is focused on the removal of the silo 3 waste while leaving the debris to be retrieved when the bulk of the material has been removed. This will be accomplished using the manipulator and long handled tools that can deploy pneumatic jaws to retrieve items that cannot be readily picked up by the vacuum unit.

The retrieval process will be monitored from the control room using remote cameras located inside the silo from the manways. This control room will be the closest manned point during retrieval operations. Transfer of the material from the silo to the stabilization plant will be effected using double contained hard piped sections.

Design Features to Minimize Risks

The double containment of the transfer piping eliminates the risk of a single pipe breach automatically shut down the transfer blowers if a blockage or breach occurs. Using the end effectors described above and maintaining adequate airflow will minimize blockages due to the material in the silo agglomerating. Limited water is used in the process, thereby reducing the probability of material holdup within the system. This reduces the schedule risk from a breakdown standpoint and reduces the risk of personnel exposure as a result of maintenance activities.

Miscellaneous electrical and instrumentation equipment such as lights and cameras will be attached to the silo entry ports to allow waste retrieval operations to be viewed on a real time basis. Real time retrieval viewing allows the operators to observe the effectiveness of the retrieval operations and anticipate if and when problems may exist and take appropriate action such as changing the manipulator end effector. The manipulator has bio-feedback to the operator.

The procedure for placement of this miscellaneous equipment on silo 3 will be fully considered during the design phase of the project, documented in construction procedures and rigorously adhered to during construction and operation. The loads associated with supporting the retrieval equipment and its housing will be supported on the support gantry and transmitted directly to the ground. Assessments will be made during the design phase of the project to ensure that the loads transmitted to the ground do not compromise silo structural integrity.

Stabilization

The Envirobond™/Envirobric™ process is particularly suited to the stabilization of the material contained within Silo 3. The Envirobond™/Envirobric™ process can achieve excellent leaching resistance and physical integrity of the finished product at high waste loadings. Preliminary testing with surrogates indicated that only 4% binder additive was required to meet TCLP LDR standards and make a solid block. The final additive make-up and waste loading will be verified during the start up testing phase.

Receipt of Feed Material

The waste from the silo will enter a separation system, which will drop out the bulk of the solid material and then return the excess air stream back to the silo. Miscellaneous items of debris encountered by the retrieval head and presenting an obstacle to retrieval will be removed during the retrieval process, either by the manipulator or other remote tools, and consigned as secondary waste.

The separator will be used to allow the waste to be continuously deposited from the retrieval vacuum system. Load cells on the feed hopper will be used to quantify the mass of waste at any given time. After collection in the cyclone the waste will be screw fed into either of the waste feed hoppers as required by the process. The hopper will be fitted with load cells such that the exact quantity of waste can be measured prior to being mixed with the binder in the mixer. The weight of waste added to the mixer will be determined as it is added to the mixer to provide both process control to determine the quantity of binder to be added to the waste and also to form the basis of payment for treating the waste.

Mixing of Feed and Additives

The binder, water and other additives, if required, will be metered into the mixer. The process will be set up for continuous throughput to a single mixer. After mixing, the waste material will be fed into an extruder volume reduction system. The volume reduction will produce a continuous stream of extruded waste bricks.

Product Material Handling

The Envirobric™ product is solid right out of the volume reduction step and can be handled immediately. The strength of the brick increases with a few hours of setting time. The leachability properties of the Envirobond™ are not dependent upon the curing of the bricks. The TCLP testing can be performed immediately after the additives are mixed in with the waste material. The product bricks are easy to manipulate in material handling systems.

The bricks will be loaded into boxes using an XYZ loader. Loaded boxes will be weighed a weigh belt. A load cell will be attached to a hoist within the airlock. This will ensure that the box weight will not exceed the 9000-pound container threshold (minus tare weight of container). The loaded box will then be sealed for interim storage.

Off-Specification Product Reprocessing

Post-treatment samples will be collected after the Silo 3 waste is mixed with Envirobond™, but prior to compaction. At this stage, the bonding agents will have immobilized the leaching characteristic of the waste and TCLP can be performed without crushing the bricks. Samples will be collected on a statistical basis. Off-specification material will be returned on a batch basis. Bricks will also be sampled on a statistical basis for physical properties. They must pass the Paint Filter Test, (SW-846 Method 9095), and must have a minimum compressive strength of 500 psi. Off-specification bricks will be sent to a hammer mill for size reduction that will prepare the rejected product bricks for reprocessing. A conveyor system will transfer the material to the mixer to be reprocessed. Because the treatment envelope is very wide, and a robust process is being used, no re-work is expected.

Waste Water System

The Envirobond™/Envirobric™ process works at low moisture contents (8% to 20%) which eliminates the need for equipment to handle wet slurries. Silo 3 material is exceeding dry, (less than 4% moisture). The majority of the daily wash water will be recycled to meet the moisture requirements to allow the waste to be extruded into bricks.

Any water generated as a res ult of process or decontamination activities will be recycled. At the close of the shutdown phase, it is anticipated that up to 500 gallons will be generated. This will be absorbed, meeting Silo 3 WAC, or packaged for off-site treatment or disposal.

FUTURE WORK

The results of the treatability showed that the Envirobond™/Envirobric™ process was successful in meeting Silo 3 WAC, and shipping and disposal criteria for the Nevada Test Site and commercial disposal facilities. Further studies of the chemical properties will be done to minimize potential interference from oxides and other chemical species that may not have been present in the sample tested. Alkalinity and oxidation-reduction potential are also important factors that will be studied to ensure that arsenic, chromium, and selenium do not become mobile as a result of chemical changes. Additional work is planned to look at the particle size, moisture and density as it relates to the formation of the Envirobric™. The effects of air transport on particle size will be addressed to ensure that after the material has been transported to the stabilization process, changes have not occurred that will affect the fabrication of the Envirobric™. Additional work will be done to define the parameters for brick formation using compaction and extrusion techniques, and the most efficient design will be selected for scale up.