Assessment of Options for the Treatment of Nitrate Salt Wastes at Los Alamos National Laboratory – 16541

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ABSTRACT

This paper summarizes the methodology used to evaluate options for treatment of the remediated nitrate salt waste containers at Los Alamos National Laboratory. The method selected must enable treatment of the waste drums, which consist of a mixture of complex nitrate salts (oxidizer) improperly mixed with sWheat Scoop®[1], an organic kitty litter and absorbent (fuel), in a manner that renders the waste safe, meets the specifications of waste acceptance criteria, and is suitable for transport and final disposal in the Waste Isolation Pilot Plant located in Carlsbad, New Mexico. A Core Remediation Team was responsible for comprehensively reviewing the options ensuring a robust, defensible treatment recommendation. The evaluation process consisted of two steps. First, a prescreening process was conducted to cull the list on the basis for a decision of feasibility of certain potential options with respect to the criteria. Then, the remaining potential options were evaluated and ranked against each of the criteria in a consistent methodology. Numerical scores were established by consensus of the review team. Finally, recommendations were developed based on current information and understanding of the scientific, technical, and regulatory situation. A discussion of the preferred options and documentation of the process used to reach the recommended treatment options are presented.

BACKGROUND

On February 14, 2014, a release of radioactivity occurred at the Waste Isolation Pilot Plant (WIPP), resulting in distribution via airborne transport of radioactivity within the repository and to the surrounding environment in the vicinity of the facility. Subsequently, WIPP personnel gained access to the area and determined that a waste drum or drums had breached. After WIPP declared a potentially inadequate safety analysis on the possibility of inadequately remediated nitrate salt-bearing waste contained in waste packages at WIPP, LANL took precautionary measures to over pack and subsequently move all remediated nitrate salt (RNS) waste drums to a Perma-Con®[2] and began monitoring the drums. Definitive photographic evidence indicated LANL RNS Drum 68660 had breached. RNS waste drums similar to those at LANL had previously been shipped to WIPP and to the Federal Cell within the waste facility in Andrews, Texas managed by Waste Control Specialists (WCS). WCS drums with this waste configuration were placed in shallow underground storage with temperature monitoring.

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[1] sWheat Scoop® is a registered trademark of Farmers Union Industries, LLC, Redwood Falls, MN.
[2] Perma-Con® is a registered trademark of Radiation Protection Systems, Inc., Groton, CT.

Precautions have been taken to protect workers, the public, and the environment from further reactions. Drums are stored under HEPA filtration and temperature controls with active fire suppression systems. Routine monitoring consists of hourly visual inspections, daily temperature measurements of standard waste boxes (SWBs) containing the RNS waste drums, and periodic sampling and analysis of the headspace gases within these SWBs.

An Administrative Compliance Order (ACO) from NMED [1] was issued to DOE and Los Alamos National Security for violations to the Hazardous Waste Facility Permit (Permit) connected to the management of nitrate salt wastes. A plan to remediate and treat the remediated daughter and unremediated parent drums pursuant to all applicable Hazardous Waste Management Regulations and Permit requirements is required. [2] [3] The Options Assessment Report [4] is an ACO requirement and provides the rationale for LANL's recommendation of the treatment options for RNS and unremediated nitrate salt wastes and includes a description of the process used to arrive at the recommendation. Viable options for treatment to render the nitrate salt wastes safe for transportation and final disposal in the WIPP repository are investigated.

INTRODUCTION

Experimental and modeling studies indicate that mixtures of metal nitrate salts (oxidizer) with sWheat® organic kitty litter (fuel) created the potential for exothermic chemical reactions. The use of sWheat® absorbent in the processing of nitrate salt wastes is the critical processing decision that led to the failure of Drum 68660, regardless of the details of the thermal processes that enabled the drum to achieve temperatures sufficient to initiate the chemical reactions.

The technical recommendation for rendering the RNS waste safe for subsequent treatment is a two-step process based on the scientific understanding gained from the Clark and Funk (2015) study [5]. First, cooling the waste is a safety measure to be performed in advance of removing the waste from its current configuration in order to sample and subsequently process the solids. Cooling drums will slow down both chemical and biological reactions thought to have led to thermal runaway. Second, mixing the RNS waste into an inorganic matrix of natural mineral zeolite like clinoptilolite will stabilize the waste. Adding zeolite to the RNS and UNS waste containers is a potential process to remove the RCRA hazardous waste characteristic (D001, ignitability) from the waste thereby meeting the WIPP Waste Acceptance Criteria (WAC). Grout is also an acceptable alternative with the important caveat that following water addition to form grout, the wetted nitrate salt/ sWheat® organic kitty litter mixture should be processed directly into concrete.[3]

[3] Results of oxidizing solids testing EMRTC Report FR 10-13 conclusively demonstrate that either zeolites (36 wt.%) or grout (55 wt.%) in proper ratios deactivate D001 characteristics per EPA SW-846, Method 1040.

OPTIONS ASSESSMENT METHODOLOGY

A methodology was developed that included an expert-based process in which a cross-disciplinary team of LANL professionals established a set of evaluation criteria and ranked the various proposed options. The Core Remediation Team convened to develop a series of options based on current waste management practices, the Clark and Funk (2015) study [5], and the availability of facilities to conduct the work.

The initial step was to establish a comprehensive list of potential RCRA treatment options for consideration. Next, a list of evaluation criteria was developed to comprehensively evaluate options against a diverse set of criteria. Then, an initial pre-screening meeting was conducted to cull the list on the basis of a decision of infeasibility of certain potential options with respect to one or more of the criteria.

Finally, the team's discussion was documented to provide the rationale for the screening decisions. The remaining potential options were then evaluated. Each option still under consideration was ranked against the criterion in a relative fashion, and numerical scores of 1-5 (a higher score was more favorable) were established by consensus. After ranking all criteria, a complete matrix of scores was determined. The final results were tabulated and the discussion and rationale for the scores were documented in a final report.

Comprehensive List of Potential Treatment Options

A range of general or industry-practice-based technologies recommended in the RCRA treatment standards (40 CFR Part 268) were used build a comprehensive list of potential treatment options. Thirteen options were evaluated. From this initial prescreening, four RCRA stabilization options were identified involving zeolite addition, zeolite addition with cementation, and wet or dry cementation. A fifth stabilization option of combined technologies, filtration and dissolution with cementation of the nitrate salt waste, was added later in the evaluation.

Table I is a list of the treatment options considered and indicates whether the option is applicable to the RNS waste, the UNS waste, debris, or any combination. Options 1 through 4 were the RCRA stabilization options and took into account scientific and technical considerations as well as facility and waste specific issues, given that the work is to be performed at LANL. Salt Dissolution With Cementation/ Stabilization was later added to the option investigation process. The descriptions represent the basis that the Core Team used in its evaluation. Options 5-13 are the other RCRA treatment options (40 CFR 268 Appendix 1) and are listed after the four RCRA stabilization options. Some options are only applicable to either the RNS or UNS waste, the RNS or UNS waste, but not for all categories of waste.

Table I. Summary of treatment options considered

Option	Description	Applicability RNS	Applicability UNS	EPA Technology Code*
RCRA Stabilization Options				
1. Stabilization Using Zeolite	Mix waste into inorganic natural mineral to eliminate ignitability potential of the waste	X	X	STABL/RHETL
2. Stabilization Using Zeolite With Cementation	Option 1, followed by production of cement waste form	X	X	STABL/RHETL
3. Stabilization Using Dry-Process Cementation	Production of cement waste form with water added only at the time of cementation	X	X	STABL
4. Stabilization Using Wet-Process Cementation	Initial water addition to eliminate potential thermal runaway reactions, followed by production of cement waste form	X		STABL/WTTRx
14. Salt Dissolution With Cementation/ Stabilization	Water addition followed by filtration and cementation process of sWheat® cake and nitrate salt solution	X		WTRRx/STABL /RHETL
Other RCRA Recommended Options				
5. Incineration	Burning of waste in a radiological incinerator	X		INCIN
6. Thermal Oxidation of Organics	Treatment of waste in air to oxidize without flame	X		RTHRM
7. Biodegradation	Biological breakdown of organics or non-metallic inorganics under aerobic or anaerobic conditions	X		BIODG
8. Chemical or Electrolytic Oxidation	Breakdown of organics through the addition of oxidation reagents	X		CHOxD
9. Chemical Reduction	Breakdown of nitrate constituents through the addition of reducing reagents	X	X	CHRED
10. Vitrification	Incorporation of waste into a glass waste form	X	X	HLVIT
11. Alternate Macro-Encapsulation	Coating of the waste with an organic polymer to reduce surface exposure	X	X	MACRO
12. Neutralization	Reagent addition to neutralize the pH	X	X	NEUTR
13. Controlled Reaction or Leaching	Removal of soluble salts by leaching with water	X	X	

* EPA Technology Code derived from 40 CFR 268.42.

LIST OF EVALUATION CRITERIA

There were 12 criteria applied to assess the various treatment options for the 60 RNS daughter containers and the 29 UNS parent containers. Since the process required a numerical score to be applied for each treatment option against each criterion, the basis for awarding a particular integer score from 1 to 5 was also defined. A list of the criteria that were applied to assess the various treatment options is shown in Table II.

Table II. List of Criteria used to evaluate the potential treatment options

Criterion	Definition of Minimum Score of 1*	Definition of Maximum Score of 5
1. Robust to Waste Stream Variability	Extremely difficult to develop a robust process	Highly likely to be a robust process
2. Ease of Permitting (Permitting Difficulties)	Extremely difficult to permit	Simple permitting process
3. Safety Basis Challenges	Extremely complex safety basis challenges	Straightforward safety basis approval process
4. Extent of Testing Required	Very onerous testing required	Straightforward testing required
5. Reduction of Toxicity, Mobility, Corrosivity, and Ignitability	Marginally effective waste form and/or difficult to package	Highly effective waste form and straightforward to package
6. Reduction of Volume	Large volume and/or large number of daughters generated	Low volume with low numbers of daughters generated
7. Short Term and Long Term Effectiveness	Effectiveness of the final waste form is questionable or indeterminate	Highly effective final waste form
8. WCS Implications	Extremely difficult to implement for WCS drums	Straightforward to implement for WCS drums
9. Scalability and Complexity	Extremely difficult to implement for drum remediation	Straightforward to implement for drum remediation
10. Facilities Challenges	Extremely difficult to implement due to Authorization Basis scope	Highly likely to implement under current LANL Authorization Basis status.
11. Schedule	Extremely time consuming	Expedited schedule is achievable
12. Cost	Extremely expensive	Cost-effective option

*If a treatment option was judged to be infeasible based on any of the criteria, it was eliminated in the initial screening and not considered further. A minimum score of 1 applied to an option that is not screened out is an unfavorable score.

SCREENING PROCESS

The Core Team evaluated the fourteen potential treatment options against the evaluation criteria leading to the recommendation of treatment options for the RNS and UNS waste streams. The evaluation occurred in two steps: A prescreening step, then a full evaluation of options not screened out in the first step. The results of the screening exercise indicate that each of the five stabilization treatment options (Options 1 through 4, and Option 14) were determined to be suitable for full evaluation, whereas the other RCRA treatment options were screened out in the initial evaluation.

Results of the Pre-Screening of Potential Options

Options 5-13 were all screened out during the pre-screening process as not viable. A summary of the results is provided below:

Option 5. Incineration. The waste is burned in a radiological incinerator. Treatment is performed in units operated in accordance with the technical operating requirements of 40 CFR Part 264 subpart O, i.e. using maximum achievable control technology. This method minimizes the mass and volume of the final waste product by destroying both the nitrate and starch components in a system with engineered controls for deflagration. The result is a highly radioactive metal oxide waste, if all of the nitrates that do not react with the cellulose decompose to a non-oxidizing solid. This operation would be very difficult to permit and is complicated by the presence of transuranics. The risk of failure to achieve the necessary safety basis and regulatory approvals was unacceptable.

Option 6. Thermal Oxidation of Organics. Waste is treated in air under high heat to oxidize fuels without flame. A heating process other than flame incineration is used to treat organic constituents of the waste stream or, secondarily, treat residues from a primary treatment process. Heating would unavoidably result in the onset of thermal runaway and further work needs to be done to ensure 60 °C is the bounding condition. This option was considered *inadvertent incineration* and is not acceptable from either a safety or regulatory basis.

Option 7. Biodegradation. Waste is treated via biologic breakdown of organics or non-metallic inorganics in units operated under either aerobic or anaerobic conditions such that a surrogate compound or indicator parameter has been substantially reduced in concentration in the residuals. Salt tolerant bacteria may be cultivated to eat the organic material. A new facility is required.

Option 8. Chemical or Electrolytic Oxidation. The waste is treated to eliminate the organics via chemical or electrolytic oxidation utilizing the following oxidation reagents (or waste reagents) or combinations of reagents: 1) hypochlorite (e.g., bleach), 2) chlorine, 3) chlorine dioxide, 4) ozone or UV light assisted ozone, 5) peroxides, 6) persulfates, 7) perchlorates, 8) permanganates; and/or (9) other oxidizing reagents of equivalent efficiency. Chemical oxidation specifically includes what is commonly referred to as alkaline chlorination. Electrochemical oxidation suffers from the low solubility of starch in aqueous solution and the necessary dilution of the waste into a large volume of aqueous solvent. This treatment process could result in a thermal runaway. The waste stream already contains oxidizing material and the goal of this treatment is to remove the oxidative properties, not enhance the waste. This option is not applicable for UNS waste since no organic absorbents are present to oxidize.

Option 9. Chemical Reduction. The waste is treated to chemically reduce the nitrate constituents utilizing the following reducing reagents (or waste reagents) or combinations of reagents: 1) sulfur dioxide, 2) sodium, potassium, or alkali salts or sulfites, bisulfites, metabisulfites, and polyethylene glycols (e.g., NaPEG and KPEG), 3) sodium hydrosulfide, 4) ferrous salts; and/or 5) other reducing reagents. Nitrates are reduced to N_2 by contacting nitrates with metal to convert nitrates to nitrites. Nitrites are reacted with amide to produce N_2 and $C0_2$. This treatment is performed in small controlled batches and concentrates TRU waste. The instability of the RNS waste is a result of the mixture of fuel with oxidants. Electrochemical oxidation suffers from the low solubility of starch in aqueous solution and the necessary dilution of the waste into a large volume of aqueous solvent. This treatment process could result in thermal runaway and the waste stream already contains oxidizing material. The goal of this treatment would enhance the waste.

Option 10. Vitrification. Waste is incorporated into a glass waste form by mixing the waste into molten glass in a melter, after which the mixture is poured and allowed to solidify and cool. Vitrified waste forms are highly durable and of uniform consistency. If the process is well controlled, all organic constituents in the RNS waste will be destroyed. However, this treatment process is equivalent to, if not more violent than, incineration. For disposal in salt at WIPP, a waste form with the durability of glass is not required.

Option 11. Alternate Macro-Encapsulation. The waste surface is coated with an organic polymer (e.g., resins and plastics) or an inert inorganic matrix to substantially reduce surface exposure to potential leaching media. Coating the oxidizing nitrate salt particles in an organic polymer would improve intimate mixing between fuel and oxidizer, potentially sensitizing the waste. Reduced susceptibility to leaching is of minimal benefit in the WIPP repository, a dry repository in bedded salt, with no groundwater intrusion and minimal natural fluids. Per EPA, this is not recommended for TRU waste.

Option 12. Neutralization. The waste is neutralized to a pH between 2 and 12. Such a treatment is likely to be part of a cementation primary treatment process or if free liquids are encountered during treatment. Both the starch and nitrostarch in RNS waste could be destroyed by adequate addition of alkaline media. While acid- or base-catalyzed hydrolysis could be used to degrade the nitrostarch component of the RNS waste, it would be difficult to monitor the progress and ensure complete destruction. The oxidizer characteristic associated with the nitrate salts in either the RNS or UNS waste remains. Neutralization is insufficient to treat the waste and must be combined with solidification or absorbent addition to be considered for removal of the D001 characteristic.

Option 13. Controlled Reaction or Leaching of Reactive Inorganic Chemicals with Water. Controlled reactions are conducted with water for highly reactive inorganic or organic chemicals with precautionary controls for protection of workers from potential violent reactions as well as precautionary controls for potential emissions of toxic/ignitable levels of gases released during the reaction. Soluble salts are removed by these reactions. This technology is similar to Option 14, but lacks the subsequent stabilization/solidification steps, which deactivate characteristics D001 and D002.

Nitrate salts in either the RNS or UNS waste could be removed by liquid/solid extraction. For the RNS waste, there is no effect on nitrated starch material, and the resulting waste would potentially be a radiological contaminated energetic fuel with no disposal path. For UNS waste, the leaching on its own would result in an aqueous waste stream that must combined with a solidification option such as cementation to be considered an adequate treatment process.

Evaluation of Potential Options

Based on the screening out of options 5 through 13 and the judgment that Options 1 through 4 and 14 were feasible, the Core Team performed a full evaluation of the latter group, which are the five RCRA stabilization options.

Option 1. Stabilization Using Zeolite. Waste is processed by removing debris prior to treatment. Then an inorganic matrix of natural mineral zeolite such as clinoptilolite is added to the RNS. The resulting mixture is not corrosive, ignitable, self-heating, or an oxidizer. Zeolite separates the waste components, reduces the potential for chemical kinetics and acts as a physical and thermal barrier against reactions. The separated debris not expected to have the D001 designation because the percent of residual reactive material is small and will be confirmed through testing. For RNS waste, drums are processed at temperatures below ambient to reduce chemical reaction risk during denesting and slow chemical kinetics potential, allowing for safe and efficient denesting and handling. The zeolite remains in the mixture and reaching physical and chemical equilibrium. Cooling does not affect the amount of water the zeolite absorbs.

Option 2. Stabilization Using Zeolite With Cementation. Waste is processed identically to Option 1 up to and including zeolite addition, at which point the ignitability and corrosivity characteristics of the waste is mitigated. The material is treated with water, additional neutralization, and cemented to produce monoliths suitable for transportation and disposal. UNS waste is processed similarly but without temperature control.

Option 3. Stabilization Using Dry-Process Cementation. Debris is removed from the RNS waste and processed smaller quantities. The RNS waste is split into smaller quantities and processed through the addition of water, neutralization, and cementation to produce monoliths that would be suitable for transportation and disposal. Cooling is not necessary since the addition of water is an endothermic reaction. Temperature controls are removed when water is added.

Option 4. Stabilization Using Wet-Process Cementation. Waste is processed by cementation, but with water addition early in the process, minimizing the flammability risk for the waste and eliminating the immediate hazard. Temperature control is removed and the wet waste is segregated and split. The daughter drums are processed by neutralization and cementation to produce monoliths suitable for transportation and disposal. The early addition of water is a safeing strategy designed for the RNS waste and is unnecessary for UNS waste.

Option 14. Salt Dissolution with Cementation/Stabilization. The salt dissolution with cementation process for RNS waste consists of waste repulped in water. Nitrates are highly soluble. The RNS drums are processed at temperatures below ambient to reduce chemical reaction risk during denesting and slow chemical kinetics potential allowing for safe and efficient denesting and handling. After denesting, the organics are separated from the mixture via filtration. sWheat® filter cake and a salt solution products are recovered in separate drums. At this stage of dissolution, TEAN is not found in the filtered cake, but rather in the liquid. Organics once dissolved in water are not combustible. The sWheat® is dissolved using caustic digestion and cemented for final preparation prior to transporting for disposal. The salt solution stream is cemented separately then transported for disposal. Addition of a base to TEAN will result in triethylamine (TEA) and the nitrate salt of the base reducing the chemical reactivity of the system.

Ranking of Options and Criteria

The core remediation team ranked each option still under consideration against the criterion in a relative fashion, and numerical scores of 1-5 (higher score being more favorable) were established. Below are the results of the ranking.

Criterion 1: Robust to Waste Stream Variability. The initial five options were reviewed for stability. Further data was examined regarding the type of cement waste forms produced by employing cementation options. Equipment and training requirements to correctly execute and consistently produce the waste forms from all options were examined. The variability of the waste from drum to drum, and within a drum, was assessed to evaluate the applicability of the treatment strategy suitable across the expected range of compositions.

After consideration of the test data, the procedural steps required, the equipment complexity, and waste stream variability, the first three options were highly likely to develop a robust process (score of 5) for both the RNS and UNS. These options involve deactivating D001/D002 for waste and debris ensured that a robust formulation could be devised to accomplish the rendering of the waste unreactive. Option 4 (Stabilization Using Wet-Process Cementation) was ranked a 3 for RNS waste due to the additional complexity of the two-week hold time after water addition, opening the possibility that low-level reactivity could vary across the drum population and complicate the process. Option 14 (Salt Dissolution With Cementation/ Stabilization) ranks a 3 because of two end streams and the waste and steel corrosion pH requirement.

Criterion 2: Ease of Permitting (Permitting Difficulty). A required modification of the Hazardous Waste Facility Permit and the degree of complexity for each treatment option required by standard RCRA permitting factors was examined. Option 4 scored a 4. Other options scored a 3. The permitting difficulty for simpler cementation is easier to execute because of the common use of cementation in the waste management industry. RCRA permitting process, schedule, and NMED review and approval would be similar for each option. The estimated extent and complexity of the submittal was the determining evaluation criterion rather than the permit modification class.

Option 1 (Stabilization Using Zeolite) is similar to the process used to prepare TRU waste containers for WIPP certification. A permit submittal requires an appropriate zeolite to inert the ignitable waste and includes final volumetric ratios. The zeolite treatment option was assigned a score of 3 for Ease of Permitting. A score of 3 was applied to the waste and debris.

Option 2 (Stabilization Using Zeolite With Cementation) combines the zeolite process with a second cementation step adding the complication of water addition and treatment by neutralization to prepare the waste for solidification with the cement. This includes Options 3 and 4. The combined steps for two processes require further technical description in the permit modification request. Potential changes to operational factors such as inspections, training, waste management operations, and emergency procedures result if additional facilities are involved in the treatment. This treatment option was assigned a score of 2 due to the increased potential for complexity in the permit modification request.
Option 3 (Stabilization Using Dry-Process Cementation) uses the same two waste management sites but limits waste processing at one facility and segregation and waste preparation at another facility. This complicates the permit requirements. The treatment option for the remediated waste stream was assigned the same score of 2 for the potential permitting complexity. The absence of the organic component in the UNS waste is less a complex technical process and the Ease of Permitting score was raised to 3 for that waste stream.

Option 4 (Stabilization Using Wet-Process Cementation) and Option 14 (Salt Dissolution With Cementation/Stabilization) use the same waste management sites and potential operational factors, implying increased operational changes associated with the permit. Cementation treatment alone in Option 4 is a simpler process and is currently approved for waste treatment. Option 14 is slightly more complex than Option 4 due to the generation and treatment of two discrete waste streams but similar in the cementation processes. The early addition of water minimizes the worker safety concerns and waste management procedures related to the oxidizer capability in the early stages of the process, a beneficial factor for permitting by potentially mitigating the degree of operational change descriptions needed to modify the permit. The need for temperature control of the waste is limited to the earliest stages of the waste treatment process, making potential permit conditions at WCRRF less complex. As a result, options 4 and 14 were assigned values of 4 and 3, respectively, for the remediated waste stream regarding permitting difficulty.

Criterion 3: Safety Basis Challenges. This criterion includes the facility features needed for radiation protection and the degree of procedure development that ensures requirements for worker safety are met. A treatment option that uses or builds from the existing safety basis analysis reduces the challenges. If facilities not previously used to treat waste are needed or if different processes are developed that are complex or require new controls, safety basis challenges increase. Option 1 (Stabilization Using Zeolite) was the simplest safety basis path forward because the operational path is already used to process nitrate salts.

Option 2 (Stabilization Using Zeolite With Cementation) and 3 (Stabilization Using Dry-Process Cementation) are identical up to the point at which zeolite is added. After that point, wastes are cemented. Because the mixing with zeolite removes the ignitability and corrosivity hazards, the any drum movement presents fewer safety basis challenges, making Option 2 (Stabilization Using Zeolite With Cementation) somewhat less onerous (from a safety basis perspective) than Option 3 (Stabilization Using Dry-Process Cementation). There is a clear separation between these options and Option 4 (Stabilization Using Wet-Process Cementation) and Option 14 (Salt Dissolution With Cementation/Stabilization), which has the challenges of the other two cementation options, but also includes movements and handling of waste to which water has been added. Due to the new additional steps Option 4 (Stabilization Using Wet-process Cementation) and Option 14 (Salt Dissolution With Cementation/ Stabilization) present the most difficult safety basis challenges and were scored a 1.

For RNS waste, there is a difference in the five options resulting in the assignment of scores of 4, 3, 2, 1, and 1 to Options 1, 2, 3, 4, and 14 respectively, for the safety basis criterion. For UNS waste, the same challenges exist, so the same scores were assigned for the first three options. Option 4 is not applicable for UNS waste or debris.

Criterion 4: Extent of Testing. The amount and complexity of sampling and analysis required for treatment are part of this criterion. Characterization of the TRU nitrate salt bearing waste stream with the D001 EPA hazardous waste number for ignitability requires that the final treated product or appropriate surrogates demonstrate the oxidizer capability has been negated by testing to SW-846 Test Method 1030, Ignitability of Solids, Test Method 1040, Oxidizing Solids, Test method 1050 Test Methods to Determine Substances Likely to Spontaneously Combust and DOT methods. Any treatment strategy requires testing. Thus, there is no scoping difference contributing to the overall score. Gas and solids sampling of the barrels was not included as it is common to all processes. The evaluation compared the amount of testing required during remediation operations and post-processing.

Cementation (all Options except Option 1, Stabilization Using Zeolite) requires achieving proper pH for the mixture for a viable grout, making pH testing mandatory during remediation. Cemented mixtures are known to dewater during storage, which adds an additional requirement[4] for tests to ensure that the solid matrix was stable and did not lose water. Thus, pH testing is not necessary or beneficial in the case of Option 1 (Stabilization Using Zeolite), and post-treatment dewatering may not be necessary when the prescribed selection of the appropriate zeolite ratio is used.

Based on these considerations, Option 1 (Stabilization Using Zeolite) scored a 5 for both RNS and UNS waste. No tests other than those requisite for waste acceptance are required. The remaining options involve cementation and require pH testing during remediation followed by surveillance for dewatering after setting. For this reason, these options all received a score of 3 for both RNS and UNS waste.

Criterion 5: Reduction of Toxicity, Mobility, Corrosivity, and Ignitability. The design and operating permit for the WIPP facility is the primary consideration for the applicability of the criteria for mobility of contaminants.[5] In a bedded salt repository,

[4] The WIPP WAC (DOE/CBFO, 2013) requires that, due to corrosivity concerns, the waste packages contain no free liquids.

[5] WIP WAC prohibits free liquid. Therefore, WIPP is not permitted to accept wastes with observable liquid that is more than 1 percent by volume of the outermost container at the time of radiography or visual examination.

the waste form is of secondary importance to the long-term performance of the repository. The waste form for all options is a solid waste confined by the waste containers. Even if the waste form dewaters over time, the amount of liquid liberated would be insufficient to facilitate transport of radionuclides through the salt bed to the accessible environment. The self-sealing salt limits the availability and transport of water into and through the repository, and minimizes the potential release of TRU nuclides. In the undisturbed repository scenarios, no significant release of actinides from the WIPP is predicted. [6] All five options meet the WIPP WAC, are an effective waste form as long as the corrosivity and ignitability characteristics of the content are removed to mitigate the safety hazard. Since, this criterion was determined to not be a discriminator among treatment options, a uniform score of 4 was applied.

Criterion 6: Reduction of Volume. The number of daughter drums generated by each option was the primary criterion used for ranking each. The estimated number of drums generated for the five options are 399, 798, 285, 342, and 285 respectively. Based on the fact that all five options increase the number of drums of waste to be disposed, the maximum number for these options was capped at 3 for Option 3 (Stabilization Using Dry-process Cementation). Scaling the remaining scores to the relative number of drums generated, Option 1 (Stabilization Using Zeolite) received a score of 2, Option 2 (Stabilization Using Zeolite With Cementation) scored a 1, Option 4 (Stabilization Using Wet-Process Cementation) scored a 2, and Option 14 (Salt Dissolution With Cementation/Stabilization) scored a 2. The corresponding scores for UNS waste, where applicable, were assigned the same values.

Criterion 7: Short Term and Long Term Effectiveness. RNS and UNS mixed with zeolite or in a concrete monolith are equally acceptable for producing acceptable waste forms if a robust, non-dewatered cemented waste form is developed. The scoring of Criterion 4 covers the development and testing of both cement waste forms. Should testing fail to reveal a cemented monolith waste form that does not undergo dewatering then Option 1 (Stabilization Using Zeolite) is the superior remediation option. If testing confirms the suitability of either type of waste form with respect to effectiveness, there is no favorable option. If enough zeolite is used, dewatering will not occur. Because of this certainty for UNS waste, scores were assigned one point higher for UNS waste than for RNS waste. All five options scored a 4 for RNS waste, and three options applicable to UNS waste scored a 5.

Criterion 8: WCS Implications. The ease of implementing a treatment process at WCS applies only to the RNS waste requiring WCS to construct and operate an on-site capability to process the waste due to the difficulty in transporting ignitable waste. The untreated waste does not meet certification of compliance for transport because it is considered ignitable and requires an exception for transportation. Comparing Option 1 (Stabilization Using Zeolite) to the three cementation options, deploying a new glove box for the single step of zeolite addition was judged to be easier than deploying new equipment for multiple steps of a cementation process. On that basis, Option 1 (Stabilization Using Zeolite) was scored a 2, and each of the cementation options was scored a 1.

Criterion 9: Scalability and Complexity. This includes the ability to treat RNS and UNS with available facilities, consideration of whether similar operations have been performed in the DOE complex, and the number and complexity of steps required. The availability of engineering controls to meet ALARA was also considered.

Option 1 (Stabilization Using Zeolite) is the easiest to implement due to the smaller number of operational steps in one facility for treatment and the precedent of having performed these operations in the past. Thus, RNS waste scored a 4. The cementation options involve more operational steps and drum transport steps. Option 3 (Stabilization Using Dry-Process Cementation) is most direct cementation option and generates the lowest number of daughter drums. Next is Option 2 (Stabilization Using Wet-Process Cementation) with one fewer step than Option 3 (but many more than Option 1) generating more daughter drums. One of the most complex, least scalable choices is Option 4 (Stabilization Using Wet-Process Cementation), involving a large number of operations and transport steps, i.e. water addition to the Perma-Con® (presenting new challenges), and the transport of drums with an introduction of significant water. Option 14 (Salt Dissolution With Cementation/ Stabilization) consists of a filtration process followed by two separate streams, nitrate solution and sWheat® cake, both requiring cementation. For these reasons, the scores issued to these four options for RNS waste were 4, 2, 3, 1, and 2 respectively.

Criterion 10: Facility Challenges. The criterion included the ability to use available sites and facilities currently operating under the approved AB for waste treatment. Option evaluation included comparison of the number of facilities used, the current operational configuration of each facility and what operation(s) are currently authorized.

Option 1 (Stabilization Using Zeolite) presented the simplest path from a facility readiness and AB perspective. For Option 1, a facility exists without modification and is already authorized for TRU waste treatment. In contrast, the three cementation options employ one additional facility and require the installation of a glove box in a Perma-Con®, with accompanying new evaluations to obtain AB approval. The cementation options are ranked significantly below Option 1 (Stabilization Using Zeolite) for this criterion. Of the four, Option 4 (Wet-Process Cementation) and Option 14 are the most challenging with respect to facilities because the addition of water requires introduction of new equipment (beyond that of the other cementation options) requiring evaluation prior to operations. The four options received scores for RNS waste of 4, 2, 2, 1, and 1 respectively. For UNS waste, the scores are one point higher than the corresponding RNS waste score for that option due to the absence of required temperature control, making the facilities challenges somewhat less onerous.

Criterion 11: Schedule. Schedule factors considered included compliance schedules, staffing requirements, and project and procedure development. Factors influencing the schedule, such as time required for permitting approvals and treatment-process facility design complexity, were not included because it was covered in other criteria. Factors influencing schedule were number of drums created, and the cycle time associated with a drum (handling to drum shipment). A lower cycle time results in a decrease in the number of drums generated requiring less storage space, potential movement, and processing time.

Option 1 (Stabilization Using Zeolite) ranked the highest among the four options due to the modest number of daughter drums created and the short drum duration. All of the cementation options have significantly longer drum durations. Options 2 (Stabilization Using Zeolite With Cementation) and 4 (Stabilization Using Wet-Process cementation) have particularly long drum durations due to the large number of steps. Option 4 has the unique requirement of a hold time on the drums after initial water addition. Option 14 (Salt Dissolution With Cementation/ Stabilization) consists of a two-part process; nitrate solution collected in one drum and sWheat® cake collected in a second drum. Both drums require cementation processing. With regard to the number of daughter drums generated, the cementation process requires leaving enough room in the drum for cement addition and mixing after splitting the RNS waste, resulting in a lengthy process of cementation being applied to a large number of daughter drums. Option 2 (Stabilization Using Zeolite With Cementation) generates a particularly large number of daughter drums, lowering this option’s rating. Option 3 (Stabilization Using Dry-Process Cementation) and Option 14 (Salt Dissolution With Cementation/ Stabilization) are the best cementation options with respect to schedule due to the relatively small number of daughter drums generated, but not as time-efficient as Option 1. The four options for RNS wastes received scores of 4, 1, 2, 1, and 2 respectively. For UNS waste, the scores applied to the three options are one point higher than the corresponding RNS waste (and debris) score for that option due to the absence of required temperature control. This would shorten the times required to complete the processing of a waste drum.

Criterion 12: Cost. Cost was not a criterion for discriminating between treatment options and was not included in the summation of scores used to rank the options.

The results of the screening and ranking process are found in Table III.

CONCLUSION

The evaluation of various processes for suitability for treating the nitrate salt wastes led to a definitive recommendation that Option 1 Stabilization Using Zeolite be pursued for both the RNS and UNS waste streams and associated debris. This result confirms the previous recommendation of Clark and Funk (2015) [5] to mix the waste with zeolite to mitigate the corrosivity and ignitability characteristics. The evaluation process was designed to be comprehensive, in terms of the variety of treatment options considered, and robust, in terms of the use of a diverse set of criteria in the evaluation. The Options Assessment Report [4] represents the formal documentation of the process for arriving at the recommended treatment option for RNS and UNS waste for consideration by NMED and DOE.

Table III. Summary results of the evaluation of treatment options vs. criteria

EVALUATION CRITERIA

	POTENTIAL TREATMENT OPTIONS	Robust to waste stream variability	Ease of permitting (permitting difficulties)	Safety basis challenges	Extent of testing required	Reduction of toxicity and mobility	Reduction in volume	Short-term and long-term effectiveness	WCS implications	Scalability and complexity	Facilities challenges	Schedule	Cost (not a primary evaluation criterion) *	SCORE
1	Stabilization Using Zeolite (remediated)	5	3	4	5	4	2	4	2	4	4	4	4	41
	Stabilization Using Zeolite (unremediated)	5	3	4	5	4	2	5	N/A	5	5	5	5	43
2	Stabilization Using Zeolite With Cementation (remediated)	5	2	3	3	4	1	4	1	2	2	1	1	28
	Stabilization Using Zeolite With Cementation (unremediated)	5	2	3	3	4	1	5	N/A	3	3	2	1	31
3	Stabilization Using Dry-Process Cementation (remediated)	5	2	2	3	4	3	4	1	3	2	2	2	31
	Stabilization Using Dry-Process Cementation (unremediated)	5	3	2	3	4	3	5	N/A	4	3	3	3	35
4	Stabilization Using Wet-Process Cementation (remediated)	3	4	1	3	4	2	4	1	1	1	1	2	25
14	Salt Dissolution With Cementation/Stabilization (remediated)	3	3	1	3	4	2	4	1	2	1	2	2	26
5	Incineration		■	■										
6	Thermal Oxidation of Organics		■	■										
7	Biodegradation			■										
8	Chemical or Electrolytic Oxidation			■										
9	Chemical Reduction			■				■						
10	Vitrification									■		■		
11	Alternate Macro-Encapsulation							■						
12	Neutralization					■								
13	Controlled Reaction or Leaching of Reactive Inorganic Chemicals With Water							■						

Note: Options developed from RCRA treatment standards are the gray-shaded rows. Red cells denote the screening out of an option based on a high degree of infeasibility with respect to that criterion. Because of the initial screened-out determination, Options 5-13 were not ranked.
*Cost not included in final score.

REFERENCES

1. New Mexico Environment Department, 2014. Administrative Compliance Order HWB-14-20, December 6, 2014.
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4. Robinson, B. A. and P. A. Stevens, 2015. Option Assessment Report: Treatment of Nitrate Salt Waste at Los Alamos National Laboratory. LANL Report LA-UR-15-27180.
5. Clark, D. L. and D. J. Funk, 2015. Waste Isolation Pilot Plant (WIPP): Chemical Reactivity and Recommended Remediation Strategy for Los Alamos Remediated Nitrate Salt (RNS) Wastes. LANL Report LA-UR-15-22393.
6. Title 40 CFR Part 191 Subparts B and C Compliance Recertification Application 2014 for the Waste Isolation Pilot Plant Appendix SOTERM-2014 Actinide Chemistry Source Term, Appendix PA-2014, Section 7.