LABORATORY PERFORMANCE TESTING OF AN EXTRUDED BITUMEN CONTAINING A

SURROGATE, SODIUM NITRATE-BASED, LOW-LEVEL AQUEOUS WASTE[a]

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ABSTRACT

Laboratory results of a comprehensive regulatory performance test program, using an extruded bitumen and a surrogate, sodium nitrate-based waste, have been compiled at the Oak Ridge National Laboratory (ORNL). The testing has shown that the relatively viscous form of oxidized bitumen that was used has been able to meet all performance requirements. Using a 53-mm Werner & Pfleiderer extruder, operated by personnel of WasteChem Corporation of Paramus, New Jersey, laboratory-scale, molded samples of ASTM D312, type III, air-blown bitumen were prepared for laboratory performance testing. A surrogate, low-level, mixed liquid waste, formulated to represent an actual on-site waste at ORNL, was used. The mixed liquid waste contained approximately 30 wt % sodium nitrate, in addition to eight heavy metals, cold cesium, and strontium. Samples tested contained three levels of waste loading: that is, 40, 50, and 60 wt % salt.

Performance test results include the 90-day American Nuclear Society (ANS) 16.1 leach test, with leach indices reported for all cations and anions, in addition to the EP toxicity test, at all levels of waste loading. Additionally, test results presented include the unconfined compressive strength and surface morphology utilizing scanning electron microscopy (SEM). Data presented include correlations between waste form loading and test results, in addition to their relationship to regulatory performance requirements.

INTRODUCTION

At the Oak Ridge National Laboratory (ORNL), ion exchange processes used to remove primarily ^{137}Cs and ^{90}Sr, as well as trace amounts of ^{60}Co and rare earths, from building process wastes results in the production of a mixed, low-level, nitric acid-based waste. The acid is heated in evaporators to recover as much acid as possible for reuse in the ion exchange circuit. Following acid recovery, the resulting slurry is made alkaline at approximately pH 12 using sodium hydroxide.

The resulting alkaline sodium nitrate solution is then pumped to storage tanks on-site at ORNL, where it is stored in tanks referred to as the Melton Valley storage tanks. The radioactive solution contains activity in the range of 9.8 E+7 to 9.8 E+8 Bq per liter and approximately 30 wt % nitrate.

Concern over the rate at which eight, 190,000-ℓ storage tanks were filling, prompted an investigation into those fixation media which were immediately available for on-site use and offered a good volumetric-reduction efficiency. Bitumen was then chosen as one of the possible fixation media which could potentially immobilize the first 378,000 ℓ of waste.

The decision process used in choosing a fixation technology or vendor involves laboratory performance testing of prospective vendors' small-scale waste form specimens. Performance testing includes compliance with test methods centered primarily around the Nuclear Regulatory Commission's (NRC's) 10 CFR 61 (1) Branch Technical Position Paper which recommends methods for demonstrating compliance with these waste form performance criteria. In addition, tests required by the Environmental Protection Agency (EPA) (2) have been performed on the waste form specimens.

BACKGROUND

During June 1986, waste form specimens were prepared at the Werner & Pfleiderer site by WasteChem Corporation personnel in northern New Jersey, using a 53-mm, twin-screw extruder. Waste form specimens were prepared in the presence of ORNL personnel to corroborate that conditions were as close as possible to those which might be encountered in the field, and then the specimens were transported to Oak Ridge for testing.

Because contamination of equipment and transportation of radioactive materials presented a major problem, a surrogate waste was prepared to represent the Melton Valley waste solution. The resulting chemical analysis of the as-prepared surrogate waste

[a]Research sponsored by the Office of Defense Waste and Transportation Management, U.S. Department of Energy, under Contract No. DE-AC05-84OR21400 with Martin Marietta Energy Systems, Inc.

used in the demonstration of the extruder technology is presented in Table I.

As shown in Table I, the waste contains metals and nonmetals of concern to the EPA and the State of Tennessee, in addition to the major salt, sodium nitrate. Currently, the waste is expected to be classified as a mixed, low-level waste, but "ultrafiltration" of the waste is expected to lower the concentration of many metals and nonmetals prior to waste fixation. Not all of these metals exist in the actual waste, but interest in the ability of bitumen to impede the leachability of all eight chemical species of current concern to the EPA caused us to include them. Although some of the chemical species are in the range of between 5 and 30 ppm in the actual waste, analytical detection limit restraints dictated the use of 50 ppm for each of the EPA chemical elements.

During the demonstration in which the specimens were prepared, three target levels of waste loading were achieved: that is 40, 50, and 60 wt % total salt. During the early stage of the demonstration, samples containing approximately 26% loading were also prepared, and some limited testing was performed on them.

TABLE I

Surrogate Waste Composition Representing the Melton Valley Nitrate-Based Waste

Species	Concentration (mg/l)
Arsenic	47
Barium	42
Cadmium	50
Calcium	13,000
Cesium	48.9
Chloride	2,200
Chromium	46
Lead	47
Magnesium	990
Mercury	38.4
Nitrate	238,000
Selenium	43
Silver	48
Sodium	77,000
Strontium	47
Sulfate	560
Density at 25°C	1.25 g/cc

Extruder Technology and Equipment

The extruder/evaporator is a one-step volume-reduction and solidification system which utilizes bitumen as the fixation medium. The extruder process equipment is very simple and consists of a direct current, geared motor which drives two nitrided steel screws that mesh together very closely. The screws are slightly offset and move in the same direction. The surfaces of the screws are designed such that they are self-wiping. The screws impart mechanical energy into the viscous bitumen, while mixing and transporting the product forward toward an orifice at the end of the machine. The screws actually effect a mixing and kneading action to the bitumen product as it moves along.

The outer steel casing of the extruder is heated in separate sections along its entire length, thereby facilitating the addition of heat to the bitumen and liquid waste. The heat lowers the viscosity of the American Society for Testing and Materials (ASTM), type III, air-blown bitumen used in the demonstration, and at the same time, evaporates the water from the waste. Typically, <1 wt % water will remain in the bitumen exiting the end of the extruder.

The electrically heated sections of the casing were maintained at controlled temperatures to evaporate the water at the desired rate. The steam produced entered domes on top of the machine, which lead to water-cooled condensers. A maximum temperature of 177°C was obtained in the hottest section, while the product exited the end of the extruder at 138°C. A schematic of the bitumen extruder process is shown in Fig. 1, and a photograph of the actual equipment showing evaporation domes, extruder barrels and power drive is presented in Fig. 2. The power drive is on the right side of the photograph. The bitumen/waste exits the extruder on the extreme left side.

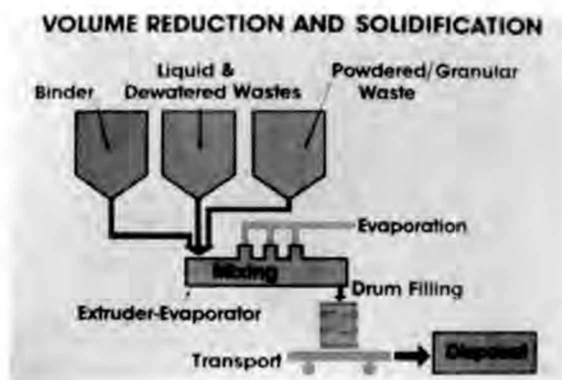

Fig. 1. Schematic of the Bitumen Extruder/Evaporator.

Fig. 2. A Photograph of the Bitumen Extruder/Evaporator.

METHODOLOGY

Waste Forms

Cylindrical waste form specimens were cast into the proper size and geometry to facilitate regulatory testing using aluminum molds with neoprene bottoms. Aluminum sheets (10 mils thick) were cut to form tubes, and aluminum-backed tape was used to hold the seams together. Disks molded from neoprene were tightly fitted into the bottom of the tubes, and a thin layer of silicone-based cement was placed on the edge of the disks to ensure a complete seal.

The aluminum molds were made longer than desired to compensate for the high thermal expansion coefficient of bitumen which causes the surface to shrink inward upon cooling. However, typical fill efficiencies have been >90% during full-scale operation. Following removal from the molds, the top of each molded sample was carefully cut and leveled using a razor knife.

The extruded bitumen product exiting the extruder unit readily flowed into the aluminum molds during the demonstration. The forms were cooled and then placed in plastic bags for storage prior to laboratory testing.

Physical Testing and Characterization

Because 10 CFR 61 requires that waste forms exhibit minimum unconfined compressive strengths of 50 psi (3.45 E+5 Pa), compression testing was performed. The recognized unconfined compressive strength test for bituminous mixtures is the ASTM method D 1074-83 (3). This test was performed on right circular cylinders of bitumen waste forms molded in soft drink cans, with a height-to-diameter ratio of 1.

The vertical rate of deformation (cross head speed) for these tests was set at 0.127 cm/min/cm of sample height. The NRC has recommended that because bitumen flows, rather than fractures, the unconfined compressive strength should be evaluated at the point where 10% deformation in specimen height occurs.

Waste form loading was determined by dissolving the bituminous waste form in dry xylene and aniline. The salts were then washed many times with solvent until it was felt that all the organic material was dissolved. The salts were then allowed to come to constant weight in the open air, where they absorbed their normal water of hydration. Correction was made for the water content of the salts in all pertinent calculations.

The waste loading is, however, reported in terms of the total weight percent salt in equilibrium with air. Samples were removed from both the top and bottom of the waste form specimens and analyzed for salt loading in an effort to see if the waste forms were homogeneous in regard to waste loading, for homogeneity is also a 10 CFR 61 regulatory requirement.

An additional consequence of the determination of the degree of waste form loading is the volumetric reduction efficiency (VRE). We have defined the VRE in this study as the ratio of the initial liquid waste volume treated to the final volume of the waste form. As a point of reference, the use of grout generally results in a VRE of approximately 0.7, which indicates approximately a 40% increase in volume, while a thermal process such as bitumenization always results in a VRE greater than unity (a volume reduction) for wastes containing volatile solvents.

The reduction efficiency has been determined on the basis of the nitrate content of the solid waste forms. Knowing the resulting analytical concentration of the feed solution, the VRE was calculated from these data.

Leaching

Two kinds of regulatory leach tests were performed in this evaluation: the EPA's EP toxicity test and the ANS 16.1 leach test (4) recommended by the NRC.

The EP toxicity tests were performed using acetic acid, according to the regulatory procedure, over 24 h. In this test, high-density polyethylene vessels were used and were connected to an automatic acetic acid delivery system composed of a burette, set point controller, and electric solenoid valve.

The leachates from the tests were analyzed for all waste species, not only the eight regulatory metals of current concern to the EPA. Analyses of the leachates from both leach tests were performed using only EPA-approved methods in an EPA-certified laboratory.

Additionally, the 90-day version of the ANS 16.1 leach test was performed on all waste forms. The leachant used in these tests was distilled water having an electrical conductivity of less than 5 µmho/cm at 25°C and a total organic content of <3 ppm.

Leach indices resulting from the use of this leach test are reported for all species after 90 days, even though this test was established primarily for radionuclides. In this way, the indices serve as a "figure-of-merit" which may be used for modeling the expected fractional release rate following disposal.

Leaching was performed in one-half liter Teflon vessels prepared in accordance with the MCC-1 cleaning method (5). The waste forms were suspended in stainless steel wire baskets in the center of the leachant. These tests are continuing past the regulatory 90 days until essentially all the nitrate has leached from the waste forms. During the extended testing, the effects of the observed physical changes occurring to the waste forms during prolonged contact with water will continue to be investigated.

Morphology

Because of interest in the possible mechanism involved in the leaching process, the morphological changes occurring to the waste form surface were investigated using SEM. Problems were encountered with the electron beam melting the waste form at 3,000 V. To circumvent the problem, photomicrographs have to be be taken as quickly as possible.

Dry xylene was used to dissolve the surface of the waste form, exposing the fixed salts below. Photomicrographs of the exposed salts were made at a magnification of 457X and 3,000 V without gold sputtering.

RESULTS

Unconfined Compressive Strength

One of the first tests performed on the waste forms was the unconfined compressive strength test. The results of the tests are shown in Table II, with the pure, salt-free bitumen sample serving as the blank.

The regulatory minimum compressive strength is 3.45 E+05 Pa (50 psi). As shown in Table II, all waste forms easily passed this minimum, including the blank. The greater resistance offered by increasing amounts of salt is evident from the upward trend in compressive strength. The waste form containing approximately 60 wt % salt was the maximum loading achievable during the demonstration.

TABLE II

Unconfined Compressive Strength Data at Ten Percent Deformation

Salt Loading (wt %)	Compressive Strength (Pa)
0.0 (blank)	1.72 E+06
41.6	2.48 E+06
50.6	3.52 E+06
60.2	4.29 E+06

Salt Loading

Salt loading is important in two different ways--a high percent loading is desirable up to the point that the leachability of the waste form is negatively impacted, and homogeneity along the length of the waste form is desirable since salt crystals will then generally be evenly coated.

Since the density of the bulk salt, sodium nitrate, is 2.3 g/cc and the bitumen density is less than one at the operating temperature, one can understand why salts might settle to the bottom of the waste form during cooling and solidification of the bitumen, especially inasmuch as thermal conductivity is low. A larger amount of salt at the bottom of the waste form may enhance leachability because the film of bitumen surrounding each salt crystal would have to be thinner, and therefore present less of a barrier toward leaching. Table III presents the results of the salt content analyses 2 cm below the top and 2 cm above the bottom of the waste forms.

TABLE III

Waste Form Loading Distribution

Target Loading (wt %)	Loading Achieved (wt %)	
	Top	Bottom
40	41.5	41.6
50	49.7	50.6
60	59.2	60.2

As shown in Table III, the waste forms were very homogeneous and close to the targeted waste loading. The harder, more viscous bitumen used in immobilizing this waste likely contributed much to the degree of homogeneity obtained, together with the mixing and kneading provided by the extruder process equipment.

Volumeric Reduction Efficiency Ratio

Analyzing the feed solution and comparing these data with the amount of nitrate present per unit volume of waste form, the VRE has been calculated. The results of these calculations are presented in Table IV.

As shown in Table IV, the VRE is high, as one might expect of a thermal process when a relatively dilute aqueous waste stream is involved. Such a reduction can offer desirable cost savings when transportation or interim storage of immobilized waste is involved.

TABLE IV

Calculated Volumetric Reduction Efficiency[a] Ratio

Salt Loading (wt %)	Efficiency Ratio (unitless)
41.6	1.51
50.6	1.68
60.2	2.43

[a]Vida Supra.

Leaching - EP Toxicity

Because the Melton Valley waste is currently a mixed waste, interest in the leachability of the eight EPA metals is of importance. Planned treatment schemes incorporating "ultrafiltration" may cause the waste not to be classed in this way prior to actual waste fixation. For this reason, regulatory EP toxicity tests were performed on all waste forms, including a waste form obtained during the fixation demonstration which contained 26.1 wt % salt. Tests performed with this waste form permitted us to establish the lower limit for waste loading and the resulting nitrate present in the EP toxicity test leachate.

Although the concentration of nitrate is not of concern in this regulatory test, we have endeavored to meet drinking water standards (6) without applying a dilution factor. The exact application of such a concentration for nitrate is still being debated among regulators, as well as the associated point of compliance in a waste disposal area. Without guidance, we have used the regulatory limit of 50-ppm nitrogen or 44.4-ppm nitrate as our upper limit. The results of our EP toxicity tests are presented in Table V.

As the data in Table V show, all waste forms at all levels of loading easily passed the EP toxicity test. The data show that for most of the metals, the concentration in the leachates was at or near the analytical detection limit.

The drinking water standard for nitrate, as applied, was exceeded for all waste forms, except for that waste specimen containing 26.1 wt % salt loading. The sensitivity of the leachability of the very mobile nitrate anion to waste form loading is probably a result of differing bitumen wall thicknesses surrounding waste salt crystals at the various levels of loading.

In an attempt to interpolate that level of salt loading likely to pass the nitrate drinking water standard as we have chosen to apply it, the data in Table V were correlated. Upon plotting the logarithm of the nitrate concentration in the leachate versus the waste form salt loading, a nearly straight line is obtained as shown in Fig. 3.

From Fig. 3, interpolation reveals that a waste form loading up to as high as 38% will, theoretically, pass the drinking water standards as applied. This

TABLE V

A Comparison of EP Toxicity Test Results as a Function of Salt Loading

Loading (wt %)	26.1	41.6	50.6	60.2	Limit
Species	Concentration (ppm)				
Arsenic	0.008	<0.005	<0.005	<0.005	5
Barium	0.017	0.010	0.024	0.049	100
Cadmium	<0.0030	0.0032	0.0081	0.024	1
Chromium	<0.010	<0.010	<0.010	<0.010	5
Lead	0.010	0.033	0.010	0.016	5
Mercury	<0.0002	0.0003	0.0003	<0.0002	0.2
Selenium	<0.0050	0.027	<0.005	0.005	1
Silver	<0.0060	<0.0060	<0.0060	<0.0060	5
Nitrate	12.9	54	160	320	44.3

observation, coupled with the fact that the exact application of the nitrate concentration limit by regulators is at present uncertain, is promising.

Leaching - ANS 16.1 Test

The NRC requires that a radionuclide possess a leach index of at least 6 when leached in accordance with the ANS 16.1 leach procedure. Because the leach index is the negative logarithm of the effective diffusion coefficient, it can be used in modeling the expected leaching behavior of any waste species if certain assumptions are made (see Ref. 4). The leach index then becomes a "figure-of-merit" used to ascertain the ability of a given waste form or treatment scheme to impede the leachability of a waste component.

Because the leach indices are based on a logarithmic relationship, each unit change in the leach index is a change in the effective diffusion coefficient of a factor of 10. The higher the leach index, the better the resistance of the waste form to

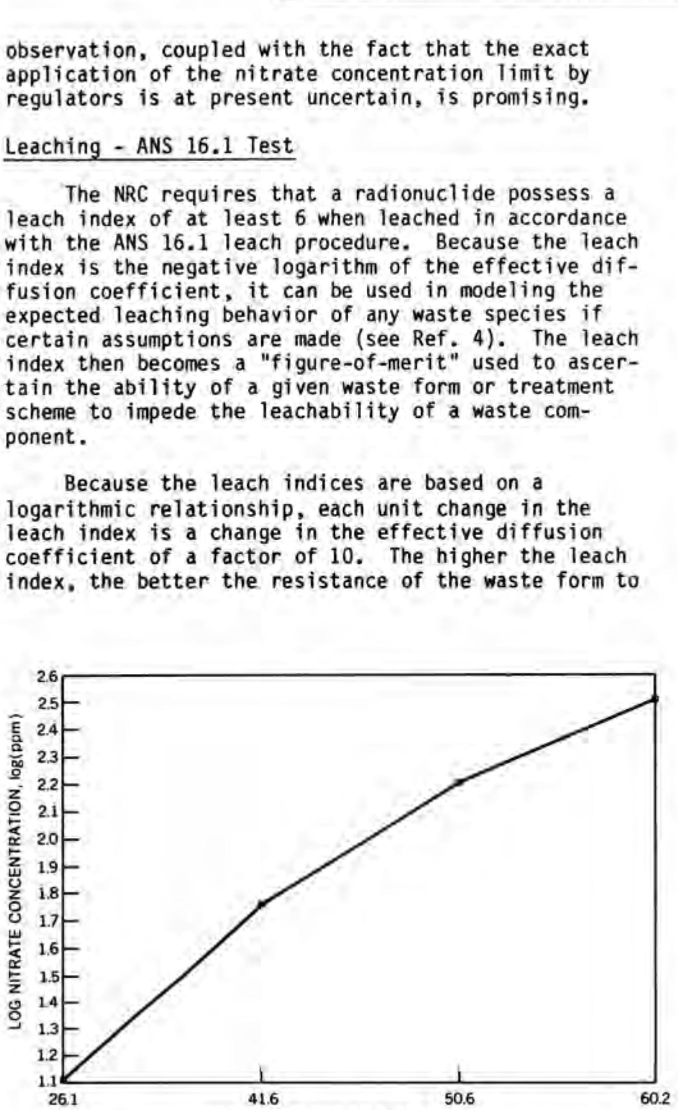

Fig. 3. Correlation of Nitrate Concentration in the EP Toxicity Test Leachate with Salt Loading.

leaching of that waste species. For a more in-depth understanding of the way in which the leach indices have been calculated, the authors refer the reader to Ref. 4.

The resultant leach indices obtained for all waste components are presented in Table VI. From the the data, one sees that both cesium and strontium exceeded the NRC's minimum leach index of 6. Applying this limit to all remaining species, even though they are not radionuclides, one sees that they have also exceeded the minimum leach index by a large margin.

The "greater than" symbols preceding some of the indices are present as a result of analytical detection limit constraints. When forced to use non-radiological species, the "less than" concentrations must become greater than symbols when reporting the indices.

With the exception of those indices in Table VI which are preceded by a greater than symbol, a general trend toward lower leach indices with increased salt loading is apparent. This apparent trend is likely the result of thicker walls of bitumen between salt crystals when salt loading is decreased.

To visualize the leaching process as a function of time, the cumulative mass fraction of nitrate leached from the bitumen waste forms is presented in Fig. 4 as a function of cumulative time. Although only nitrate is shown, the leaching of all waste species followed the same general shape of this curve. The use of data in which concentrations were consistently below analytical detection limits is of limited value, and so only nitrate is presented as an example.

Since the mass fractional release of waste species is assumed to be diffusion controlled in the interpretation of the ANS 16.1 test procedure, a square root of time relationship is frequently employed when the fractional release is plotted versus this parameter. A straight line relationship is expected when diffusion control is, in fact, the limiting process in the leaching mechanism when data are plotted, as long as the specimen approximates a semi-infinite medium. For this reason, the cumulative mass fractional release is plotted against the square root of time in Fig. 5.

An interesting point to note in Fig. 5 is the positive deviation of the curve after approximately 45 days. This deviation has been found to be real, for data currently being obtained past 90 days (not presented here) reveals that the curve turns upward with time. A similar change in the leaching rate after approximately the same amount of time is reported in the literature when leaching emulsified bitumen waste forms (8).

TABLE VI

Tabulation of Average Leach Indices After Ninety Days of Leaching in Distilled Water

Loading (wt %)	41.6		50.6		60.2	
	Average[a]	Std Dev[b]	Average[a]	Std Dev[b]	Average[a]	Std Dev[b]
Arsenic	>10.9	0.8	>11.1	0.8	>11.3	0.8
Barium	10.7	0.5	10.2	0.4	9.8	0.3
Cadmium	>10.7	0.3	10.6	0.3	10.4	0.5
Calcium	10.5	0.4	9.8	0.3	9.4	0.3
Cesium	> 8.0	0.4	> 8.4	0.7	> 8.6	0.8
Chloride	> 9.7	0.7	> 9.7	0.5	> 9.6	0.4
Chromium	>10.3	0.8	>10.5	0.8	>10.7	0.8
Lead	9.7	1.1	10.0	0.9	10.2	0.9
Magnesium	10.7	0.5	10.5	0.5	10.4	0.5
Mercury	>13.7	0.8	>13.9	0.8	>14.1	0.8
Nitrate	10.6	0.5	9.8	0.2	9.3	0.3
Selenium	>10.9	0.8	>11.1	0.7	>11.2	0.7
Silver	>10.8	0.8	>11.0	0.8	>11.2	0.8
Sodium	10.4	0.4	9.5	0.2	9.2	0.3
Strontium	10.7	0.5	10.0	0.3	9.4	0.3

[a]Arithmetic average of three replicates.
[b]Root-mean-square (7) of the standard deviations of the replicates.

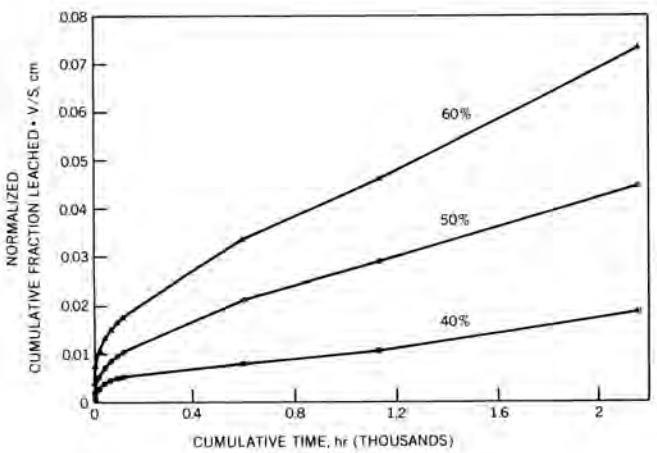

Fig. 4. Plot of the Cumulative Fractional Release of Nitrate as a Function of Cumulative Time.

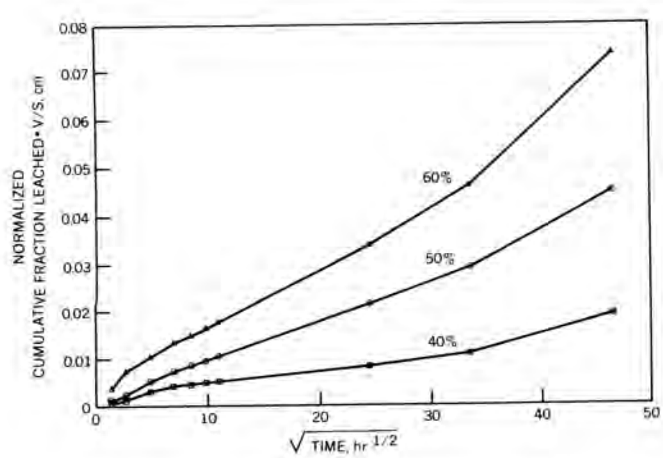

Fig. 5. Plot of the Cumulative Fractional Release of Nitrate as a Function of the Square Root of Time.

Morphology

Bitumen-based waste forms containing water-soluble salts are known to swell as a result of osmotic forces which are in effect when the waste form is immersed in water (9). These forces can be related to those involved when two solutions of different concentration are separated by a semipermeable membrane.

The vapor pressure of pure water at 25°C is 23.8 mm of mercury, while the vapor pressure of water above a saturated solution of sodium nitrate is only 17.8 mm of mercury at this temperature (10). This difference in pressure can result in an osmotic pressure maximum as high as 390 atm when calculated using a modified Van't Hoff relationship (11). Figure 6 is a schematic which shows pictorially what is believed to be occurring during the leaching process.

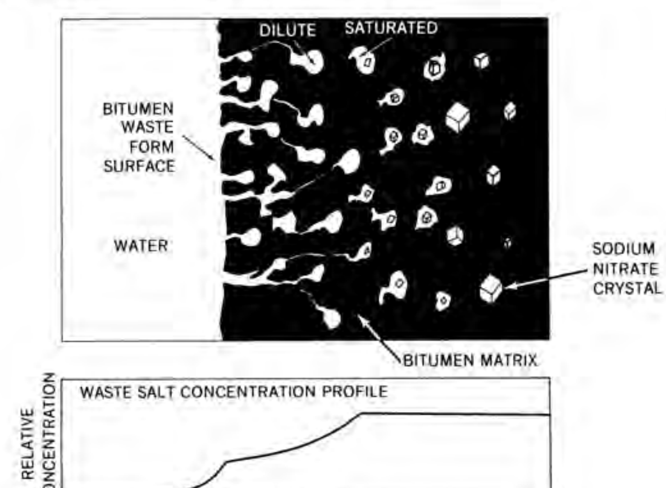

Fig. 6. Proposed, Staged Sequence of Sodium Nitrate Leaching from a Bitumen Matrix.

Figure 6 depicts leachant water passing through the bitumenous material and into an area containing salt crystals. When the water dissolves the salt, the resulting saturated solution expands into a cavity, forming interconnecting channels to the outer surface.

The leach data from this performance testing study appear to substantiate the belief that at lower waste loadings, when a very homogeneous waste form is involved, the waste particles are surrounded by a thicker wall of bitumen, thereby offering resistance to the leaching process. In an effort to observe the immobilized salts and their arrangement in the bitumen host matrix, SEM was utilized.

Figure 7 shows the salts embedded in the bitumen matrix when using organic solvent to dissolve the surface bitumen, revealing the salts and their relative

Fig. 7. A SEM Photomicrograph of a Solvent-Cleaned Surface of a Bitumen Waste Form Revealing Immobilized Nitrate Salts at a Magnification of 457X.

positions to each other. The photomicrograph shows salt crystals in the size range of between 10 and 20 µm. The crystals do not appear to be symmetrically arranged with walls of bitumen individually separating each crystal; rather, they appear to be bunched together with small pockets of void space found at random.

CONCLUSIONS

Regulatory performance testing of extruded bitumen has shown that the relatively viscous form of oxidized bitumen used has been able to meet all required performance tests. The extruder technology used to prepare the test specimens was shown to combine both superior physical and thermal processing capabilities in one unit, thereby permitting the mixing of high-viscosity materials in an extremely homogeneous manner.

The extruder bitumen process has been able to achieve high waste loading and still result in a waste form capable of offering superior resistance to the leachability of those current EPA metals of concern while showing promising results for very problematic nitrate.

The high VRE observed with this type of thermal waste fixation process undoubtedly offers potential transportation and storage cost savings. Long-term leach testing is continuing in order to observe the effects of the physical changes occurring to the waste forms during prolonged contact with water.

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