CONF-870306--4

LABORATORY PERFORMANCE TESTING OF AN EXTRUDED BITUMEN CONTAINING A SURROGATE, SODIUM NITRATE-BASED, LOW-LEVEL AQUEOUS WASTE*

A. J. Mattus
Oak Ridge National Laboratory
P. O. Box P
Oak Ridge, Tennessee 37831

and

M. M. Kaczmarsky
WasteChem Corporation
One Kalisa Way
Paramus, New Jersey 07652

CONF-870306--4

DE87 004272

To be presented at the Waste Management 1987 Meeting
Tucson, Arizona
March 1-5, 1987

For publication in the
Proceedings of Waste Mangement

*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.

DRAFT

LABORATORY PERFORMANCE TESTING OF AN EXTRUDED BITUMEN CONTAINING A SURROGATE, SODIUM NITRATE-BASED, LOW-LEVEL AQUEOUS WASTE

A. J. Mattus[1]
M. M. Kaczmarsky[2]

[1]Martin Marietta Energy Systems, Inc.
P. O. Box P
Oak Ridge, Tennessee 37831

[2]WasteChem Corporation
One Kalisa Way
Paramus, New Jersey 07652

ABSTRACT

Laboratory results of a comprehensive, regulatory performance test program, utilizing an extruded bitumen and a surrogate, sodium nitrate-based waste, have been compiled at the Oak Ridge National Laboratory (ORNL). Using a 53 millimeter, Werner & Pfleiderer extruder, operated by personnel of WasteChem Corporation of Paramus, New Jersey, laboratory-scale, molded samples of type three, 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, containing approximately 30 weight percent sodium nitrate, in addition to eight heavy metals, cold cesium and strontium was utilized. Samples tested contained three levels of waste loading: that is, forty, fifty and sixty weight percent salt.

Performance test results include the ninety day 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 also include the unconfined compressive strength and surface morphology utilizing scanning electron microscopy. Data presented

includes correlations between waste form loading and test results, in addition to their relationship to regulatory performance requirements.

INTRODUCTION

At the Oak Ridge National Laboratory, ion exchange processes used to remove primarily cesium 137 and strontium 90, as well as trace amounts of cobalt 60 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 neutralized to approximately pH 12 utilizing sodium hydroxide.

The resulting alkaline, sodium nitrate solution is then pumped to storage tanks on site at ORNL where it is stored in what are called the Melton Valley storage tanks. The radioactive solution contains activity in the range of between 0.01 to 0.1 Curies per gallon and approximately 30 weight percent nitrate.

Concern over the rate at which eight, fifty-thousand gallon 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 mediums which could potentially immobilize the first one-hundred thousand gallons of waste.

The decision process involved in choosing a fixation technology or vendor, involves laboratory performance testing of prospective vendor's, small-scale waste form specimens. Performance testing has involved compliance with test methods centered primarily around the Nuclear Regulatory Commission's (NRC) 10 CFR 61 (1) Branch Technical Position

Paper on 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 of 1986, waste form specimens were prepared at the Werner & Pfleiderer site by WasteChem Corporation personnel in northern New Jersey, utilizing a 53 millimeter extruder. Waste form specimens were prepared in the presence of ORNL personnel to ensure that conditions were as close as possible to those which might be encountered in the field, after which the specimens were transported to Oak Ridge for testing.

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

As shown in Table I, the waste does contain heavy metals of concern to the EPA and 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 of these heavy metals 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 heavy metals of current concern to the EPA caused us to include them. Although some of the metals which are in the actual waste range between 5 and 30 ppm, analytical detection limit restraints dictated the use of 50 ppm for each of the EPA heavy metals.

TABLE I

Target Surrogate Waste Composition Representing the Melton Valley Nitrate-Based Waste

Salt	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 gm/cc

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

Extruder Technology and Equipment

The extruder process equipment is actually very simple and basically consists of a direct current, geared motor which drives two nitrided steel screws that mesh together very closely. The screws are slightly off set and move in the same direction. The surfaces of the screws are so close that they are self-wiping. The screws impart mechanical energy into the viscous bitumen, while mixing and transporting the product forward towards 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 it's entire length, thereby facilitating the addition of heat to the bitumen and liquid waste. The heat lowers the viscosity of the ASTM Type III, air blown bitumen used in the demonstration, while at the same time, evaporates the water from the waste. Typically, less than one weight percent 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 was removed by 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 and power drive to the left side of the photo is presented in Fig. 2.

METHODOLOGY

Waste Forms

Cylindrical waste form specimens were casted into the proper size and geometry to facilitate regulatory testing using aluminum molds with "Neoprene" bottoms. Ten mil, aluminum sheets 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 insure a complete seal.

The aluminum molds were made longer than desired by approximately one and a half mold diameters. This was necessary because the high thermal expansion coefficient of bitumen causes the surface to shrink inward upon cooling. 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 to prevent the uptake of water from the surrounding air.

Physical Testing and Characterization

Since 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 American Society for Testing and Materials

(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 one.

The vertical rate of deformation (cross head speed) for these tests was set at 0.05 inches/min/inch of sample height (0.127 cm/min/cm of height). The NRC has recommended that since bitumen flows, rather than fractures, the unconfined compressive strength should be evaluated at the point where ten percent 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. The water content of the salts was corrected for 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 regards 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, being indicative of approximately a 40 percent increase in volume, while a thermal process such as bitumenization always results in a VRE greater than unity, that is a volume reduction.

Volume 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 types of regulatory leach tests were performed in this evaluation: the Environmental Protection Agency's EP Toxicity test, and the NRC's ANS 16.1 leach test (4).

The EP Toxicity tests were performed using acetic acid as per the regulatory procedure, over twenty-four hours. 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 leachate from the tests were analyzed for all waste species, not only the eight regulatory metals of current concern to the EPA. Analysis of the leachates from both leach tests were performed using only EPA approved methods in a EPA certified laboratory.

Additionally, the ninety-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 less than 3 ppm.

Leach indices resulting from the use of this leach test are reported for all species after ninety 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 wire baskets fabricated from 316 stainless steel in the center of the leachant. Due to physical changes occurring to the surface of the waste forms, these tests are continuing past ninety days. Leaching will continue until all the nitrate has leached from the waste forms in an effort to investigate the effects of the physical changes observed.

Morphology

Due to interest in the possible mechanism involved in the the leaching process, the morphological changes occurring to the waste form surface were investigated using scanning electron microscopy (SEM). Problems were encountered with the electron beam melting the waste form at three-thousand volts; to circumvent the problem photomicrographs had 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 three-thousand volts, without gold sputtering.

Visual light photographs were also taken to show the extent of the surface changes that were occuring. Waste forms containing approximately sixty percent loading are shown before and after ninety days of leaching.

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 the table, all waste forms easily passed this minimum, including the blank, type III bitumen without salt. The greater resistance offered by increasing amounts of salt loading is evident from the upward trend in compressive strength. The waste form containing approximately sixty percent loading was the maximum loading achievable during the demonstration.

Salt Loading

Salt loading is important in two different ways, in that 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 partly for the same reason.

Because the density of the bulk salt, sodium nitrate is 2.3 gm/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 since the thermal conductivity is very poor. 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 towards leaching. Table III presents the results of the salt content analyses two centimeters below the top and two centimeters above the bottom of the waste forms.

TABLE II

Table of Unconfined Compressive Strength Data at Ten Percent Deformation

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

TABLE III

Waste Form Loading Distribution

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

As Table III shows, the waste forms were found to be very homogeneous and close to the targeted waste loading. The harder, more viscous ASTM type III 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.

Volume Reduction Efficiency Ratio

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

As the table shows, the volume reduction efficiency 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.

Leaching-EP Toxicity

Because the Melton Valley waste is a mixed waste at present, 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 percent loading. Tests performed with this waste form permitted us to establish the lower limit in regards to 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 strived to meet drinking water standards (6)

TABLE IV

Calculated Volumetric Reduction Efficiency *
Ratio

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

* _Vide Supra_

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 shows, 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 is however exceeded for all waste forms except for that waste specimen containing 26.1 percent 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 was 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 percent will theoretically pass the drinking water standards as applied. This observation, coupled with the fact that the exact application of the nitrate concentration limit by regulators is at present uncertain is promising.

TABLE V

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

Species	Loading,(%) Concentration, (ppm)				
	26.1	41.6	50.6	60.2	Limit
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

Leaching- ANS 16.1 Test

The NRC requires that a radionuclide posses a leach index of at least six when leached in accordance with the ANS 16.1 leach procedure. Because the leach index is the negative logarithm of the effective diffusion, it can be used in modeling the expected leaching behavior of any waste species if certain assumptions are made, as outlined in 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.

Since the leach indices are based upon a logarithmic relationship, each unit change in the leach index is a change in the effective diffusion coefficient of ten. The higher the leach index, the better the resistance of the waste form to impede 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 below. From the data one sees that both cesium and strontium exceeded the minimum leach index of six. Upon applying this limit to all remaining species, even though they are not surrogate 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 nonradiological species, the "less than" concentrations must become "greater than" symbols when reporting the indices.

TABLE VI

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

Loading(%)	41.6		50.6		60.2	
Species	Average*	Std Dev**	Average*	Std Dev**	Average*	Std Dev**
Arsenic	> 10.9	1.4	>11.1	1.4	>11.3	1.4
Barium	10.7	0.9	10.2	0.6	9.8	0.6
Cadmium	> 10.7	1.4	10.6	0.6	10.4	0.9
Calcium	10.5	0.7	9.8	0.5	9.4	0.5
Cesium	> 7.1	1.9	> 7.8	2.0	> 8.4	1.8
Chloride	> 9.7	1.2	> 9.7	0.8	> 9.6	0.6
Chromium	> 10.3	1.4	>10.5	1.4	>10.7	1.4
Lead	9.7	1.9	10.0	1.6	10.2	1.6
Magnesium	10.7	0.9	10.5	0.8	10.4	0.9
Mercury	> 13.7	1.4	>13.9	1.4	>14.1	1.4
Nitrate	10.6	0.9	9.8	0.4	9.3	0.5
Selenium	> 10.9	1.4	>11.1	1.3	>11.2	1.2
Silver	> 10.8	1.4	>11.0	1.4	>11.2	1.4
Sodium	10.4	0.7	9.5	0.3	9.2	0.4
Strontium	10.7	0.9	10.0	0.6	9.4	0.5

* Arithmetic average of three replicates.
** Geometric mean of the standard deviations of the replicates.

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

In order 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, 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.

Because the mass fractional release of waste species is assumed to be diffusion controlled in the ANS 16.1 test procedure, a square root of time relationship is usually employed when the fractional release is plotted versus this parameter. A straight line is expected when diffusion control is in fact the limiting process in the leaching mechanism, when this type of data is plotted. For this reason, the cumulative mass fractional release is plotted against the square root of time in Fig. 5 below.

An interesting point to note in Fig. 5 is the positive deviation of the curve after approximately forty-five days. This deviation has been found to be real, for data currently being obtained past ninety days, and not presented here, does reveal that the curve begins to turn upward with time, presumably as physical changes in the bitumen waste form surface occurs. 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 (7). Documentation of early stages of the physical change will be presented and discussed below.

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 (8). 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 mmHg, while the vapor pressure of water above a saturated solution of sodium nitrate is only 17.8 mmHg at this temperature (9). This difference in pressure can result in an osmotic pressure maximum of as high as 390 atmospheres when calculated using a modified van't Hoff relationship (10).

The authors feel that these forces are acting to expand the outer surfaces of the waste forms into a spongy-like material with a vastly greated surface exposed to the water outside. Figure VI is a schematic which shows pictorially what is believed to be occurring during the leaching process.

In Fig. 6 it is assumed that leachant water passes through the bitumenous material and into the area containing the salt crystals. When the water dissolves the salt, the resulting saturated solution expands into a cavity. At some point in time, the expanding cavity either ruptures or tears to form interconnecting channels to the outer surface. This action results in expansion of the bitumen surface outward forming a spongy outer coating with a hard surface below.

With time the spongy layer grows as the water moves further into the waste form but progressively at a slower rate due to a diminished concentration gradient. Waste forms containing 60.2 percent salt are shown in Fig. 7 below after leaching for ninety days. The photograph shows the starting, unleached specimen on the left, and the expanded spongy surface on the right, with the soft material removed to show the still hard surface below.

The leach data from this performance testing study appears to substantiate the belief that at lower and lower waste loadings, when a very homogeneous waste form is involved, the thicker the walls of bitumen surrounding the waste particles the better. In an effort to observe the immobilized salts and their arrangement in the bitumen host matrix, a scanning electron microscope(SEM) was utilized.

Figure 8 below shows the salts in the bitumen matrix. Organic solvent dissolved surface bitumen revealing the salts below, and their relative positions to each other in the host matrix.

Figure 8 shows salts in the size range of between ten and twenty microns. The salts do not appear to be symmetrically aranged with walls of bitumen individually separating each crystal, at least not on this scale; 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 utilized has been able to meet all required performace 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 heavy metals while showing promising results for very problematic nitrate.

The high volumetric reduction efficiency 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.

REFERENCES

1. 10 CFR 61.55, Code of Federal Regulations, "Waste Classification - Low-Level Waste Licensing Branch Technical Position on Radioactive Waste Classification," Rev. 0, May, 1983.

2. SW-846, U.S. Environmental Protection Agency, "Test Methods for Evaluating Solid Waste - Physical/Chemical Methods," 2nd Edition, 1982.

3. ASTM D 1074-83, "Compressive Strength of Bituminous Mixtures," The American Society of Testing Materials, Annual Book of ASTM Standards, 1983.

4. ANS 16.1, American Nuclear Society Standards Committee, American Nuclear Society, "Measurement of the Leachability of Solidified Low-Level Radioactive Wastes by a Short-Term Procedure," Final Draft, February 6, 1986.

5. MCC-1, "Static Leach Test Method-Draft," MRB-0326, Materials Characterization Center, May, 1984.

6. 40 CFR 141, U.S. Environmental Protection Agency, "National Interim Primary Drinking Water Regulations," Code of Federal Regulations, July, 1983.

7. H. W. Godbee, J.H. Goode, and R. E. Blanco, "Development of a Process for Incorporation of Radioactive Waste Solutions and Slurries in Emulsified Asphalt, "Environmental Science and Technology, 2, pp 1034-40, 1968.

8. K. Broderson, "The Influence of Water Uptake on the Long-Term Stability of Conditioned Waste," Radioactive Waste Management-A Series of Monographs and Tracts, Testing, Evaluation and Shallow Land Burial of Low and Medium Radioactive Waste Forms, Harwood Academic Publishers, New York, NY, Volume 13, 1986.

9. National Research Council, International Critical Tables, McGraw-Hill Book Company, Inc., Volume III, First Edition, 1928.

10. D. Farrington and R. A. Alberty, Physical Chemistry, John Wiley & Sons, Inc., New York, NY, Third Edition, pp 223-24, 1956.

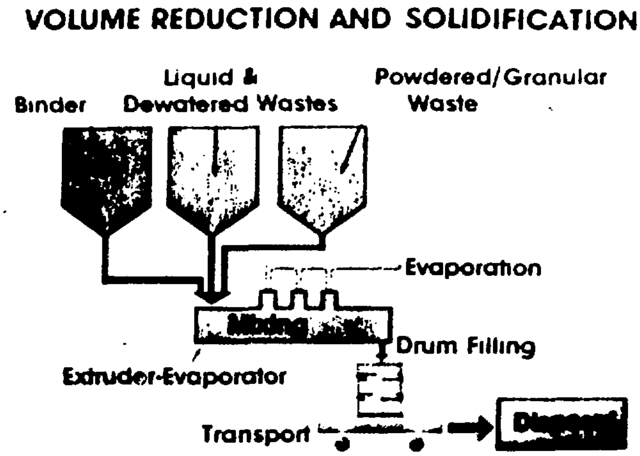

Fig. 1. Schematic of the bitumen extruder process.

Fig. 2. A photograph of the bitumen extruder.

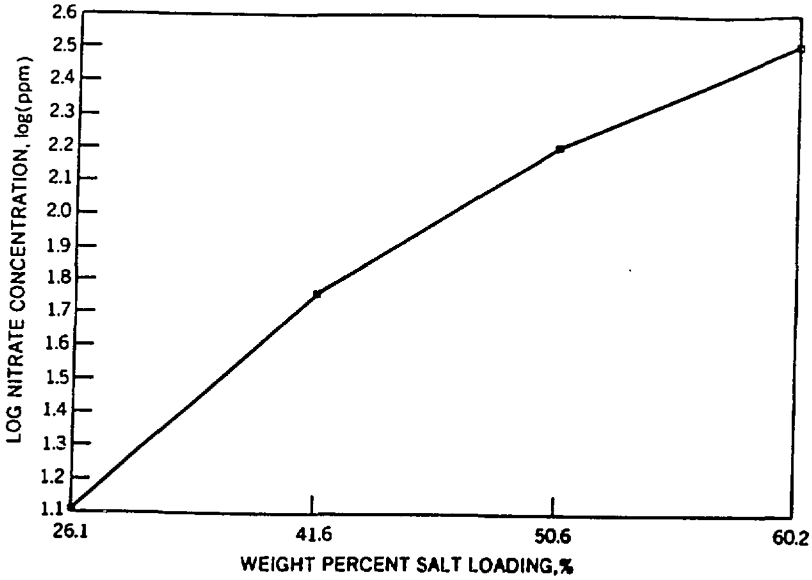

Fig. 3. Correlation of nitrate concentration in the EP Toxicity test leachate with salt loading.

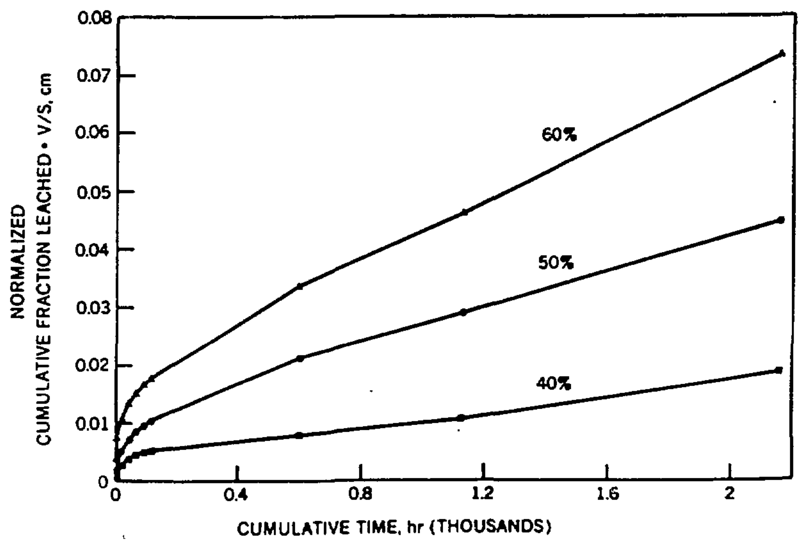

Fig. 4. Plot of the cumulative fractional release of nitrate as a function of cumulative time.

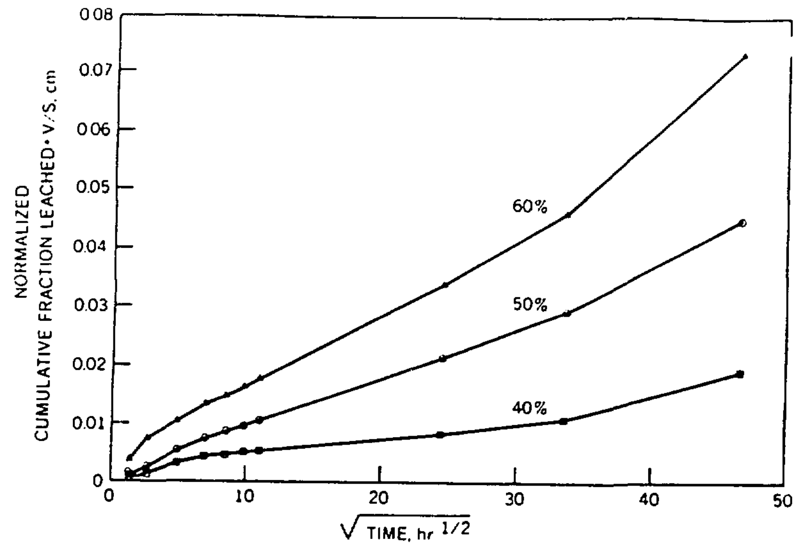

Fig. 5. Plot of the cumulative fractional release of nitrate as a function of the square root of time.

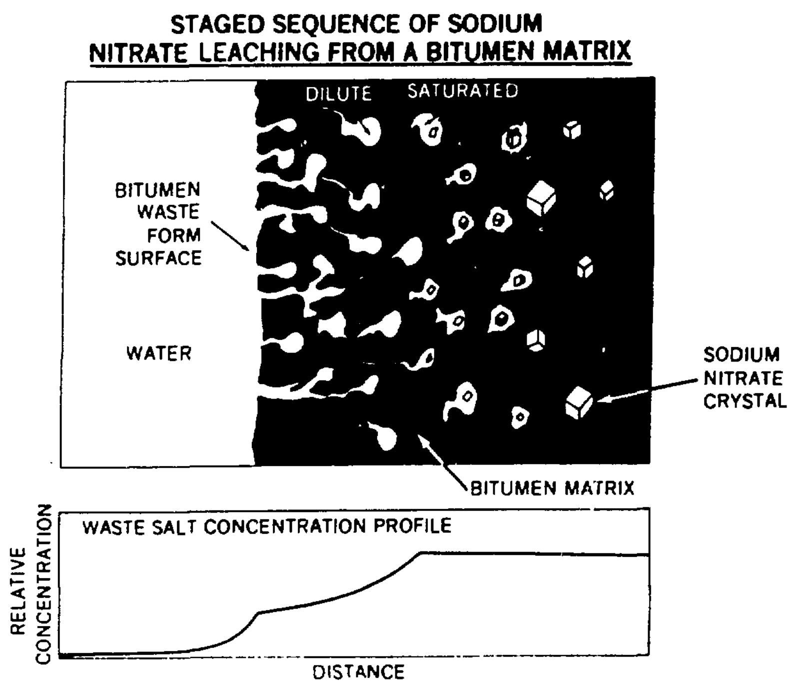

Fig.6. Proposed, staged sequence of sodium nitrate leaching from a bitumen matrix.

Fig.7. A photograph of bitumen waste forms after ninety days of leaching showing surface changes and expansion.

Fig.8. An SEM photomicrograph of a solvent-cleaned surface of a bitumen waste form revealing the immobilized nitrate salts below.