URANIUM MILL TAILINGS STABILIZATION

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ABSTRACT

Uranium mill tailings pose a potential radiation health hazard to the public. Therefore, stabilization or disposal of these tailings in a safe and environmentally sound way is needed to minimize radon exhalation and other environmental hazards. One of the most promising concepts for stabilizing U tailings is the use of asphalt emulsion to contain radon and other hazardous materials within uranium tailings. This approach is being investigated at the Pacific Northwest Laboratory. Results of these studies indicate that a radon flux reduction of greater than 99% can be obtained using either a poured- on/sprayed-on seal (3.0 to 7.0 mm thick) or an admixture seal (2.5 to 12.7 cm thick) containing about 18 wt% residual asphalt. A field test was carried out in June 1979 at the Grand Junction tailings pile in order to demonstrate the sealing process. A reduction in radon flux ranging from 4.5 to greater than 99% (76% average) was achieved using a 12.7-cm (5-in.) admix seal with a sprayed-on top coat. A hydrostatic stabilizer was used to apply the admix. Following compaction, a spray coat seal was applied over the admix as the final step in construction of a radon seal. Overburden was applied to provide a protective soil layer over the seal. Included in part of the overburden was a herbicide to prevent root penetration.

INTRODUCTION

The milling of uranium ore produces large quantities of waste (mill tailings) which remain potentially hazardous for a very long time due to the long half-lives of the radionuclides present. Two potentially hazardous radioactive decay products are radium-226 (half-life 1620 years) a solid, and radon-222 (half-life 3.8 days), a radioactive gas which is considered to present the most significant exposure risk.

Based on projected U.S. nuclear generating capacity, 490 million metric tons (MT) of tailings will be produced by the year 2000 using conventional milling.[1] These tailings would be in addition to the 107 million MT of tailings at currently active mill sites at the end of 1977 and 24 million MT of tailings at inactive sites. Because of potential radiation health hazard to the public, methods to stabilize or dispose of the tailings in a safe and environmentally sound way are needed in order to minimize radon exhalation and minimize other environmental hazards.

Proposed requirements for uranium tailings disposal include placing no less than 3 m (10 ft) of cover material over the tailings.[1] This cover material must not include mine waste or rock that contains elevated levels of radium. This technique might minimize human exposure from inhalation and ingestion, but it is not considered a totally satisfactory solution based on economics and the availability of cover material. Other techniques include the use of clay liners and covers with a soil cover. Placement of tailings in abandoned underground and open pit mines is also being considered.

An alternative approach would be to apply a cost-effective cover material that would reduce radon exhalation to background levels and remain stabile for at least 1000 years. The Pacific Northwest Laboratory (PNL) is working on such an alternative.[2,3,4,5] The Department of Energy has contracted PNL to evaluate the use of asphalt emulsion sealants to retain radium and other hazardous elements in uranium tailings and to provide a barrier over the tailings to prevent radon exhalation to the atmosphere. Figure 1 illustrates the general concept of stabilizing or sealing a tailings pile above or below grade using the asphalt emulsion sealing procedure.

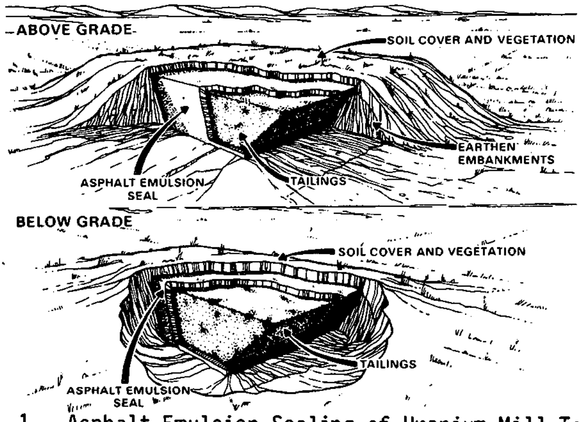

Fig. 1. Asphalt Emulsion Sealing of Uranium Mill Tailings

In order for a stabilization or sealing material to last for millenia it must be inert, remain pliable, and not be affected by its surrounding environment. Since no materials have been tested for greater than 19 to 100 years, we cannot provide long-term stability data.

We do know that asphalt, the primary constituent of asphalt emulsion, is present in very old (4800 to 5400 years) ornaments, figurines, and statues. Many ceremonial objects have been excavated and recovered in excellent condition, attesting to the potential long-term stability of asphalt under anaerobic burial conditions.

The use of cationic asphalt emulsion to retain radon and other potentially hazardous materials within uranium tailings is being investigated in the laboratory and in field tests. Laboratory studies include uranium tailings characterization, asphalt emulsion formulation, radon diffusion measurements, assessment of seal stability, and techniques to prevent plant or animal intrusions. The field studies include reviewing and evaluating application technology and conducting field tests using the most promising application technology to apply an effective seal. The effectiveness of the asphalt emulsion seal to contain radon is being established by monitoring radon exhalation with time. The stability of the seal is being evaluated to determine the effects of mechanical abuse, root penetration, and chemical environment.

This paper discusses the progress of this project, including laboratory and field studies, and summarizes the status of the sealing procedure for controlling radon release from uranium tailings. The costs of potential tailings seals are also discussed.

ASPHALT EMULSION

Asphalt emulsion consists of asphalt, water, and an emulsifier (the surface-active agent or surfactant [a]) which are

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a Surfactants possess the unique property of altering the surface energy of their solvents, usually lowering rather than increasing the surface energy. Surface-active chemicals are soluble substances that markedly change the properties of their solvents and the surfaces they contact. The three basic types of chemical surface-active agents are classified according to their dissociation characteristics in water: anionic, nonionic, and cationic surfactants.

combined together in a colloid mill to form a homogeneous mixture of small asphalt droplets suspended in water.[5] The quality of asphalt and water used to make the emulsion are very important. However, the most important component of any asphalt emulsion is the emulsifier.

To be an effective emulsifier for asphalt, the surfactant must be water soluble and must possess a proper balance between hydrophilic and hydrophobic properties. When used in combination with an acceptable asphalt, a good quality water, and adequate mechanical mixing, the emulsifier is the major factor which influences initial emulsification, emulsion stability, and ultimate field performance.

The asphalt emulsions considered for sealing tailings are cationic asphalt emulsions which have positively charged droplet surfaces. The positively charged surface of the asphalt droplets adheres to the negatively charged tailings as shown in Fig. 2. The surface charge (zeta potential) of cationic asphalt emulsions ranges from +12 to +130 mV. The choice of cationic emulsion depends on the surface area (particle size distribution) present in the material to be sealed. Different particle types, and sizes require different choices of cationic emulsion in order to obtain the proper bonding, set time, and penetration.

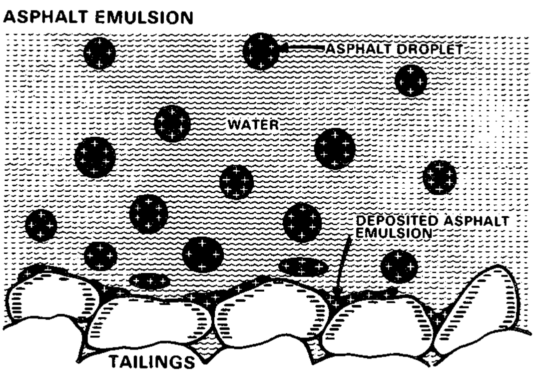

Fig. 2. Asphalt Emulsion Deposition on Uranium Tailings

LABORATORY STUDIES

The overall objective of the laboratory studies is to investigate various asphalt emulsion sealants to contain radon and other potentailly hazardous materials like radium within the uranium tailings. Characterizing uranium tailings, formulating the seal, and measuring radon diffusion are the primary activities.

Uranium tailings are analyzed for physical and chemical characteristics that could have influence on formation of a radon-tight seal including particle size, chemical composition, pH, and moisture content.

Particle size (and thus surface area) of the tailings varies widely and can have a significant effect on seal formation. For example, the high-surface-area materials (slimes) contain considerable clay-like material which is very difficult to coat with the emulsion. Also, some tailings contain a very narrow range of particle sizes, which makes compaction a potential problem.

Moisture content and pH have a direct effect on the bonding mechanism. For example, very dry soil can dehydrate the emulsion prematurely.

Laboratory Radon Measurements

Radon diffusion measurements are performed to determine the effectiveness of various asphalt emulsions in producing a radon seal. The primary system used to test the seals is shown in Fig. 3. Radon gas is passed below the seal in addition to the radon generated by the tailings. This allows for a greater concentration of radon below the seal and if required a pressure can be applied to the bottom of the seal. Radon diffusion measurements are usually run for up to two weeks unless a major leak occurs. Any radon that diffuses through the seal is adsorbed on activated carbon which is immersed in a dry ice-alcohol bath to improve its adsorption efficiency to greater than 99%. After the specified test time, the carbon is removed, sealed and counted. The radon content of the carbon is indirectly determined by counting the gamma activity of the Bi-214 daughter product. From this data the radon fluxes are determined.

Examples of radon flux reductions obtained using poured-on seals are tabulated in Table I. Cationic asphalt emulsion prepared with Armak Co. Redicote E-63, E-65, and E-4868 emulsifiers was used to make the seals.

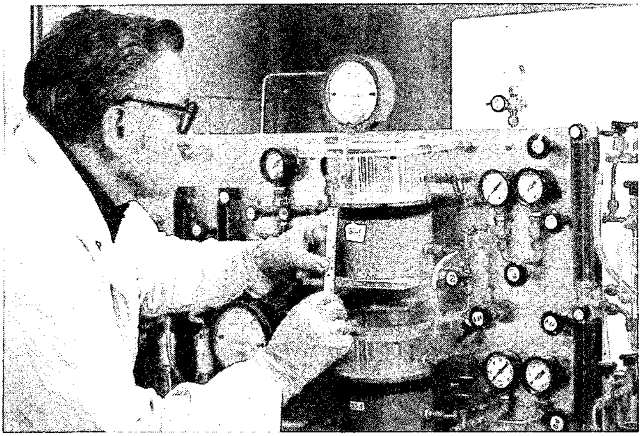

Fig. 3. Operation of Radon Diffusion Test Apparatus

Table I. Radon Flux Reduction Using Poured-On Asphalt Emulsion Seals

Sample	Seal Thickness, mm	% Flux Reduction
Vitro-Site at Salt Lake City, UT	9	99.9%
Ambrosia Lake, NM	9	99.9%
Mexican Hat, UT	6	99.3%
Monument Valley, AR	9	99.8%
Shiprock, NM	9	99.9%
Tuba City, AR	9	99.9%
Falls City, TX	9	99.9%
Grand Junction - 1	9	99.9% Pressurized Seal
Grand Junction - 3	9	99.9% Pressurized Seal
Grand Junction - 4	9	99.9% Pressurized Seal

All flux reductions were greater than 99.3%. However, poured-on seals are not adequate for the field since they do not have enough mechanical stability. Therefore, admixtures of Grand Junction tailings and asphalt emulsion were tested using the primary radon diffusion test apparatus. Admixtures containing from 10 to 20 wt% residual asphalt were compacted at about 5.6 kg/cm^2 (80 psi) and tested. Admixtures containing 10- to 12-wt% residual asphalt stopped neither water vapor or radon; a 12- to 14-wt% residual asphalt admix only sealed out water vapor. Starting at 14-wt% residual asphalt, a marked reduction in radon flux is noted. At a 16- to 20-wt% residual asphalt content, greater than 99.9% reductions in radon fluxes were obtained.

FIELD STUDIES

The overall objective of the field studies is to demonstrate the effectiveness of a stabilization or sealing procedure using asphalt emulsion to contain radon. Techniques for applying the asphalt emulsion seal are being investigated at selected tailings sites. The objectives of the field tests are to obtain sufficient operating data to evaluate the technical and economic feasibility of the most promising application techniques.

During June 1979, a field test was conducted at the Grand Junction tailings site. The primary emphasis of this test was to test a stabilization procedure consisting of site preparation (contouring, watering, compacting), asphalt emulsion application using a hydrostatic stabilizer and compactor to form the seal, and overburden application.

A 82.5-m x 82.5-m (275-ft x 275-ft) site was prepared for the sealing test by removing a 5-cm to 15.2-cm (2- to 6-in.) cover of overburden that contained too much clay to make an adequate seal, then contouring the site for drainage, watering, and compacting.

Following some preliminary seal application equipment tests, a total of 4328 m^2 (1.07 acre) was sealed using a BOMAG MPH100 Hydro-static Stabilizer as shown in Figs. 4 and 5. Cationic asphalt emulsion prepared with Armak Co. Redicote E-4868 emulsifier was used for the admix seal. A few hours after the emulsion was applied, the tailings-emulsion mixture was compacted using a vibratory compactor to form an admixture seal.

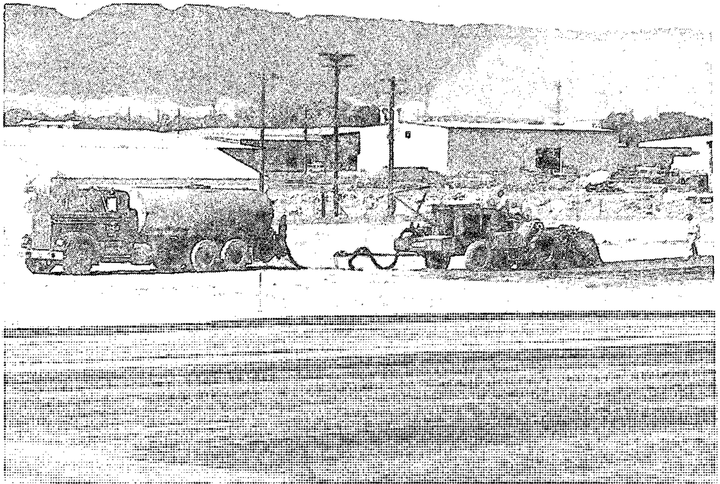

Fig. 4. BOMAG MPH100 Hydrostatic Stabilizer and Distributor Truck Applying Asphalt Emulsion to Tailings

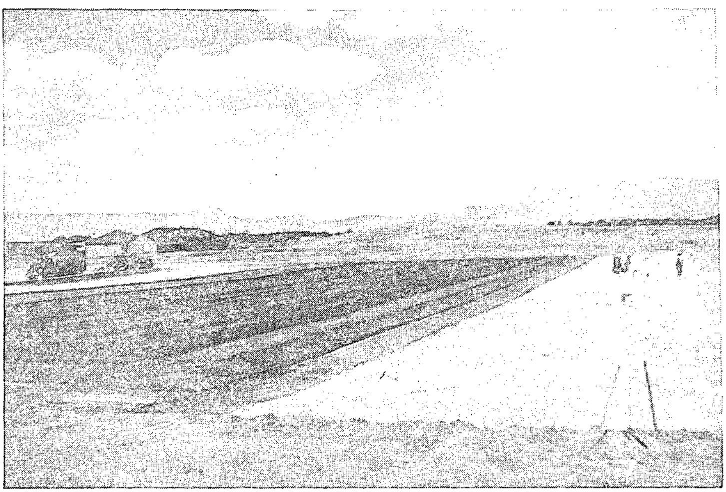

Fig. 5. Application of Asphalt Emulsion to Tailings

Several application rates were tried, including a double-pass application where about 8-wt% residual asphalt was applied on the first pass followed by an additional 8-wt% residual asphalt on the second pass.

After application and after the majority of the water evaporated, the admix was compacted 4 to 6 times using a vibratory compactor to form the admixture seal. The next step was to apply a 3.2-l/m^2 (0.7-gal/yd^2) spray coat seal (Fig. 6) to fill any micro-cracks that might have formed during compaction. We've found that a well-compacted surface is important to control admixture depth thereby maintaining consistent seal asphalt content. Water application makes the tailings easier to compact. Water application over the admixture seal is necessary for proper bonding between the admixture seal and the spray coat seal.

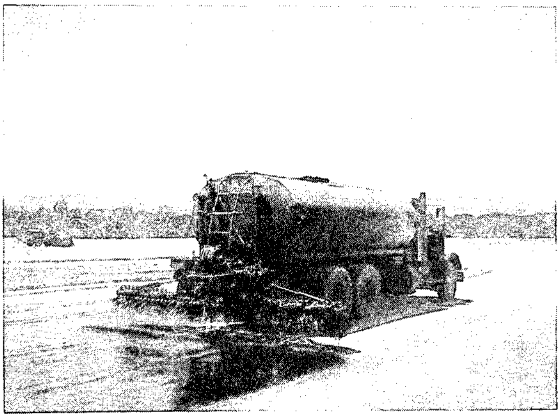

Fig. 6. Asphalt Emulsion Spray Coat Seal Application over Admixture Seal

Equipment modifications to both the hydrostatic stabilizer and distributor truck will be investigated during 1980 along with alternative application techniques such as a pug mill/paver and slurry sealer.

Field Radon Flux Measurements

The effectiveness of the asphalt emulsion seal was determined by measuring radon exhalation from the test area before and after the seal was applied. The system used for the radon measurements is illustrated in Fig. 7 and pictured on the field test site in Fig. 8.

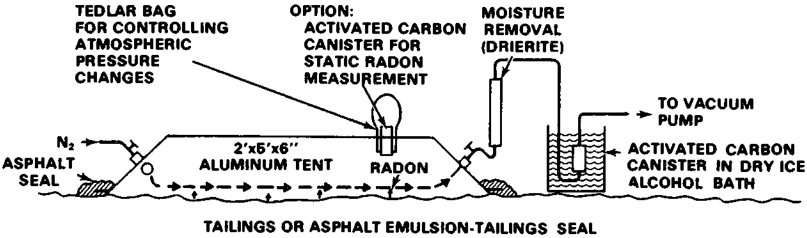

Fig. 7. Diagram of the Radon Measurement System Used for Field Tests

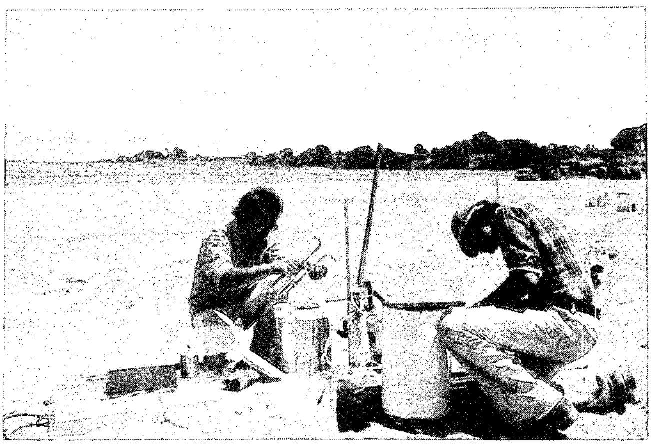

Fig. 8. Radon Measurement System Used at Grand Junction Field Test

Nitrogen passed over the tailings or seal picks up the radon and carries it to an activated carbon canister that is kept in a dry ice/alcohol bath. After a specified time, the activated carbon is removed, allowed to reach equilibrium and counted. Radon fluxes are determined from this data.

Radon flux from the Grand Junction tailings pile test site ranged from 12 to 2400 pCi/m^2-s. The average flux was 270 pCi/m^2-s, while the mean was 73 pCi/m^2-s. The large range of fluxes is due to the high radium content of the slimes compared to the sand.

Radon fluxes after the seal was applied were reduced by 4.5 % to 99.9% with average flux reduction of 76%. Considering that we did not obtain the residual asphalt required for a total seal (18 to 20 wt%), the test was a success. In most cases, areas covered with fog seal were sealed. Areas where the seal failed indicated an average 104% increase in flux. This is caused by the lateral diffusion of radon to a crack or breach in the seal; however, this increase in flux would not have a significant effect on overall radon concentration above the seal.

CONCLUSIONS

The general conclusions that can be made as a result of both laboratory and field tests are as follows:

- Cationic asphalt emulsion can be used effectively to stop radon exhalation from uranium tailings by either pouring/spraying on or admixing with the tailings. Proper selection of the emulsion depends on the physical-chemical properties of the tailings or soil to be sealed.

- Admixture seals can be potentially applied to tailings piles using conventional stabilizing equipment with some modifications.

- Both laboratory and field tests indicate the potential to reduce radon exhalation to below the proposed EPA standard of 2 pCi/m^2 sec (annual average flux). Field test radon reduction at the Grand Junction tailing test site averaged 76%.

- An admix containing about 20-wt% residual asphalt is required to achieve a total seal.

- Laboratory cyclic freeze/thaw tests (0 to 22°F) did not affect seal integrity in poured-on seals 3.0 to 7.0 mm thick.

- Exposing laboratory seals to 10^5-R gamma radiation did not have any effect on seal integrity.

- Long-term stability of the seal is affected by the nature of the environment surrounding the seal. For example, if the seal were exposed to sunlight, degradiation would occur and the seal would not last for a thousand years.

- Maintaining overburden over the seal provides erosion control. Revegetation or a rip-rap (rock) cover could be used to prevent soil erosion. Addition of selected herbicides can control root growth and prevent seal penetration.

- In order to meet the proposed EPA standard of 2 pCi/m^2-sec annual average flux a radon reduction of greater than 99% is required for most U tailings.

REFERENCES

1. U.S. Nuclear Regulatory Commission. 1979. Draft Generic Environmental Impact Statement on Uranium Milling, NUREG-0511 Vol. 1.

2. Koehmstedt, P. L., Hartley, J. N., and Davis, D. K. 1977. Use of Asphalt Emulsion Sealants to Contain Radon and Radium in Uranium Tailings. BNWL-2190, Pacific Northwest Laboratory, Richland WA.

3. Hartley, J. N., Koehmstedt, P. L., and Esterl, D. J. Aspahalt Emulsion Sealing of U Mill Tailings. PNL-SA-8125, Presented at the Second Symposium on U Mill Tailings Management, Fort Collins, CO, November 19-20, 1979.

4. Koehmstedt, P. L., Hartley, J. N., Esterl, D. J. Asphalt Emulsion Sealing of U Mill Tailings. PNL-SA-7736, Presented at the Materials Research Society International Symposium on the Scientific Basis for Nuclear Waste Management, Boston, MA, November 27-30, 1979.

5. Hartley, J. N., Koehmstedt, P. L., and Esterl, D. J. 1979. Asphalt Emulsion Sealing of Uranium Mill Tailings, (In Progress) Pacific Northwest Laboratory, Richland WA.