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3,298,961

CONCENTRATION AND CONTAINMENT OF RADIOACTIVITY FROM RADIOACTIVE WASTE SOLUTIONS IN ASPHALT

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No Drawing. Filed Aug. 26, 1965, Ser. No. 482,950
2 Claims. (Cl. 252—301.1)

The present invention relates to the disposal of radioactive waste solutions. More particularly, it relates to, and has for a principal object to provide a process for the concentration and containment of radioactivity from intermediate level radioactive waste solutions. The invention described herein was made in the course of, or under, a contract with the U.S. Atomic Energy Commission.

For the purposes of this invention, an intermediate level waste solution is one which contains radionuclides in excess of the maximum permissible levels as established by the International Commission on Radiological Protection, provided the total radiation emanating from such radionuclides is not sufficient to adversely degrade asphalt.

Public health and safety considerations prevent release of the majority of radioactive waste solutions to the environment without first assuring that the radiotoxicity is within biologically acceptable values. Within this framework, a simple, inexpensive, and effective method is needed to concentrate and to retain as a stable insoluble solid the radionuclide content of large volumes. As a practical matter, any process for treating radioactive waste solutions to remove radionuclides and other salts usually results in a solution which is still radioactive, but contains lesser amounts of radionuclides. The success of any method of treating and handling radioactive waste solutions is measured by (a) the degree to which the radionuclides are reduced in the treated solution, and (b) the effectiveness of the containment of the separated nuclides, as well as (c) the economics of the total process.

It is, therefore, an object of this invention to provide a process of treating an intermediate level radioactive waste solution to effectively concentrate and contain the radionuclide and salt content wherein said process is characterized by its simplicity, ease of handling, and overall economy.

The objects of this invention are met by a process which comprises contacting an intermediate level radioactive waste solution, as previously defined, with an emulsified asphalt composition, evaporating the water in the resultant mixture at a temperature no greater than about 160° C., maintaining the temperature of the dry asphalt product at a flow temperature, and then flowing the resultant asphalt composition into a storage container.

To place the present invention in its proper content, it should be noted that the concentration and containment of radionuclides of radioactive sludges with asphalt is known. A sludge is an aqueous precipitate of solid particles which contains the radioactivity. The water content of sludges is essentially non-radioactive. The object in using asphalt in such a case is to reduce the volume of the sludge by removing its water content in order to contain the radioactive precipitated phase within the volume of the asphalt. In that sense, the process is similar to the one described here. However, the water content of a viscous sludge-asphalt mixture can only be removed by heating said mixture to a temperature in excess of 200° C. while vigorously agitating the mixture. Moreover, water removal is accompanied by sputtering which adds to the technical difficulty and increases the safety hazards. In addition to this, water removal at these high tempera-

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tures presents an ever-present danger of carrying over excessive volatile radionuclides with the steam distillate. In contrast to this, the process of the present invention contemplates removal of water and containment of radionuclides from a soluble mixture of a radioactive solution with an emulsified asphalt composition, heating the mixture with nominal stirring to evaporate the water and finally raising the temperature to about 160° C. Under these conditions, water removal by evaporation takes place at a temperature near the boiling point of the solution. As the water is removed, the emulsion is destroyed and the particles of asphalt begin to agglomerate. However, before complete agglomeration has taken place, substantially all of the water will have been removed under the relatively mild conditions described without violent sputtering.

After the water is removed, the resultant asphalt product will be found to contain virtually all of the solid content originally in solution, including the radionuclides, within a relatively small asphalt volume. In some cases, the collected aqueous condensate will be found to contain so little radioactivity as to be safely discharged into soil environments. On the other hand, where the radionuclide content is above biologically safe threshold values, previous treatment with the emulsified asphalt still constitutes an effective process treatment because the aqueous condensate is now in condition for further treatment by standard ion exchange adsorption or precipitation means to remove substantially all of the radionuclide content.

After water removal, the asphalt is maintained at a temperature at which it will flow freely from the evaporator. Most asphalt compositions containing up to 60% solids resulting from the treatment of radioactive waste solutions will flow freely from the evaporator at 130° C. If the solid content is appreciably greater, higher temperatures are needed to obtain a free-flowing product. For example, an asphalt loaded with 80% by weight solids will not flow readily until a temperature of 195° C. is reached.

The asphalt emulsion compositions useful for the purposes of this invention are commercially available products which are normally used in the maintenance and construction of roads and pavement. More particularly, the asphalt paving emulsions useful in removing and containing radionuclides from an aqueous solution are those defined in the United States Government Federal Specification SS-A-674b, dated November 25, 1965, and SS-A-00674c dated August 20, 1962, as issued by the Materials Specification Department of the General Services Administration Agency of the United States Federal Government.

The emulsified asphalt compositions useful for this invention may vary widely within the framework of the aforementioned specification in order to attain the objects of this invention. It is most desirable to use an emulsified asphalt which yields a resilient product at room temperature after it has been loaded with solids. A satisfactory product for practicing our process is that asphalt emulsion defined in the aforementioned Federal specifications as type RS-2, a rapid setting, high viscosity emulsion, a composition normally used in the surface treatment of roads. This type of asphalt emulsion contains about 63 weight percent asphalt, 2% emulsifying agent, and the remainder water.

Having defined our invention in general terms together with operating parameters, the following embodiment is provided by way of example, and is not to be construed as limiting the scope of our invention.

EXAMPLE

In the course of separating nuclear fuel values by liquid-liquid solvent extraction techniques derived from the dis-

solution of spent nuclear fuel elements, large volumes of aqueous stripping solutions are generated. The strip solutions contain large amounts of salting out agents, as well as fission product radionuclides. Such strip solutions are generally classified as intermediate waste solutions, as previously defined, and constitutes a major problem of disposal. The process of this invention is eminently suitable for use in removing and containing the major percentage of inorganic salts and radionuclides from such strip solutions into a biologically safe and easily handled asphalt mass, as will be hereinafter described.

One pound of an RS-2 emulsified asphalt, as previously defined, was mixed with mild stirring with 2.67 pounds of an intermediate waste solution in an evaporator unit containing means for collecting an aqueous condensate. After evaporating the water, the resultant mixture was heated to a temperature of 160° C. The aqueous condensate was collected during evaporation. At the end of this time all of the water had evaporated and a solid viscous mass of liquid asphalt containing solids remained in the evaporator. The asphalt was poured from the evaporator at a temperature of 130° C. into a container and allowed to cool. The following data represents a material balance breakdown of a representative run conducted in accordance with this invention.

Int. Level Waste	Asphalt	Product	Vapor
Salts—38.5%-----	-----	60-62%-----	-----
Water—61.5%-----	35%-----	-----	100%-----
Asphalt-----	65%-----	38-40%-----	-----

EVAPORATOR FEED

	Inter- mediate Level Waste	RS-2 Asphalt	Products	
			Aqueous Condensate	Asphalt Product
Vol., ml.-----	300-----	163-----	300-----	154-----
Density, g./cc.-----	1.282-----	0.96-----	1.00-----	1.50-----
Total weight, gm.-----	385-----	156-----	300-----	230-----
Sodium-----	130 g./l.-----	-----	-----	16.96 w. per cent.-----
Aluminum-----	4.62 g./l.-----	-----	-----	0.60 w. per cent.-----
Sulfate-----	28.0 g./l.-----	-----	-----	3.65 w. per cent.-----
Chloride-----	1.71 g./l.-----	-----	-----	0.22-----
Hydroxide-----	37.9 g./l.-----	-----	-----	4.94 w. per cent.-----
Nitrate-----	226 g./l.-----	-----	-----	29.48 w. per cent.-----
Water-----	853.77 g./l.-----	336 g./l.-----	1,000 g./l.-----	44.22 w. per cent.-----
Asphalt plus emulsifier-----	-----	624 g./l.-----	-----	9.39 curies per gal.-----
Total Activity-----	4.82 curies per gal.-----	-----	2.81×10 ⁻⁵ curies/gal.-----	-----

RADIOACTIVITY BREAKDOWN

	Percent In Feed	Aqueous Condensate, Percent
Cs-137-----	99-----	99.9-----
Ru-106-----	1.0-----	0.1-----

Others:

Volume reduction

$$\frac{\text{volumes of waste solution}}{\text{volume of asphalt product}} = 1.95$$

Material balance

In:

In. level waste-----	g-- 385
Emulsified asphalt-----	g-- 156
Total-----	g-- 541

Out:

Condensate-----	g-- 300
Product-----	g-- 230
Total ¹ -----	g-- 530

$$\frac{530 \text{ g. In}}{540 \text{ g. Out}} \times 100 = 98.2\%$$

Product composition

Salts-----	G. 128.5
Asphalt-----	101.5
Total-----	230

$$\frac{\text{g. Salt Loading}}{\text{g. Asphalt}} = \frac{128.5}{101.5} = 1.27 \frac{\text{g. Salt Loading}}{\text{g. Asphalt}}$$

$$\frac{128.5 \text{ g. Salt}}{230 \text{ g. Product}} \times 100 = 55.9\% \text{ salts}$$

¹ Balance of material is that coating the surfaces of the evaporator and stirrer.

The asphalt product, as shown in the above data, contains 60% solids and essentially all of the inorganic salt content of the feed solution. This asphalt product was then leach tested in static water over a period of several months by examining the radionuclide content of the leachant collected at intervals of time during this period. The leaching solutions were tested for their cesium-137 and ruthenium-106 content, and showed that the average leach rate for cesium-137 was about 5×10^{-4} g./cm.² of exposed surface per day; and for ruthenium-106, the average leach rate was 7×10^{-6} g./cc. per day. It was found that essentially all of the inorganic salts as well as the Cs-137 and Ru-106 content was incorporated into the asphaltic mass, as shown in the material balance data above. While the aqueous condensate could not be discharged directly into natural soil or streams, standard treatment by flow through readily available ion exchange resins such as strong acid phenolic resin will easily remove sufficient radionuclide content to provide a biologically safe aqueous effluent.

This example demonstrates the value of the process in treating radioactive waste solutions containing large amounts of inorganic salts to concentrate and contain the major percentage of the solids content in a readily available asphalt material. The solid asphalt product is an effective container for the solids and the aqueous effluent is now in condition for disposal to soil or natural stream environments, or easily converted to a biologically safe solution by standard ion-exchange or complexing techniques. While the utility of the process has been demonstrated for treatment of a solution which is heavily loaded in inorganic salts, the advantages of the process will be realized in treating any intermediate level radioactive waste solution.

Having thus described our invention, we claim:

1. A method of concentrating and containing the solid content of an intermediate level radioactive solution which comprises mixing said solution with emulsified asphalt, evaporating essentially all of the water content of said mixture at a temperature no greater than 160° C., and then flowing the resultant asphalt composition into a container to solidify in a desired configuration.

2. A method of concentrating and containing the solid content of an intermediate level radioactive solution which

comprises mixing said solution with emulsified asphalt, evaporating essentially all of the water content of said mixture at the boiling point of the said mixture, and then flowing the resultant asphalt composition into a container to solidify in a desired configuration.

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