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CHEMICAL BEHAVIOR AND GROUND MOVEMENT OF SELECTED RADIONUCLIDES

RADIOACTIVE WASTE

KEYWORDS: *HAPO, radioactive waste disposal, radionuclide migration, sediments, radioactive waste storage, tanks, leaks, ground water, liquid wastes, solidification*

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Waste management practices at Hanford are based on 30 years of experience and special studies. Aqueous high-level wastes are being converted to salt cakes in underground tanks to reduce the potential for loss of liquid high-level radioactive waste due to tank failure. If wastes enter the ground they are sorbed in the Hanford sediments and become fixed in place by natural processes. Water from the equivalent of a thousand years of rainfall in one deluge is not likely to move the radioactive materials such as plutonium, strontium, and cesium to the water table.

The radioactive waste management program at Hanford has resulted in extensive study regarding the chemical behavior and ground movement of radionuclides in Hanford sediments. The major features of the waste solidification program for Hanford high-level waste are shown in Fig. 1. The stored liquid waste is processed to remove most of the long-lived heat-emitting isotopes, ^{90}Sr and ^{137}Cs.

The removal of these isotopes is not complete but is carried out to reduce the heat output in the salt cake so that it can be safely stored in the existing tanks with no adverse impact on the containment structure. The strontium and cesium will be doubly encapsulated and placed in water basins for interim and/or extended interim stor-

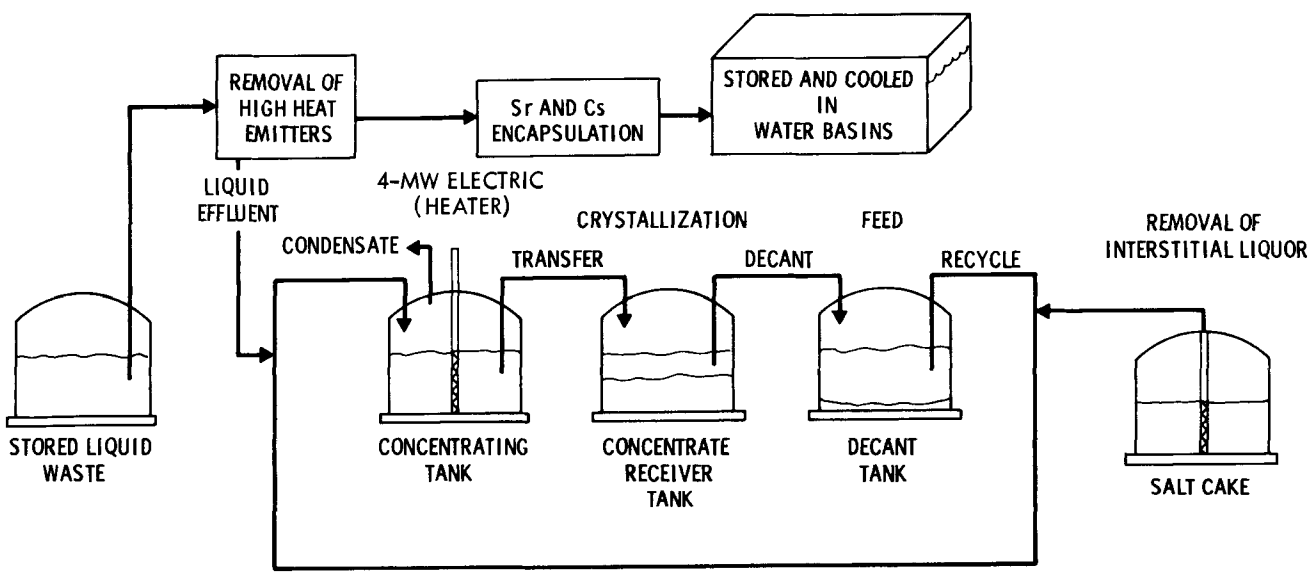


Fig. 1. Hanford high-level waste solidification program.

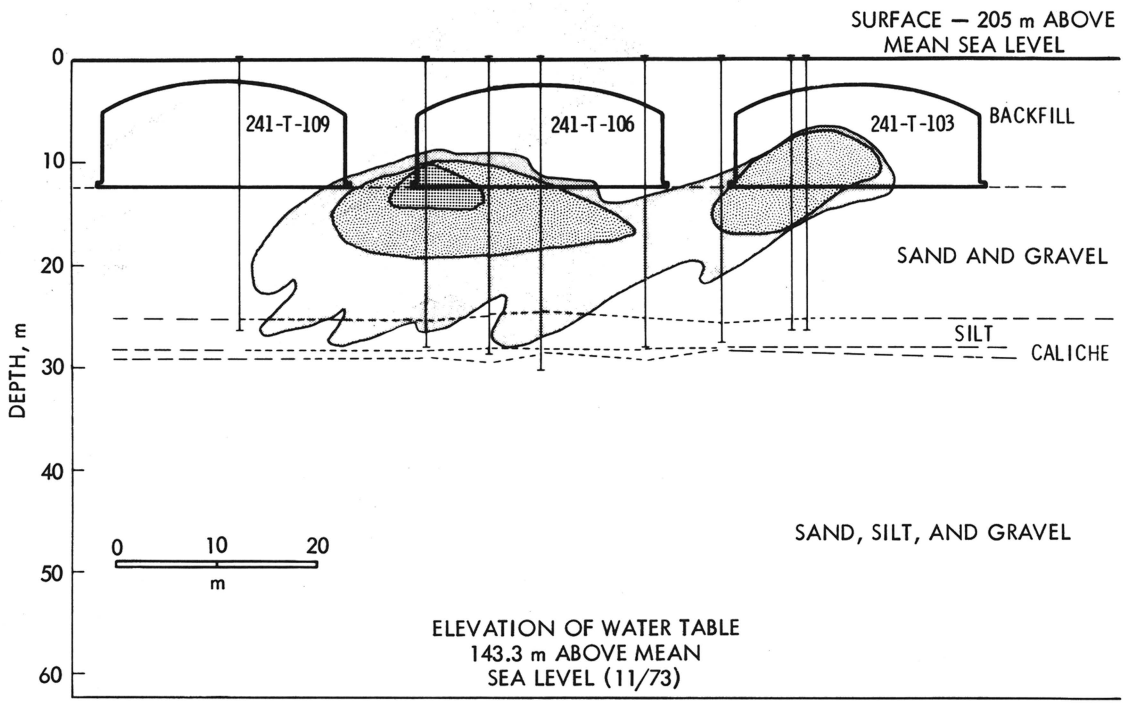
age. After the cesium and strontium are removed to the extent required, the liquid waste is then concentrated by evaporation to produce a salt cake that is much less mobile than the liquid waste. The leak that occurred in 1973 was solution that had been treated for cesium and strontium removal and was being held in storage awaiting the evaporation treatment. The waste solidification program was developed to reduce the potential for loss of liquid high-level waste due to tank failure.

The knowledge regarding the chemical behavior and movement of radionuclides in Hanford sediments is extensive. By using the knowledge available regarding the sediments under the leaking tank, the knowledge of how liquids move in these sediments under partially saturated flow conditions, and the knowledge of how the radionuclides chemically behave in the Hanford sediments, it was predicted[1] that the leak solution would be contained in the soil well above the water table. The Hanford sediments, as depicted in Fig. 2, are not uniform in character with depth; rather, they consist of sands, gravels, and silts; and, in the case of the T Tank Farm, a caliche layer. This is a deposit that is quite like concrete but was made by nature following the deposition of the sediments which took place in the distant past.

The prediction also was made that the long-lived radioisotopes, plutonium, ^{90}Sr, and ^{137}Cs, would not migrate as far as the waste liquid. Several factors were used in making this prediction.

In alkaline systems, plutonium oxide has a very low solubility. The small fraction of plutonium which remains in solution is resorbed on the sediments by ion exchange reactions. The mobility of the plutonium ions in a saturated groundwater flow system is so low that it is difficult to measure.

In these alkaline systems, strontium carbonate also has a very low solubility and a very low mobility in a groundwater flow system. Laboratory and field tests have shown that strontium moves at about 1/100th the rate of groundwater, cesium at about 1/1000th, and the plutonium ion migration rate is orders of magnitude less than cesium. The rates of sorption and desorption (ion exchange) of these radionuclides in the sedi-



Pu - PRECIPITATES AS OXIDE

Sr - COPRECIPITATES AS CARBONATE (IN HIGH CAUSTIC WASTES)

Cs - HELD ON SEDIMENTS BY ION EXCHANGE AND CHEMICAL REACTIONS

Pu, Sr, Cs - REACTS WITH SILICIC ACID TO FORM SILICATES

Pu, Sr, Cs - REACTS WITH CLAYS TO FORM ALUMINOSILICATES

Fig. 2. Tank leak distribution in sediments.

ments are dependent upon the chemical characteristics of the infiltrating solutions and the minerals present in the sediment.[2]

These nuclides are held on sediments more than one hundred feet about the regional water table and would require an amount of water equivalent to 1000 years of accumulated rainfall passing through these sediments for any of them to reach the water table.

The radionuclide distribution in the sediments from material leaked in 1973 was determined by core drilling, sampling, and analysis. The results are shown in Fig. 2 and verified the earlier prediction.[1] There are three areas identified. The smaller, inner zone shows the extent of cesium movement, the intermediate zone shows the extent of strontium movement, and the outer zone depicts the extent of liquid infiltration into the sediments based on ruthenium data. In alkaline systems, some chemical species of ruthenium, such as ruthenate and perruthenate, move with aqueous solutions. The plutonium content of the leaked material was exceedingly low, and no plutonium was detected in samples taken. In the alkaline system, plutonium is held within a few centimeters of the tank. It is worthy of note that plutonium, cesium, and strontium are more mobile in acidified sediments. At Hanford the sediments are alkaline (ph = 8 to 9.5). In the past, when acidic solutions were placed in Hanford sediments, they were neutralized by the highly alkaline sediments. The movement of radionuclides in acid solutions was thereby restricted.

In summary, under partially saturated flow conditions such as we experience with leaks, the plutonium, cesium, and strontium nuclides do not migrate large distances from the point of entrance into the soil. Continued passage of liquid through the soil does cause movement of the radionuclides; however, the rate of such movement is much less than the rate of liquid movement. This varies with the soil and the chemical characteristics of the liquid.

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