LA-7379-MS
Informal Report

UC-70
Issued: June 1978

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UNITED STATES
DEPARTMENT OF ENERGY
CONTRACT W-7405-ENG. 36

MOVEMENT OF FLUIDS AND PLUTONIUM FROM SHAFTS AT LOS ALAMOS, NEW MEXICO

by

William D. Purtymun, Raymond Garde, and Richard Peters

ABSTRACT

The movement of fluids and plutonium from wastes disposed in shafts drilled into rhyolite tuff were studied under normal and test conditions. During normal operations of a waste treatment plant at the Los Alamos Scientific Laboratory (LASL), a $Fe(OH)_3$ (ferric hydroxide) sludge is mixed with cement to form a paste and disposed into the shafts. Under these conditions there was no indication of movement of fluids into the tuff; however, there was some movement of paste into open joints that intersected the shaft. As a special test, some $Fe(OH)_3$ sludge without cement was put into an experimental shaft. In this case, fluids moved a few meters from the shaft. The fluids carried trace amounts of plutonium from the shaft, but an inventory indicated more than 99% remained adsorbed or attached to the sludge in the shaft.

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I. INTRODUCTION

Industrial waste from the LASL Plutonium Processing Facility is treated at a plant to remove plutonium and other radioactive contaminants.[1] The wastes are treated by chemical flocculation, sedimentation, and filtering processes, which produce an effluent that is released to the environment and a contaminated $Fe(OH)_3$ sludge that is mixed with a cement paste (waste-cement paste). The waste-cement paste is pumped into shafts at a controlled burial ground adjacent to the treatment plant.[2] These shafts are generally 2.4 m in diameter and range from 4.6 to 19.8 m deep.

The burial ground is located on the Pajarito Plateau, which was formed by a series of ashflows and ashfalls of rhyolite tuff.[3] At the site, the tuff is about 240 m thick and is underlain by a fanglomerate composed of cobbles and boulders in a matrix of sand and gravels. The main aquifer of the Los Alamos area lies at a depth of 350 m within the fanglomerate and there are no known perched aquifers above it.[4]

This study was conducted in two parts: (1) to determine the movement of fluids (water containing treated chemicals) and radioactive contaminants from a shaft filled with waste-cement paste under normal operating conditions, and (2) to determine the movement of fluids and plutonium under test conditions from $Fe(OH)_3$ sludge released into a smaller (60-cm-diam) shaft without the addition of cement. The second part of the study was experimental as sludge is not normally released or disposed of in this manner at LASL.

II. MOVEMENT OF FLUIDS AND RADIONUCLIDES FROM A WASTE CEMENT PASTE FILLED SHAFT

Test holes (TH) 10 cm in diameter and 29.6 cm deep were drilled at the shaft area in early 1968, prior to the development of the disposal field, to establish the geology and hydrology of the tuff. The shafts are located in the 18-m-wide berm between seepage ponds, which were used for effluent disposal in the late 1940s and early 1950s. Normally, the fluid content of the tuff in undisturbed areas ranged from 4 to 6% by volume.[5,6] However, because of infiltration of effluents from the ponds, the fluid content of the tuff in cuttings from the test holes, drilled prior to the development of shafts, ranged from 7 to 12% by volume.

A shaft, located about 60 cm north from TH-7, was drilled to a depth of 17.7 m and filled with contaminated cement paste and capped by April 1969. The cement-paste volume released into the shaft was 85 m^3, of which about 28 m^3 was fluids. Test hole TH-7A, located about 60 cm from the eastern edge of the shaft, was completed after the shaft was filled. Cuttings from both test holes were analyzed for fluid content and gross-alpha and -beta activity.

The fluid content of cuttings from the test holes ranged from 8 to 14% by volume in the upper 3 m, reflecting the infiltration of fluids from precipitation. Prior to use of the disposal shafts, the fluid content of the tuff at TH-7, from a depth of 3 to 29.6 m, ranged from 7 to 10% by volume. After the shaft was filled, the fluid content of the tuff at TH-7A, from a depth of 3 to 24 m, ranged from 4 to 8% by volume, showing a decrease in the fluid content of 3 to 6% (Fig. 1). Below a depth of 24 m there was no significant change in fluid content of the tuff. The decrease in the fluid content of the tuff adjacent to and below the bottom of the shaft is apparently caused by the hydration of the cement. During the hydration process of the waste-cement paste, fluid was extracted from the tuff.

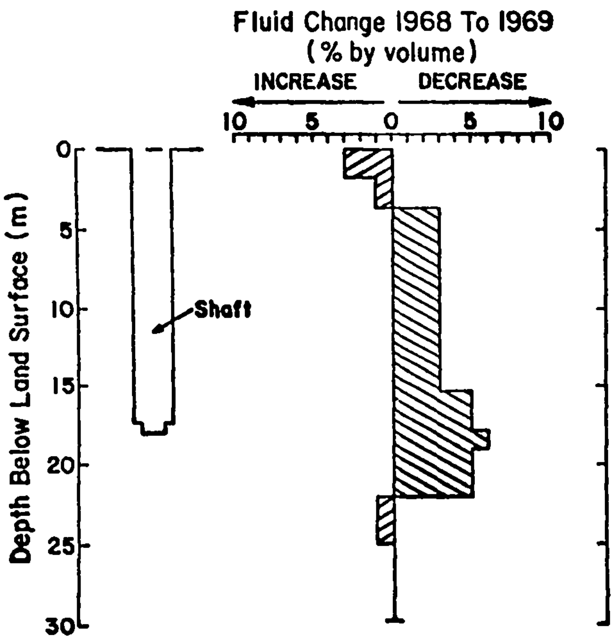

Fig. 1.
Change in fluid content of tuff adjacent to shaft prior to and after shaft was filled with cement paste.

The gross-alpha activity of cuttings from TH-7 ranged from 2 to 5 pCi/g, reflecting naturally occurring radioactive isotopes. There was no significant change in the activity in cuttings from TH-7A, which was drilled after the shaft was filled with contaminated cement paste.

Gross-beta activity ranged from <1 to 3 pCi/g in cuttings from TH-7 (1968). Activity in cuttings from hole TH-7A (1969) indicated only one significant change; the sample from the interval 23.5 to 25.0 m contained gross-beta activity of 40 pCi/g. The activity was lower, by a factor of 10 or more, above and below that depth interval. This is attributed to the test hole intersecting an open joint connected to the shaft, which was filled with the cement paste from an adjoining shaft. Such cement-paste filled joints have been observed when other shafts have been drilled adjacent to shafts that were previously filled and capped.

III. MIGRATION OF FLUIDS AND PLUTONIUM FROM SLUDGE

A small volume, 2.9×10^3 ℓ, of $Fe(OH)_3$ sludge was used to study the movement of fluids and plutonium in the tuff. This study is also applicable in evaluating the containment of similar sludge (dewatered to 35% solids) that is packaged in 208-ℓ drums and stored or buried in disposal pits dug into the tuff.

A 60-cm-diam, 6.1-m-deep shaft was used for this study. Four test holes (X series), 12.2 m deep, were drilled adjacent to the shaft (Fig. 2). The fluid content and plutonium concentrations from the cuttings were determined as baseline data. The test holes were later used to monitor the movement of fluids for this study. The subsurface fluid measurements were made by neutron logging equipment prior to and after each disposal event. About nine months after the study began, three test holes (C-series) were cored to a depth of about 11 m to recover cores for analyses to evaluate the movement of plutonium from the sludge.

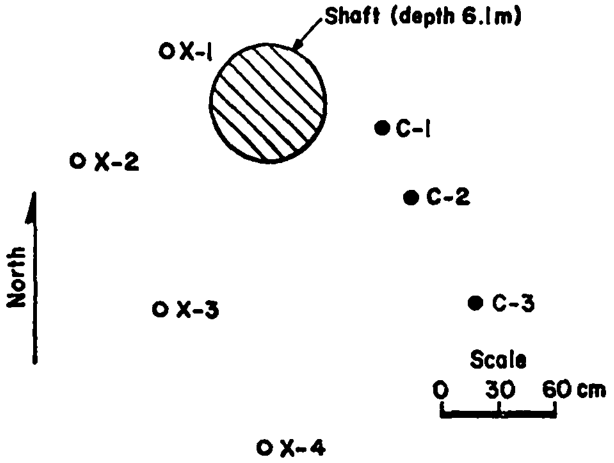

Fig. 2.
Plan view of shaft and test holes, X-series for monitoring fluid contents of tuff and C-series cored for plutonium data.

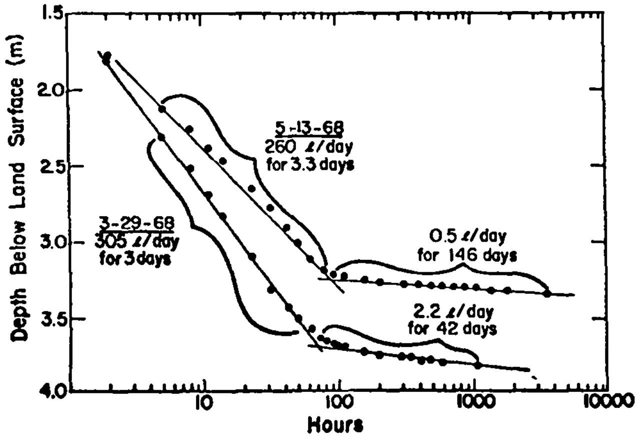

Fig. 3.
Fluid level declines in shaft receiving sludge on March 29 and May 13.

A. Movement of Fluids

About 1.5×10^3 ℓ of sludge, with a fluid content of 82%, were pumped into the shaft on March 29, 1968. The rate of sludge-level decline, measured with a continuous water-stage recorder, was about 305 ℓ/day for the first three days and about 2.2 ℓ/day for the remaining 42 days (Fig. 3). Twenty-eight days after the study began, the fluids had moved outward from the shaft 1 m and downward about 2 m (Fig. 4). The maximum fluid content of the tuff, about 30 cm from the shaft, increased to about 12% by volume from a pretest measurement of 4 to 6% by volume.

An additional 1.4×10^3 ℓ of sludge, with a fluid content of about 83%, were pumped into the shaft on May 13, 1968. The rate of sludge level decline was about 260 ℓ/day for 3.3 days, then sharply decreased to 0.5 ℓ/day for the remaining 146 days the recorder was in operation (Fig. 3). Seven days following the second release of sludge into the shaft, the fluids had moved outward from the shaft 1.5 m, while the downward movement remained at about 2 m. The fluid content of the tuff 30 cm from the shaft increased from about 12 to 16% by volume.

Fine particles in the sludge in the first release filled pore space in the tuff in the walls and the bottom of the shaft (Fig. 4). As a result, the fluid loss rate was slower during the second release and the accumulation of sludge at the bottom of the shaft caused the fluids to move horizontally from the shaft.

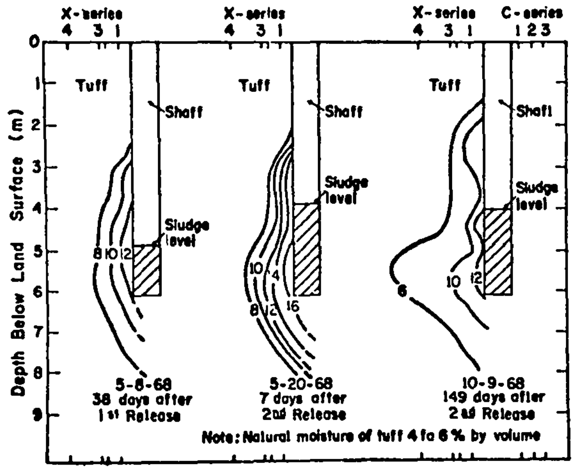

Fig. 4.
Cross section of shaft area showing isofluid contents (per cent by volume) in tuff.

About 150 days after the second release of sludge, the fluids had moved horizontally into the tuff more than 3 m. The vertical movement remained at about 2 m. The fluid content of the tuff about 30 cm from the shaft decreased from 16 to 12% by volume as the fluids were distributed to a greater distance from the shaft.

The porosity of the ashflow tuff ranges from 35 to 45% by volume, with fluid contents of less than 16%. Thus the tuff is not saturated and movement of fluid is under unsaturated conditions. Abrahams described the energy relationship of fluid to tuff with an average porosity of 40% by volume.[7] The movement of fluids in the capillary size pores can be explained by one or more of the processes noted in Table I. The fluid content of the tuff near the shaft was 12% by volume after the initial release and increased to about 16% after the second release. The main movement processes in this fluid content range are capillarity and gravity. When the fluid content decreased below 10%, the major movement processes were diffusion and capillarity. The lack of vertical movement of fluids and the fluid content of the tuff during the initial period of the test indicated the major process of movement was by capillarity aided somewhat by gravity. During the later periods, the moisture content adjacent to the shaft decreased and the movement was mainly by diffusion and capillarity (Fig. 4).

The total volume of fluids in the tuff within the 8% contour on October 9, 1968, was estimated at 3.1×10^{3} ℓ, of which 1.6×10^{3} ℓ were fluids in the tuff prior to the study and 1.5×10^{3} ℓ were fluids that had moved into the tuff from the sludge (Fig. 3). The two releases of sludge contained about 2.9×10^{3} ℓ of fluids; thus the inventory accounts for about 60% of the fluids released. The remaining 40% of the fluids are retained in the shaft with the sludge or may have moved in the lower moisture range beyond the 8% contour. The major mechanism for transporting plutonium from the sludge was the fluid movement into the tuff.

B. Movement of Plutonium

Predisposal concentrations of ^{238}Pu and ^{239}Pu in the tuff were <0.02 pCi/g (analytical limits of detection) as determined from cuttings from the four X-series test holes (Fig. 2). The plutonium concentration in the 2.9×10^{3} ℓ of sludge pumped into the shaft was about 45×10^{3} pCi/ℓ, or about 1.3×10^{-4} Ci. About 90% of the plutonium was ^{239}Pu.

Cores analyzed from the C-series test holes, taken about seven months after the second release of sludge into the shaft, showed ^{238}Pu was detectable only in hole C-1, 30 cm from the shaft, at a depth of 4.6 to 5.5 m. The ^{239}Pu was detectable out to about 60 cm from the shaft at a depth of 5 to 5.5 m (Table II). The core from 1.8 to 2.1 m of hole C-1 contained 0.17 pCi/g of ^{239}Pu. However, this does not indicate differential movement between the ^{238}Pu and ^{239}Pu. The ^{239}Pu was detectable at greater distances from the shaft because of the larger amount of that isotope in the sludge.

The plutonium concentrations decreased outward from the shaft, following the same general pattern as the movement of fluids. The total plutonium (^{238}Pu and ^{239}Pu) in hole C-1 ranged from 0.33 to 1.52 pCi/g in the interval 4 to 5.5 m, where the fluid content of the tuff reached a maximum of 16% by volume on May 20, seven days after the second release (Fig. 5). The interval 4 to 5.5 m is opposite the sludge remaining in the shaft, which also may have contributed fine particles with attached plutonium that moved out with the fluids.

Measurable amounts of plutonium were found in about 1.3 m^3 of tuff adjacent to the shaft. The inventory in the tuff was estimated using two volumes (density of the tuff 1.47 g/cm^3) and two concentrations. The inner volume had an average concentration of 1.1 pCi/g and a mass of 9.8×10^{5} g (0.67 m^3), and the outer volume a concentration of 0.26 pCi/g and a mass of 9.3×10^{5} g (0.63 m^3). The amount of plutonium in the tuff was estimated at 1.1×10^{-6} Ci. Compared to the 1.3×10^{-4} Ci of plutonium contained in the $Fe(OH)_3$ sludge disposed in the shaft, the inventory indicates less than 1% of the plutonium moved from the shaft into the tuff.

The plutonium and sludge were probably retained by the tuff in part by the filtering of fine particles in the fluids by capillary size pores and in part by adsorption (ion exchange) in the tuff.

TABLE I

MOVEMENT PROCESSES AT VARIOUS FLUID CONTENTS IN TUFF

Movement Process	Fluid Content (% volume)	Tension (bars)
No apparent movements	<5	---
Diffusion & capillarity	5 ≃ 10	15.2
Capillarity & gravity	10 ≃ 22	0.34
Drainage by gravity	22 ≃ 39	---

IV. SUMMARY

The major transport mechanism for radionuclide migration from a sludge released into shafts in the tuff is by movement with fluids associated with the

TABLE II

PLUTONIUM ANALYSES FROM C-SERIES TEST HOLE
NOVEMBER 1968

		Picocuries per Gram					
		^{238}Pu			^{239}Pu		
From	To	C-1	C-2	C-3	C-1	C-2	C-3
1.8	2.1	<0.02	<0.02	---	0.17	<0.02	---
2.7	3.0	0.04	0.02	---	<0.02	<0.02	---
3.4	3.7	0.06	---	<0.02	0.02	---	<0.02
4.0	4.3	0.05	<0.02	---	0.28	<0.02	---
4.6	4.9	0.15	<0.02	<0.02	0.86	<0.02	<0.02
5.2	5.5	0.12	<0.02	<0.02	1.4	0.06	<0.02
5.8	6.1	<0.02	<0.02	<0.02	0.06	<0.02	<0.02
6.4	6.7	<0.02	<0.02	<0.02	0.03	<0.02	<0.02
7.3	7.6	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02
8.8	9.1	<0.02	<0.02	---	<0.02	<0.02	---
10.7	11.0	<0.02	---	---	<0.02	---	---

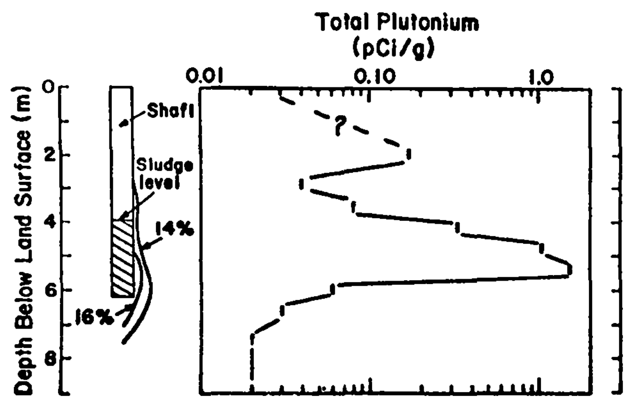

Fig. 5.
Cross section of shaft area showing maximum fluid content of tuff and sludge level in shaft in relation to total plutonium in cores from hole C-1.

wastes. Test holes drilled adjacent to a cement-paste filled shaft indicate no loss of fluids into the tuff from the hydration of the cement-paste mixture. Thus, movement or transport of contaminants is very unlikely because of the lack of fluids. Open joints in the tuff intersecting the shaft will allow early movement of the contaminated cement paste; however, upon hydration of the cement in the joint, this movement ceases.

The $Fe(OH)_3$ sludge released into a shaft during the study contained fluids that moved into the adjacent tuff, carrying some plutonium into the tuff with it. Trace amounts of ^{239}Pu were found about 60 cm from the shaft; however, the inventory indicates that 99% of the plutonium was retained in the sludge remaining in the shaft. Plutonium that moved with fluids in the tuff was probably retained by the filtering properties and adsorption (ion exchange) in the tuff.

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