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LA-UR -89-3049

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DE90 000601

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SUBMITTED TO: present to Migration 89, Monterey, CA, November 6-10, 1989, proceedings to be published in Radiochemica Acta.

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MIGRATION OF FISSION PRODUCTS AT THE NEVADA TEST SITE: DETECTION WITH AN ISOTOPIC TRACER

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Radionuclide migration/^{101}Rh/^{102}Rh

Abstract

Researchers at Los Alamos National Laboratory are studying the migration of fission products away from explosion cavities formed by underground nuclear tests at the Nevada Test Site. In some cases, the isotopic composition of the the fission products or activation products associated with a particular test are distinctive and we may identify them many years after the event. In this paper we describe a case in which we used rhodium isotopes to identify the source of radioactive material that had moved some 350 m from the explosion site.

1. Introduction

1.1. The Nevada Test Site (NTS)

The NTS encompasses an area of about 3600 km^2 located in southern Nevada. This region is semi-arid, with the mountains receiving less than 30 cm of moisture per year and the valleys even smaller amounts. The elevation of the NTS varies from about 1200 m to about 2100 m. Rock types in this region include Precambrian and Paleozoic carbonates, Tertiary ash flow and ash-fall tuffs and rhyolites, and alluvial fill in the valleys. Hydraulic conductivity in the carbonate aquifer below Yucca Flat is estimated to be 1- to 100-m/yr; similar values are estimated for the conductivity in the brecciated rhyolites of Pahute Mesa. A more complete description of the geology and hydrology of the NTS is contained in Ref. [1].

Testing of nuclear devices began at the NTS in 1951. Since 1962 all United States nuclear tests have been underground. Over 500 such tests have been conducted at the NTS [2]. Approximately 30% of these were below the water table which lies 100- to 650-m below the land surface. Collapse of material into the explosion cavity often results in a crater at the surface.

1.2. The Hydrology/Radionuclide Migration Program (HRMP)

In 1973 a program was started at the NTS to investigate the fate of radionuclides deposited underground by nuclear testing. Of particular interest were questions concerning the possible movement of these radionuclides within or outside the boundaries of the NTS. The HRMP research project involves personnel from Los Alamos National Laboratory, Lawrence Livermore National Laboratory, the US Geological Survey, the Desert Research Institute, and the US Department of Energy. Our research includes both laboratory and field studies. We attempt to take advantage of the unique opportunities at the NTS provided by the many underground emplacements of a variety of radioactive materials.

1.3. A Case Study of an Unusual Situation

In 1984 when an emplacement hole for a future nuclear test was being drilled, tritium was detected in the formation water contained in the layered tuffs below the water table. Subsequent analyses of the water and of filtered material indicated the presence of fission products and a trace of plutonium. Since it is very unusual to encounter radioactive materials during the drilling of emplacement holes at the NTS, we wished to identify the source of these materials and to determine their mode of transport to that location. Four previous tests in the area were identified as possible sources of the radioactive materials, though all were hundreds of meters distant. The presence of rhodium isotopes in the transported material made it possible to identify the test which produced the source material. The circumstances which gave rise to this study and other aspects of our work at the NTS are described in annual reports (for example, see Ref. [3]).

2. Sampling and Data Acquisition

The initial samples from the contaminated site were small-volume grab samples of water taken during the drilling operation. Tritium was detected by liquid scintillation counting. Later we collected 2-liter water samples in evacuated tubes; in them we confirmed the presence of tritium and also measured ^{85}Kr, a fission product. We collected a number of barrels (208 liters) of muddy water from the emplacement hole, filtered the water through 50 nm pore size Nuclepore filters, and evaporated the filtrate to dryness. Both the filtrate residue and the filters were counted on Ge(Li) gamma ray analyzers. Some portions of the filtrate were analyzed for ^{90}Sr by beta counting and for plutonium by mass spectrometric techniques.

Our analyses showed that the water contained tritium, ^{85}Kr, ^{90}Sr, ^{106}Ru, ^{125}Sb, and ^{137}Cs. Particulates collected on the filters contained ^{106}Ru, ^{125}Sb, ^{137}Cs, and a trace of ^{102}Rh. The trace of plutonium detected through mass spectrometry had an isotopic composition indicative of early, above-ground testing. We believe this indicated a small amount of airborne contaminate had gotten into the water sample. Bore hole gamma logs showed that the radioactivity was confined to a relatively narrow zone at a depth of almost 650 m. After detecting rhodium in the residue on the filters, we separated rhodium from the other components and analyzed it by Ge(Li) spectroscopy. Our separation scheme generally followed that outlined in Ref. [4]. The separation required dissolution of the filters and residue, precipitation of RhI_3, formation of $[Rh(CN)_6]^{3-}$ complex, scavenging with hydroxide and sulfide, and precipitation of $Cu_3[Rh(CN)_6]_2$. Finally, the purified rhodium chloride solution was counted in a Ge(Li) well detector which was surrounded by a large NaI(Tl) detector operated in an anticoincidence mode to suppress Compton radiation.

3. Source Identification

3.1. Timing with Rhodium Clocks

When rhodium is added to a nuclear test device, rhodium isotopes of mass 102 and 101 are produced by neutron reactions. The decay properties of these isotopes are shown in Table 1.

Since the two isomers of ^{101}Rh are formed by similar reactions, the metastable to ground state ratio at the time of production is nearly a constant for all nuclear devices. This ratio, measured at a later time, can be used to calculate the age of the sample for as long as the shorter lived metastable state can be detected (approximately 60 days). Similarly, the two isomers of ^{102}Rh can be used to date samples up to about seven years old. The ratio of ^{101}Rh to ^{102}Rh (particularly the two longer lived isomers) can vary by at least a factor of 10 and is device specific. This ratio changes only slowly with decay and can be measured for 20 years or longer. It also can be used to identify debris depending on the uniqueness of the particular device and its rhodium loading.

There is some uncertainty about the values of the half-lives of ^{101g}Rh and the longer lived of the ^{102}Rh isomers. The values shown in Table I are those we used in our analyses; they were determined by one of us (JSG) by following the decay over a period of 5 years. There is also some uncertainty as to which of the ^{102}Rh isomers is the ground state. For the purpose of this paper it is adequate to identify them as ^{102}Rh(ℓ) and ^{102}Rh(s) for the longer- and shorter-lived species, respectively. Futhermore, since we are able to compare samples from the contaminated site with recently recounted samples from each of the suspected sources, the values of the long half-lives are not essential to our analysis.

3.2. Review of Possible Sources

There had been four tests conducted in the vicinity of our study area prior to the drilling of the emplacement hole. We were fortunate to have on hand rhodium samples from debris recovered from each of these tests. Fig. 1 contains portions of the gamma spectra from each of these sets of test debris; these spectra were taken at approximately the same time as the spectrum from the sample being analyzed, which is shown in Fig. 2. The spectra in Fig. 1 illustrate both the disappearance with time of ^{102}Rh(s) and the different ratios of ^{101}Rh/^{102}Rh(ℓ).

3.3. Determination of the Origin of the Sample Material

A comparison of the spectra in Figs. 1 and 2 shows very clearly that the sample spectrum most closely resembles the spectrum of test site D. A more quantitative evaluation of the data leads to the same conclusion, as illustrated in Table II. We concluded that the sample material was very similar to the debris from test site D. This conclusion was based on the absence of ^{102}Rh(s) in the sample spectrum and the similarity of the ratio ^{101}Rh/^{102}Rh(ℓ) in the sample spectrum and in the spectrum of debris from test site D. Since rhodium contributed only one minor peak to the gamma spectrum of the sample debris, chemical separation of rhodium from the other material was essential. The separated sample required counting times as long as 4000 minutes to obtain statistically valid data.

Of particular interest to us is the indication that rhodium moved some 350 m away from its initial source. We do not know whether this movement occurred slowly over the 9 years from the time of the test or occurred rapidly (perhaps by a pressure pulse injection in a fracture) at the time of the nuclear detonation. We are continuing to seek evidence which will allow us to determine the means by which the rhodium and other fission products moved. It should be noted that the data do not prove that the tritium and fission products found at the contaminated site all arrived with the rhodium. However, the fact that the radioactive material seems to be confined within a small zone does argue against general contamination from multiple sources.

4. Summary

Fission products and tritium were found at the site of an emplacement hole being drilled at the NTS. This unusual discovery stimulated a study of the area and resulted in the identification of the source of the contaminating material. The presence of ^{101}Rh and ^{102}Rh isotopes in the sample and at the suspected sources allowed us to determine the actual source from among several possibilities. Chemical isolation of rhodium and Ge(Li) gamma spectroscopy were essential for the success of our analyses. The mechanism by which the rhodium migrated 350 m is still under investigation. The numerous underground nuclear tests at the NTS provide a unique opportunity to study migration of fission products in igneous rock.

References

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Table 1. *Decay data for ^{101}Rh and ^{102}Rh isotopes*

Isotope	Half-Life	Principle γ-ray energies (kev)
^{101m}Rh	4.34 d	306.77, 544.85
^{101g}Rh	4.04 ± 0.04 yr	127.21, 198.0, 325.2
^{102}Rh(ℓ)	3.82 ± 0.04 yr	418.3, 475.0, 631.1, 697.1, 766.8, 1046.6, 1112.7
^{102}Rh(s)	207.9 d	468.6, 475.0, 556.5, 628.0, 1103.4, 511 ($_+\beta$)

Table 2. *Rhodium isotopic composition of samples*

Test	^{102}Rh(s) in 1986	$\frac{^{101g}\text{Rh}/^{102}\text{Rh}(\ell)\text{ at shot time in test debris}}{^{101g}\text{Rh}/^{102}\text{Rh}(\ell)\text{ at shot time in emplacement hole debris}}$
A	measurable	1.96
B	measurable	1.03
C	measurable	1.13
D	not measurable	1.00
emplacement hole	not measurable	

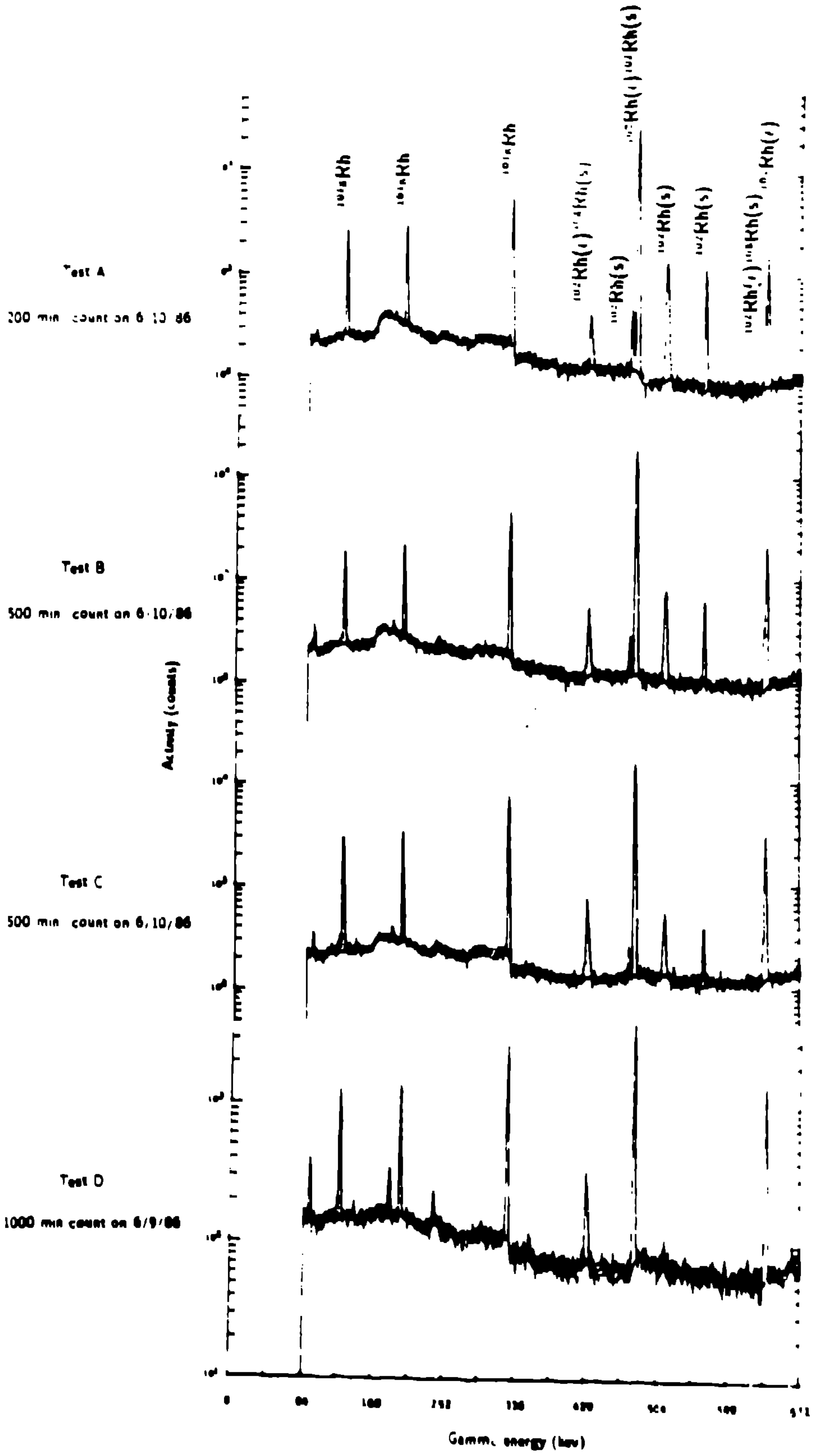

Fig. 1. Gamma spectra of test debris.

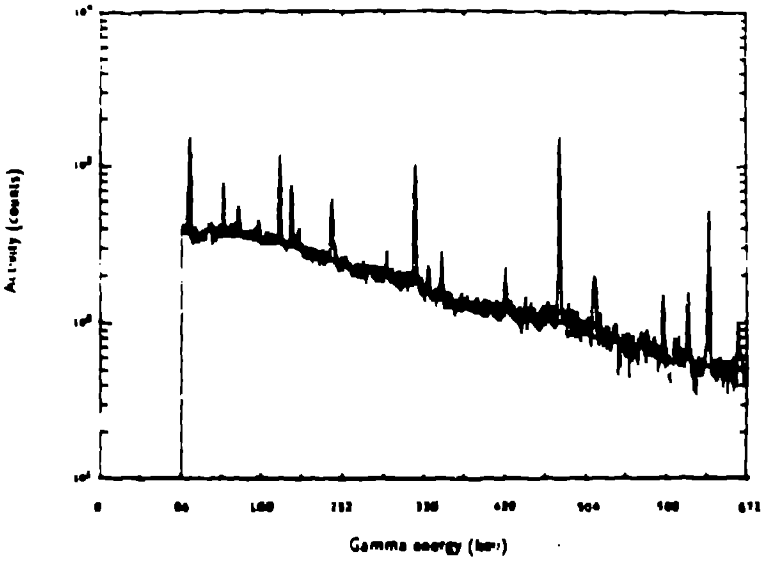

Fig. 2. Gamma spectrum of sample.
4043 min. count on 6/6/86.