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SEABED DISPOSAL PROGRAM: GEOCHEMICAL AND SEDIMENTOLOGICAL
STUDIES OF NORTH PACIFIC SEDIMENTS

Progress Report
for Period January 1, 1976 - March 31, 1976

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April 1976

Prepared For

THE U.S. ENERGY RESEARCH AND DEVELOPMENT ADMINISTRATION
UNDER CONTRACT NO. E(11-1)-2689

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Abstract

Studies of sorption equilibria between end-member (illite-rich and smectite-rich) clays from the MPG-2 area (33°20'N, 151°00'W) and ThCl_4 solutions in 0.7M NaCl have shown that partitioning is both concentration and temperature dependent. Preliminary data indicate that the partition coefficient is inversely proportional to the solution concentration, and increases by about a factor of 3 from 15 to 85°C.

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Introduction

As part of a multidisciplinary, multi-institutional study to evaluate the bed of the deep-sea as a repository for the permanent disposal of high-level radioactive waste, we are determining the barrier properties of the sediments and the long-term physical stability of two areas in the central North Pacific. Factors guiding the selection of test areas and the overall plan of attack for the Seabed Disposal Program are outlined in reports by Bishop and Hollister (1974) and Anderson et al (1975).

Progress for the first three quarters of this fiscal year has been summarized in an earlier report (C00-2689-3).

Progress January - March 1976

Our efforts this quarter have been concentrated on the first studies of the sorption properties of MPG clays. From our earlier work, we know that the MPG sediments range from quartz - illite-rich clays of predominantly eolian origin near the sea floor, to smectite-rich clays containing variable phillipsite and ferromanganese hydroxyoxides at depths of 5 - 12 meters. For the sorption experiments, we are using bulk samples of typical end-member sediments. The illitic-rich sample, designated Q1, is from 20 - 25 cm in core Y74-3-66LG (33°12.8'N, 150°58.8'W). The smectite-rich sample, designated T2, is from 867 - 1083 cm in core Y74-3-58P (33°10.1'N, 150°54.1'W). Spot checks of additional samples will be made during the study to ensure that the experimental results are applicable to the full range of sediments in the MPG areas.

The first experiments utilize ThCl_4 solutions in 0.68 ionic strength NaCl (to simulate the ionic strength of seawater but temporarily to avoid its complexity). These experiments are designed to assess the effects of temperature and ion concentration on partition coefficients. To date, temperatures in the range 15-85°C and concentrations in the range 2×10^{-2} to $4 \times 10^{-6}\text{M}$ have been tested.

Experimentally, 0.1g of sediment which has been split quantitatively from a bulk sample is equilibrated with 0.68M NaCl solution, centrifuged, then resuspended in 25ml of ThCl_4 in NaCl solution. After agitation for 24 hours at the specified temperature, the sample is centrifuged and re-equilibrated at the specified temperature. A pipette equipped with a $0.45\mu\text{m}$ filter is used to sample the supernatant liquid for Th analysis by colorimetry (concentrations above 10^{-4}M) or γ -spectrometry (using a Th^{228} spike in the test solution).

Preliminary data indicate that the partition coefficient is strongly concentration dependent ($K \approx k.C_s$ for Q1 at 15°C, where K is in ml/gm, C_s is in moles, and k, the proportionality constant, is about 0.1 for the units cited). This inverse relationship leads to extreme sorption at the diffusional front from a limited source (such as a canister), resulting in very slow migration of Th ions through the sediment (rates of about 1 meter per 100,000 years for a $1\mu\text{M}$ solution, for example).

The effect of a temperature increase is to increase the partition coefficient (about a factor of 3 from 15 to 85°C). Experiments in progress are designed to determine the functionality of the temperature effect, and its dependence on the solution concentration.

Mineralogy seems to be the least important variable examined thus far. The

smectite-rich clay has slightly higher partition coefficients than the illite-rich one, but the differences are small in comparison to the temperature and concentration effects.

Work underway substitutes seawater for the NaCl solution and extends the present series of experiments to additional temperatures and lower Th concentrations. Experiments just beginning will assess the sorption behaviour of U under oxidizing conditions.