

23. URANIUM RESULTS FROM HOLE 504B WATER SAMPLES¹

Michael P. Bacon, Woods Hole Oceanographic Institution, Woods Hole, Massachusetts

ABSTRACT

Measurements of U concentration and $^{234}\text{U}/^{238}\text{U}$ ratio were made in five water samples from Deep Sea Drilling Project Hole 504B. The results indicate that a loss of U had occurred either during sampling or during the storage of the samples, probably as a result of adsorption by rust particles or by the walls of the sampling system. Within analytical uncertainty, the $^{234}\text{U}/^{238}\text{U}$ ratios in the samples were identical to those in unreacted seawater. Thus, it is not possible to detect any U exchange that may have occurred during the reaction of the solutions with the basement formation. Improvement in sampling technique is a necessity for future studies of uranium and probably other trace elements in drill hole water samples.

INTRODUCTION

It is known that significant amounts of seawater uranium are added to oceanic basalts during low temperature alteration (Aumento, 1971; Thompson, 1973), and MacDougall (1977) has suggested that this process may represent the principal oceanic sink for uranium. Measurements of radioactive disequilibrium in altered basalts (Bacon, 1978; MacDougall et al., 1979) have shown, in some samples, excess ^{234}U ($^{234}\text{U}/^{238}\text{U}$ activity ratio > 1.00), reflecting the 14% ^{234}U enrichment of seawater. However, in other samples, especially the more intensely altered ones, ^{234}U deficits have been found, indicating a loss of ^{234}U formed *in situ* by radioactive decay. Bacon (1978) has suggested that the net effect of the low temperature weathering of basalt is the removal of uranium from seawater but that the removal is accompanied by a virtual isotope fractionation such that ^{234}U tends to be left in solution. He also suggests that, in contrast to these effects, which take place at low temperatures, some uranium may be leached from basalts during hydrothermal alteration, possibly with the preferential loss of ^{234}U . The purpose of the analyses reported here was to determine directly the net effect of the rock/water interactions on the composition of seawater by measuring the uranium concentration and isotopic composition of reacted seawater samples.

The samples analyzed for this investigation were collected by the packer method. As shown by Mottl, Anderson, et al. (this volume), the water's chemical composition and tritium content indicated that the samples from Hole 504B had varying proportions of two components: unreacted surface seawater used to flush the hole at the time of sampling and reacted surface seawater drawn from pores in the rocks surrounding the hole. The reacted surface seawater had apparently been pumped into the basement formation during earlier operations and had reacted with the basalt over a period

of several days at 80°C. During this reaction the solutions gained Ca and Si and lost Mg and K.

One-liter aliquots of the samples were received in polyethylene bottles. They had never been filtered or acidified. Small amounts of powdered basalt and pipe rust were present, but most had settled out before the aliquots were taken. There was no attempt to remove the small amounts of solid that remained. At the start of the analysis, each sample was acidified to a pH of 2 with HCl, spiked with a known amount of calibrated ^{236}U tracer and a solution containing 50 mg FeCl_3 , and bubbled for several hours with filtered air. Uranium was concentrated by co-precipitation with $\text{Fe}(\text{OH})_3$ after the sample solution was neutralized with NH_4OH . Uranium was then isolated and purified by anion exchange and solvent extraction. Thin sources for alpha spectrometry were prepared by electroplating the uranium on silver disks.

RESULTS AND DISCUSSION

The results are given in Table 1, where the packer samples (PS) are numbered in the order of decreasing proportion of reacted seawater. Thus, Sample PS-1 was expected to have the most altered composition. How-

Table 1. Uranium results.

Sample	U ($\mu\text{g}/\text{kg}$)	Activity Ratio ($^{234}\text{U}/^{238}\text{U}$)
SW-1 ^a	3.27 \pm 0.06	1.16 \pm 0.02
SW-2 ^a	3.19 \pm 0.06	1.19 \pm 0.02
PS-1	1.88 \pm 0.03	1.14 \pm 0.02
PS-2	2.04 \pm 0.04	1.15 \pm 0.02
PS-3	1.65 \pm 0.03	1.17 \pm 0.02
PS-4	1.64 \pm 0.04	1.15 \pm 0.03
PS-5	1.73 \pm 0.03	1.17 \pm 0.02

^a Aliquot of normal seawater (S = 34.673‰) collected in the eastern equatorial Pacific (9°36'N, 89°9'W) at a depth of 125 m. From the results of Ku et al. (1977), the expected values are 3.24 \pm 0.19 $\mu\text{g}/\text{kg}$ for U and 1.14 \pm 0.03 for $^{234}\text{U}/^{238}\text{U}$.

¹ Cann, J. R., Langseth, M. G., Honnorez, J., Von Herzen, R. P., White, S. M., et al., *Init. Repts. DSDP*, 69: Washington (U.S. Govt. Printing Office).

ever, it was not possible to determine the actual percentages of the two components present in the samples, because the composition of the reacted component was not known. I also analyzed two normal seawater samples that had been collected in the eastern equatorial Pacific and found good agreement with accepted values for uranium concentration and isotopic composition (Ku et al., 1977). On that basis the uranium concentration in the unreacted component (surface seawater, for which $S = 33.8\%$) would have been $3.1 \pm 0.2 \mu\text{g}/\text{kg}$.

The uranium concentrations found in all of the drill hole samples are lower than they are in unreacted seawater, indicating that a loss of uranium had occurred. However, there is no correlation between the concentration of uranium and the proportion of the reacted seawater component. Therefore, the loss of uranium seems to have occurred either during the sampling operation or later, during storage, and it is not possible to determine the effect of reaction in the basement formation. The loss of uranium was probably due to adsorption by the rust particles that were unavoidably present or by the walls of the sampling system. Since most of the particles initially present in the samples had settled out before the aliquots were taken, the uranium lost by adsorption to them would not have been recovered on acidification. Other trace elements are likely to be similarly affected by this sampling problem, indicating the need for improvement in sampling technique. Possible improvements would consist of lining the sample cham-

bers with an inert material such as Teflon and installing prefilters in the sampling ports.

None of the reacted seawater samples show $^{234}\text{U}/^{238}\text{U}$ ratios that differ significantly from the normal seawater value. Thus, I do not find evidence for the preferential leaching of ^{234}U previously suggested (Bacon, 1978), although the length of time for reaction was obviously very short.

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