

## MOBILITY AND RETENTION OF $^{60}\text{Co}$ IN SOILS IN COASTAL AREAS

Yasunori Mahara<sup>1)</sup> and Akira Kudo<sup>2)</sup>

- 1) Visiting scientist, Division of Biological Sciences, National Research Council of Canada, from Central Research Institute of Electric Power Industry, Abiko, Japan.
- 2) Research Officer, Division of Biological Sciences, National Research Council of Canada, Ottawa, Canada, K1A 0R6, and visiting professor, University of Ottawa.

The operation of more than 20 commercial nuclear reactors, at coastal sites in Japan during the past several years, has produced a tremendous amount of radioactive wastes which have saturated the waste storage facilities at the reactor sites. Because of present Japanese safety regulations, the wastes cannot be disposed of permanently underground nor on the ocean floor. Two methods of disposal (either into the ocean floor or underground) have been much discussed, but still more information is needed on the movements of radionuclides in the environments. This is definitely so for the movements of radionuclides underground, especially in underground water-flow systems. The lack of information includes data on and the mechanisms of the chemical and/or biological transformation of radioactive materials during the movements under various geochemical conditions.

Among the operating wastes from electricity generating nuclear stations, radioactive cobalt ( $^{60}\text{Co}$ ) is a major radionuclide in quantity and as a radiological hazard. It has a relatively long half life (5.3 years) and emits two gamma rays with high energy (1.17 Mev and 1.33 Mev). Furthermore, cobalt is one of the essential elements to sustain life in animals and plants (for example, as Vitamin B<sub>12</sub>) and hence is easily accumulated by various organisms, if available to them (1,2).

This paper summarizes the results of our previous investigation and reports some recent findings of  $^{60}\text{Co}$  interactions in seawater-sediment systems, especially pertaining to the difference in the mobility of  $^{60}\text{Co}$  under aerobic and anaerobic conditions. Furthermore, the difference in the mobility of  $^{60}\text{Co}$  in seawater and in freshwater was investigated under the influence of various environmental factors. Details of the investigation will be reported elsewhere (3).

### MATERIALS AND METHODS

A series of laboratory experiments was conducted simulating some of the underground environments at coastal areas. The interaction of  $^{60}\text{Co}$  with various sediments in both freshwater and seawater systems was observed for a considerable period under either aerobic or anaerobic conditions. The degree of  $^{60}\text{Co}$  mobility, after it had interacted with water and sediments, was determined by measuring radioactivity in the water phase and in the sediment phase.

The reversible (and irreversible) nature of  $^{60}\text{Co}$  interaction between water and sediments was evaluated by a quick change of the surrounding environments. The  $^{60}\text{Co}$ , for example, after having

interacted under anaerobic conditions for the first 30 days, was suddenly introduced into aerobic conditions for the next 30 days. On the other hand, the  $^{60}\text{Co}$ , kept under an aerobic condition for the first 30 days, was moved into a condition of anaerobic for a further 30 days. To achieve the new anaerobic and aerobic conditions, nitrogen and oxygen gases were bubbled respectively into the systems.

To evaluate some of the physical and chemical characteristics of the highly mobile  $^{60}\text{Co}$  produced in a seawater-sediment system, the  $^{60}\text{Co}$  was separated from the rest of the system by means of a dialysis membrane (24 Å pore size), a chemical extraction (dithizone-benzene), and a sorption (various sediments). Details of the physical and chemical characteristic of the sediments used were reported elsewhere (4-6).

## RESULTS AND DISCUSSION

The magnitude of  $^{60}\text{Co}$  mobility under four different environmental conditions is illustrated in Fig. 1. The mobility in the system of the sand sediments and freshwater under aerobic condition was used as a base line for a comparison. The mobility of  $^{60}\text{Co}$  increased at pH values between 6 and 8 in the following order: (1) in freshwater under aerobic conditions, (2) in seawater under aerobic conditions, (3) in freshwater under anaerobic conditions, and (4) in seawater under anaerobic conditions. It is seen that the  $^{60}\text{Co}$  in the system of seawater under anaerobic conditions is by far the most mobile.

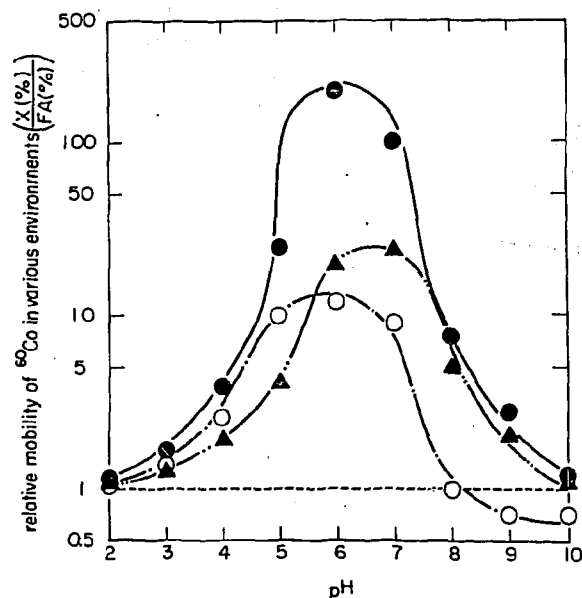


Fig. 1. Magnitude of  $^{60}\text{Co}$  mobility in various environments:  $\blacktriangle$ -...- $\blacktriangle$  X = freshwater under anaerobic,  $\bullet$ -...- $\bullet$  X = seawater under anaerobic,  $\circ$ -...- $\circ$  X = seawater under aerobic, - - - -FA = freshwater under aerobic.)

The irreversible nature of  $^{60}\text{Co}$  derived from the changes in the redox environments was observed. In other words, the initial conditions such as aerobic or anaerobic conditions in the sediment and water systems where  $^{60}\text{Co}$  was initially discharged seemed to determine the magnitude of  $^{60}\text{Co}$  mobility. In Fig. 2, characteristic variation of  $^{60}\text{Co}$  mobility was shown when incubating anaerobic conditions were suddenly converted into aerobic, and vice versa. The following results were observed in a series of these experiments; (i) if  $^{60}\text{Co}$  initially had a high mobility under anaerobic conditions, it could have the high mobility even if surrounding environments were changed drastically, and (ii) if  $^{60}\text{Co}$  initially had a low mobility under aerobic conditions, it would keep the low original mobility even if surrounding conditions were changed.

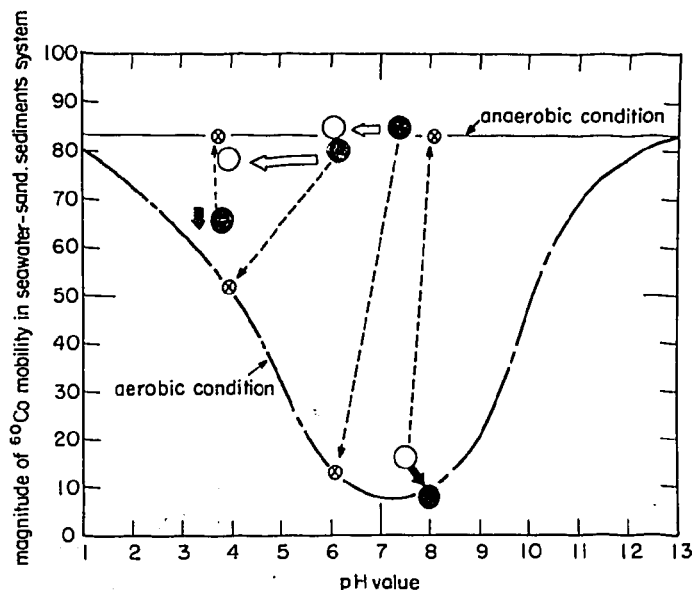


Fig. 2. Mobility of  $^{60}\text{Co}$ , in the system of seawater and the sand sediments, with respect to changes in pH. (Incubating condition was changed suddenly from aerobic to anaerobic, or vice versa.) The following five symbols (○, ●, ⊗, ⇌, →) indicate respectively: the magnitude of  $^{60}\text{Co}$  mobility under aerobic conditions; mobility under an anaerobic condition; mobility of reversible  $^{60}\text{Co}$  expected by changes in the incubating condition; direction of change from aerobic to anaerobic conditions; and direction of change from aerobic to anaerobic conditions.

A highly mobile form of  $^{60}\text{Co}$  produced in the seawater system under anaerobic conditions could pass through a dialysis membrane freely (more than 99%). This result indicated that the mobile form of  $^{60}\text{Co}$  consisted of a mixture of ionic cobalt, cobalt compounds with low molecular weight, and cobalt adsorbed (or absorbed) on the surface of very fine particulates whose diameter is less than  $24 \text{ \AA}$ .

A comparison was made between ionic form of cobalt ( $\text{CoCl}_2$ ) and the cobalt which was highly mobile and could pass through a dialysis membrane freely. The purpose of this comparison was to define some of

the characteristics of the highly mobile cobalt produced in the system of seawater under anaerobic conditions. Figure 3A shows a result of dithizone-benzene extraction for two forms of the cobalt at various pH values. The highly mobile cobalt was not extracted at all by the chelating agent with an organic solvent, though almost all of the ionic form was extracted at pH values between 4 and 8.

There is a clear difference between two forms of the cobalt in sorption to a fresh sand sediment. Again, the highly mobile cobalt was not reactive to the sediments, though the ionic form was sorbed nearly 100% by the sediments at pH values between 5.5 and 7, Fig. 3B. Unfortunately, the precise nature of the highly mobile cobalt has not yet been determined, but the result of these experiments suggest that it is stable and moves freely with the water in the flow systems of underground environments at the coastal areas.

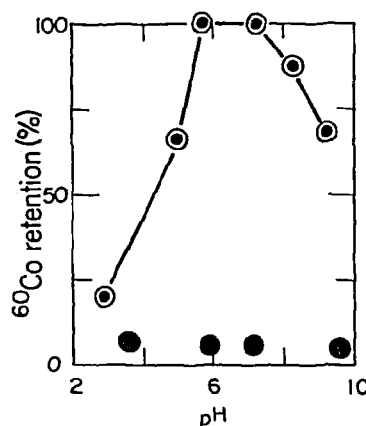
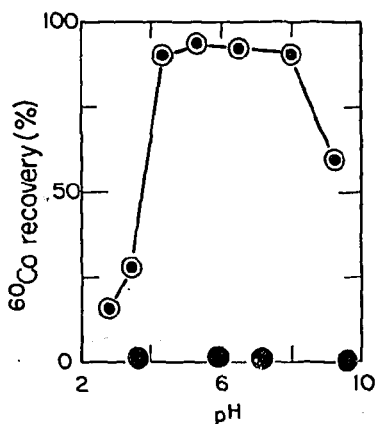


Fig. 3-A. Comparison between recovery of mobile  $^{60}\text{Co}$  in seawater under anaerobic conditions by dithizone-benzene and recovery of ionic  $^{60}\text{Co}$ .

Fig. 3-B. Comparison between sorption of mobile  $^{60}\text{Co}$  in seawater under anaerobic conditions on the sand sediments and sorption of ionic  $^{60}\text{Co}$ .

(●;  $^{60}\text{Co}$  in seawater under anaerobic, ○; ionic  $^{60}\text{Co}$ )

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