Chemical behavior of ^{7}Be in cooling-waters
~ Colloid formation in accelerator facilities and experimental radiation fields ~

Cooling water is an indispensable component of accelerator facilities as it enables the cooling of various instruments such as electromagnets, RF cavities, targets, and particle detectors. At high-energy and high-intensity accelerators such as J-PARC, radioactivity in the cooling water, produced by high-energy radiation poses serious problems for radiation management. Treatment of radioactivity in cooling water is more difficult than that of radioactivity in solid components such as targets and magnets; that is because behavior of radionuclides in water depends on their chemical states. In particular, the presence of colloidal species complicates the behavior of radionuclides in water cooling systems. Therefore, the characteristics and formation process of radioactive and non-radioactive colloidal species in cooling water were studied by a team of KEK Radiation Science Center, Kyoto University Research Reactor Institute, and Chiba University.

In water cooling systems at accelerator facilities, deionized water is used as the cooling medium; the ion-exchange resin units installed in the water-loop maintain water quality by removing soluble ionic species. The ion-exchange resins are expected to collect both non-radioactive ionic species and ionic radionuclide species. However, ^{7}Be (a major radionuclide produced by the spallation of oxygen in water molecules) in the cooling water for electromagnets at proton accelerator facilities was found to be partially passing through the ion-exchange resin units and circulating in the water cooling systems. This is in contrast with the well-known fact that Be species in water exist as Be(II) cations, Be^{2+} and $Be(OH)^{+}$, and can be easily removed by cation-exchange resins. This group considered that the incomplete adsorptivity of ^{7}Be on the ion-exchange resin was caused by formation of colloidal ^{7}Be species and discussed the validity of this consideration.

The size fraction of colloids associated with ^{7}Be in the cooling water for electromagnets at the 12 GeV proton accelerator facility, KEK, was measured by ultrafiltration technique in order to investigate the existence of radioactive colloids. Figure 1 shows these measured size fractions; this figure indicates that approximately 60% of ^{7}Be in the circulating water exists as fine colloids whose sizes range from 3 to 7 nm. Furthermore, electron microscopic analyses proved that the cooling water for electromagnets contains very fine particles (Fig. 2)

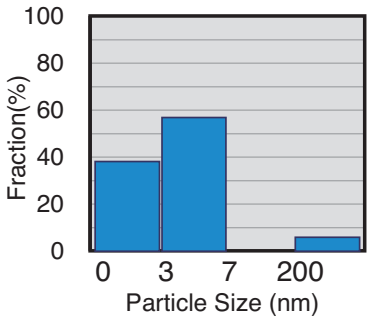

Fig. 1. Non-colloidal (0–3 nm) and colloidal fractions for different size ranges (3 – 7, 7 – 200, and >200 nm) of ^{7}Be in the cooling water for electromagnets at the 12 GeV proton accelerator facility, KEK.

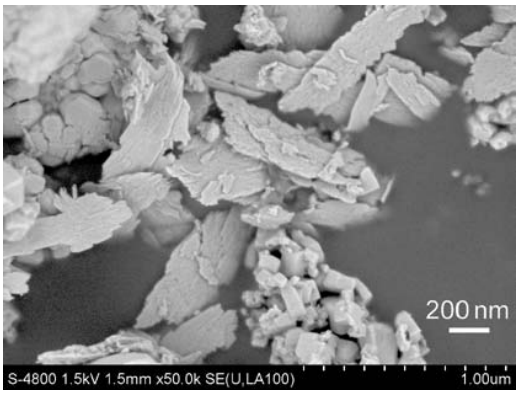

Fig. 2. Electron microscope image of particles formed in the cooling water for electromagnets and RF cavities at the J-PARC MR facility.

and that these particles are mainly composed of copper oxides (CuO or Cu_2O). These observations imply that ^{7}Be-related colloids are formed by two steps: (1) formation of copper-oxide colloids in water by corrosion of copper tubing material and (2) adsorption of ^{7}Be on the copper-oxide colloids. Various metal components come in direct contact with the cooling water in the accelerator facilities; hence, it is possible to create metal-oxide colloids for other materials such as stainless steels, iron, aluminum alloys, and various particular metals that are used for targets or accelerator components.

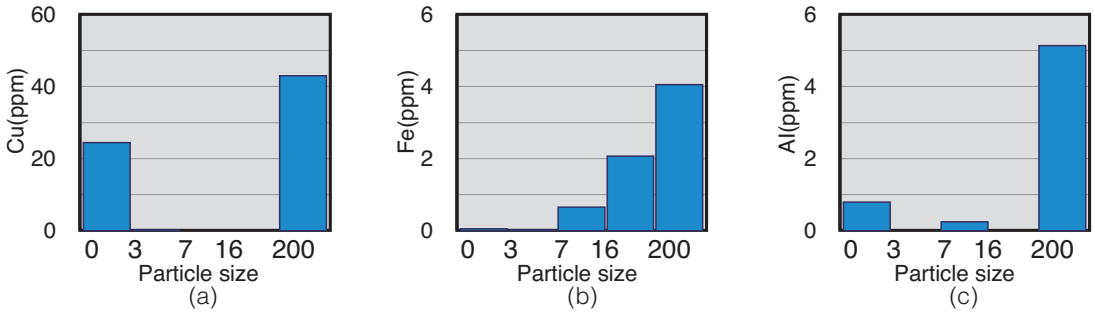

Fig. 3. Non-colloidal (0–3 nm) and colloidal concentrations for different size ranges (3–7, 7–16, 16–200, and >200 nm) of (a) Cu, (b) Fe, and (c) Al in water sealed into the respective pure-metal containers (Inside: Φ10.7 mm × 298 mm) after irradiation by high-energy photons* for 4h.
* Bremsstrahlung generated by 30 MeV electron beam striking a Ta target assembly.

The first process, formation of metal-oxide colloids, was investigated under two experimental conditions that simulate the environment in accelerator facilities. First, pure water samples sealed into the metal containers (Cu, Fe, and Al) were irradiated by bremsstrahlung generated by a 30 MeV electron beam striking a Ta target assembly. Figure 3(a)–(c) shows the non-colloidal and colloidal concentrations of Cu, Fe, and Al in water after the irradiation experiments. High concentrations of metal elements were transferred to the water phase, and creation of particles was clearly observed. The particle-size profiles are dependent on the metal materials. The results suggested that the irradiation of high-energy photons promotes the corrosion of metal surfaces and the generation and growth of colloidal species in water. Second, Cu metal particles dispersed in water were irradiated by several doses (0 – 3.3 kGy/h, 87 h) of γ-rays from a ^{60}Co radiation source. A similar tendency was confirmed in this condition. Creation of large colloids was facilitated by high doses of γ-ray irradiation. The chemical form of radiation-induced copper oxide (I) (Cu_2O) colloids was identified by X-ray diffraction analysis. These results indicate that Cu is oxidized in water under intense radiation and transferred to the water phase as Cu_2O colloids.

The second process, adsorption of radionuclides on metal oxide colloids, was investigated in the case of adsorption of Be(II) ions on CuO colloids, by placing the model of ^{7}Be in the cooling water for electromagnets. Aqueous Be(II) solution was mixed with CuO nanoparticles, and the adsorption behavior of Be(II) on CuO was

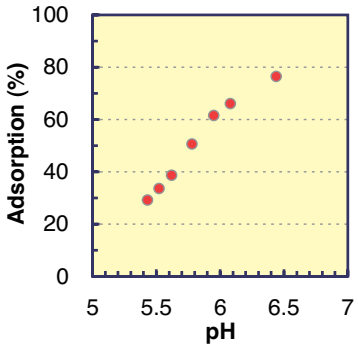

Fig. 4. Adsorption (%) of Be(II) on CuO nanoparticles in water as a function of pH. CuO nanoparticles (Average size: 48 nm) : 0.50 g; Water: 20 ml.

quantitatively examined. Figure 4 shows the dependence of adsorption (%) of Be(II) on CuO as a function of pH; this figure indicates that the adsorption of Be(II) on CuO monotonously increased with the pH of the solution. The detailed analysis of adsorption data measured under various conditions confirmed that Be(II) is adsorbed on CuO particles by a complex formation of –OH groups terminated at the CuO surface according to the following equation.

$$\text{CuO particle}\!-\!(OH)_n + Be^{2+} \rightleftharpoons \text{CuO particle}\!-\!(O)_n\!-\!Be^{(2-n)+} + n\,H^{+} \quad (n=1-3)$$

The information obtained from the present work will be valuable for establishing radiation safety at future high-energy/high-intensity accelerator facilities.