Adsorption Behavior of Beryllium(II) on Copper-oxide Nanoparticles Dispersed in Water: A Model for ^7Be Colloid Formation in the Cooling Water for Electromagnets at High-energy Accelerator Facilities

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The adsorption behavior of Be(II) on CuO nanoparticles dispersed in water was studied as a model for colloid formation of radioactive ^7Be nuclides in the cooling water used for electromagnets at high-energy proton accelerator facilities. An aqueous Be(II) solution and commercially available CuO nanoparticles were mixed, and the adsorption of Be(II) on CuO was quantitatively examined. From a detailed analysis of the adsorption data measured as a function of the pH, it was confirmed that Be(II) is adsorbed on the CuO nanoparticles by complex formation with the hydroxyl groups on the CuO surface ($>\text{S-OH}$) according to the following equation:

The surface-complexation constants corresponding to the above equilibrium, $\beta_{(s,2)}$ and $\beta_{(s,3)}$, were determined for four types of CuO nanoparticles. The $\beta_{(s,2)}$ value was almost independent of the type of nanoparticle, whereas the $\beta_{(s,3)}$ values varied with the particle size. These complexation constants successfully explain ^7Be colloid formation in the cooling water used for electromagnets at the 12-GeV proton accelerator facility.

Keywords Colloid, copper oxide, beryllium(II), complexation, accelerator, cooling water

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Introduction

Cooling water is an indispensable component at particle accelerator facilities that is required for the cooling of various components, such as electromagnets, targets, and particle detectors. Various radionuclides are produced in the cooling water due to nuclear reactions under an intense radiation field. Appropriate treatment of radionuclides in water is essential for the safe operation of extremely high-energy and/or high-intensity accelerator facilities; the operation of such facilities has been initiated in recent years,¹⁻³ while others are under construction or are in the design stage⁴ in various regions of the world.⁵⁻¹¹

Beryllium-7 (^7Be ; $T_{1/2} = 53.1 \text{ d}$)¹² is one of the most dominant gamma-ray emitting nuclides produced in the cooling water at high-energy accelerator facilities. Our previous studies focused on analysis of the existing forms and behavior of ^7Be in various cooling water systems at high-energy proton accelerator facilities.⁸⁻¹¹ These investigations demonstrated that a part of ^7Be in the circulating water was present as fine colloids with

diameters of several nanometers. The presence of colloidal species complicates the behavior of ^7Be in the water circulation system.^{7,10,11} In order to establish an appropriate treatment for ^7Be in cooling water, the mechanism for the formation of the colloidal ^7Be species must be clearly understood.

Although the chemical composition of the ^7Be -related colloids in the cooling water at the accelerator facilities has not been clarified, it has been found that metal oxide or metal hydroxide colloids are generated in the cooling water by the corrosion of metal components. In the case of the cooling water system for the electromagnets at a 12-GeV proton accelerator facility, elemental analyses, electron microscope observations, and powder X-ray diffraction spectrometry clarified the dominant colloidal materials to be copper(II) or (I) oxides (*i.e.*, CuO or Cu₂O).¹¹ Oxygen-free copper is used as a coil material in the electromagnets. The cooling water flows through the Cu coil tubes and corrosion is possible at the large Cu/water interface area. Considering these facts, the formation process of ^7Be -related colloids in the cooling water for electromagnets is proposed to proceed *via*: (1) the formation of copper-oxide colloids by corrosion of copper in water and (2) the adsorption of ^7Be on the copper-oxide colloid surfaces.

The adsorption of ionic species on metal oxides in aqueous media is an important process in the transport of trace elements

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Table 1 Physical properties of CuO nanoparticles

	CIK	2810NH	US3063	US3065
Manufacturer	C. I. Kasei. Co., Ltd.	SkySpring Nanomaterials, Inc.	US Research Nanomaterials, Inc.	US Research Nanomaterials, Inc.
Purity, %	99.9	99	99.95	99
Average particle size/nm ^a	48	40	25 – 55	< 80
Density/g cm ⁻³ ^a	6.3	6.3 – 6.49	6.4	6.4
Surface –OH concentration/mol g ⁻¹ ^b	(3.7 ± 0.1) × 10 ⁻⁵	(3.3 ± 0.1) × 10 ⁻⁵	(1.1 ± 0.1) × 10 ⁻⁴	(8.0 ± 0.2) × 10 ⁻⁵
Average particle size (nm) dispersed in water ^b	600 ± 50	4000 ± 700	1000 ± 100	1000 ± 80

a. Cited from supplier's data.

b. Obtained in the present work.

and radionuclide species in various natural and engineering systems. The mechanism of adsorption of various metal ions by certain metal oxides, such as Al₂O₃, Fe₂O₃, FeOOH, and SiO₂, has been studied extensively.^{13–17} On the other hand, copper oxides have not been studied as potential adsorbents for metal ions or radionuclides because these oxides have been considered to be less important in these application fields. The adsorption of the Be(II) ion on metal oxides has also not been extensively investigated to date. Previous studies show that the mechanism of adsorption of metal ions on metal oxide particles can be explained by complex formation between the surface –OH sites on the metal oxides and metal ions in water.^{13–17} It has been found that the surface complexation constants of some metal oxides for metal ions and the hydration constants of metal ions show good correlation.¹⁷ Be(II) is recognized to be readily hydrated,^{18–20} and thus it can be expected that the surface –OH sites of metal oxides should have a large adsorptivity for Be(II) in water.

In the present work, the adsorption of Be(II) ions on copper(II) oxide (CuO) nanoparticles dispersed in water was quantitatively evaluated. By mixing aqueous Be(II) solutions with four kinds of CuO nanoparticles, the distribution ratio of Be(II) between the CuO nanoparticles and water was determined, and the complexation equilibria between the surface –OH sites and Be(II) ions were analyzed based on the pH dependence of the distribution ratio. The surface complexation constants were determined for four types of CuO nanoparticles. The results are discussed.

Furthermore, the mechanism of colloid formation of ⁷Be in the cooling water for the electromagnets at a high-energy proton accelerator facility is discussed by using the complex formation between the colloidal CuO particles and Be(II) in water as a model.

Experimental

Reagents and chemicals

Four kinds of CuO nanoparticles, denoted CIK, 2810NH, US3063, and US3065, were purchased and used for these studies. The manufacturers and physical properties of these particles are summarized in Table 1 and discussed in detail in the Results and Discussion section. Aqueous standard solutions of Be(II) were purchased from Kanto Chemical as 1000 mg/L atomic absorption standards, and were used after appropriate dilution. Water was purified with a Milli-Q Labo system (Millipore). Analytical-grade potassium hydrogen phthalate reagent was dried at 110°C for 4 h before use as a standard solution for acid-base titrations. Other chemicals were

purchased as analytical-grade reagents, and used without further purification.

Characterization of CuO nanoparticles dispersed in water

The surface –OH concentration per unit mass of the CuO nanoparticles was determined. CuO nanoparticles were mixed with appropriate amounts of water in a 10 mg cm⁻³ ratio of CuO to water. A 50 cm³ (CIK, 2810NH) or 20 cm³ (US3063, US3065) aliquot of the CuO/water suspension was placed in a glass beaker, set in a water bath (25 ± 0.2°C), and stirred with a magnetic stirrer. Acid-base titration experiments were performed by adding aliquots of a calibrated aqueous NaOH solution (1.23 × 10⁻² mol dm⁻³) or an aqueous H₂SO₄ solution (7.87 × 10⁻³ mol dm⁻³) to the CuO/water suspension. The pH of the suspension was measured with a glass electrode. Blank experiments were carried out using the same amount of pure water instead of the CuO/water suspension. By comparing the titration curves of the CuO/water suspension and pure water, the surface –OH concentration per unit mass of CuO nanoparticles (mol g⁻¹) was determined.

The zeta potential and particle size distribution of the CuO nanoparticles dispersed in water were measured by laser Doppler electrophoresis and dynamic light scattering (DLS) using a Horiba SZ-100 nanoparticle analyzer.²¹

Adsorption experiments of Be(II) on CuO nanoparticles in water

Aqueous solutions containing 1.0 × 10⁻⁷ mol dm⁻³ Be(II) were prepared. The pH of each solution was adjusted in the range of 5.5 to 6.6 with 0.01 mol dm⁻³ MES–NaOH buffer. The solution (20 cm³) was mixed with CuO nanoparticles (0.050 – 0.50 g) in a plastic vial. The vial was mechanically shaken for 48 h at 25 ± 0.2°C. The CuO nanoparticles were removed from the suspension by using a centrifugation-type ultrafiltration unit (Vivascience Vivaspin 20; MWCO 1000 kDa) with an estimated pore size of 16 nm based on the correlation between the molecular weight and estimated molecular size.²² The concentration of Be in the filtrate solution was determined by graphite furnace atomic absorption spectrometry with a Hitachi Z-5000 atomic absorption spectrometer, and the distribution ratio of Be(II) between the CuO nanoparticle phase and the aqueous phase was calculated. The pH of the filtrate solution was also measured with a glass electrode.

Results and Discussion

Properties of CuO nanoparticles dispersed in water

Certain physical parameters of the CuO nanoparticles used in the present study (under dry conditions) were specified by the

Fig. 1 Zeta potential *versus* pH plots for CuO nanoparticles dispersed in water. CuO nanoparticles: (A) CIK; (B) 2810NH; (C) US3063; (D) US3065; ionic strength: 0.01.

manufacturers. Herein, the zeta potentials, average particle diameters, and surface -OH concentrations were determined for the CuO nanoparticles in the CuO/water suspension state.

It is well known that the acid-base properties of -OH sites on metal oxide surfaces in water can be represented as the reactions expressed as follows¹³⁻¹⁷:

Figures 1(A) – 1(D) show the zeta potential of four kinds of CuO nanoparticles as a function of the pH at an ionic strength of 0.01. A steep decrease in the zeta-potential was observed around pH = 5 and 9 for all CuO nanoparticles. These results demonstrate that the acid-base reactions corresponding to Eqs. (1) and (2) progress around pH 5 and 9, respectively. Thus, the surface -OH sites on CuO are in the neutral >S-OH form in the pH range from 5 to 9. The particle sizes in the aqueous dispersion were found to be larger than those measured in the dry state, which implied aggregation of the CuO nanoparticles in water. Although the particle sizes differed for the four kinds of CuO nanoparticles, the respective particle sizes were nearly constant over the experimental pH range. Table 1 summarizes the manufacturer-specified physical parameters for the CuO nanoparticles in the dry state as well as the surface -OH concentration and the average particle diameter determined herein for the CuO/water suspension.

The following section discusses the properties of Be(II) adsorption by the CuO nanoparticles in the pH range from 5.5 to 6.6, where the surface -OH groups are in the neutral >S-OH form, and both the surface -OH concentration and the particle size are nearly constant.

Distribution behavior of Be(II) between water and CuO nanoparticles

In the present study, the partitioning of Be(II) between water and the CuO nanoparticles was evaluated in terms of the distribution ratio, *D*, defined by

$$D = \frac{\text{Amount of Be adsorbed on unit mass of the particles (mol g}^{-1}\text{)}}{\text{Concentration of Be in the aqueous phase (mol dm}^{-3}\text{)}}. \quad (3)$$

For all nanoparticles, the aqueous pH dependence of the distribution ratio is shown in Fig. 2. The log *D* value increases monotonously with increasing pH, and the magnitude of *D* at a given pH is dependent on the kind of nanoparticle. The *D* value at pH 6.0 follows the order: US3063 > US3065 > CIK > 2810NH. This order is consistent with the order of the surface -OH concentration (mol g⁻¹) of the CuO nanoparticles. The correlation between the distribution behavior of Be(II) and the surface -OH concentration of the CuO nanoparticles implies that the surface -OH sites participate in the sorption of Be(II) on the CuO nanoparticles.

The sorption of Be(II) on the CuO nanoparticles was evaluated based on the surface complexation model, in which the surface -OH sites on metal oxides form complexes with metal ions in

Fig. 2 Plots of $\log D$ as a function of the pH for the adsorption of Be(II) on various CuO nanoparticles in water. CuO nanoparticles (mass): ■, US3063 (0.050 g); ◇, US3065 (0.050 g); ●, CIK (0.50 g); △, 2810NH (0.50 g); Aq. soln., 20 cm³; initial Be(II) concentration, 4.5×10^{-8} – 9.2×10^{-8} mol dm⁻³; pH, 5.6–6.4; shaking time, 48 h.

water.¹³ In this study, the experimental data shown in Fig. 2 were analyzed according to the complexation equilibrium described below, in which the surface $>\text{S-OH}$ sites react with Be^{2+} to form $(>\text{S-O})_n\text{Be}^{(2-n)+}$ on the CuO surface:

The surface complexation constants, $\beta_{(s,n)}$, are defined by

$$\beta_{(s,n)} = \frac{[(>\text{S-O})_n\text{Be}^{(2-n)+}]_s [\text{H}^+]^n}{[>\text{S-O}]_s^n [\text{Be}^{2+}]}. \quad (5)$$

Here, $[\text{X}]$ and $[\text{X}]_s$ denote the concentration of a material X in the aqueous phase (mol dm⁻³) and that in the nanoparticle phase (mol g⁻¹), respectively. The coordination number of the surface $-\text{OH}$ groups to one Be atom corresponds to n ($n = 1 - 4$ for Be(II)). In the pH range of the present experimental condition, three kinds of Be(II) species (Be^{2+} , $\text{Be}(\text{OH})^+$, and $\text{Be}(\text{OH})_2$), are formed in the aqueous phase, depending on the pH. When the hydrolysis of Be(II) in the aqueous phase is considered, D can be expressed as Eq. (6), and further transformed to Eq. (7) using the hydrolysis constants of Be(II) in water ($K_1 = [\text{Be}(\text{OH})^+][\text{H}^+]/[\text{Be}^{2+}] = 10^{-5.7}$,¹⁹ $\beta_2 = [\text{Be}(\text{OH})_2][\text{H}^+]^2/[\text{Be}^{2+}] = 10^{-11.68\ 20}$):

$$D = \frac{\sum_{n=1}^4 [(>\text{S-O})_n\text{Be}^{(2-n)+}]_s}{[\text{Be}^{2+}] + [\text{Be}(\text{OH})^+] + [\text{Be}(\text{OH})_2]}, \quad (6)$$

$$D = \frac{\sum_{n=1}^4 \beta_{(s,n)} [>\text{S-OH}]_s^n / [\text{H}^+]^n}{1 + K_1/[\text{H}^+] + \beta_2/[\text{H}^+]^2}. \quad (7)$$

The following equation is derived from Eq. (7):

$$\log D + \log (1 + K_1/[\text{H}^+] + \beta_2/[\text{H}^+]^2) = \log (\beta_{(s,1)} [>\text{S-OH}]_s / [\text{H}^+] + \beta_{(s,2)} [>\text{S-OH}]_s^2 / [\text{H}^+]^2 + \beta_{(s,3)} [>\text{S-OH}]_s^3 / [\text{H}^+]^3 + \beta_{(s,4)} [>\text{S-OH}]_s^4 / [\text{H}^+]^4). \quad (8)$$

Fig. 3 Plots of $[\log D + \log (1 + K_1/[\text{H}^+] + \beta_2/[\text{H}^+]^2)]$ as a function of the pH for the adsorption of Be(II) on CuO nanoparticles in water and least-squares fitting analyses based on Eq. (9). CuO nanoparticles: ■, US3063; ◇, US3065; ●, CIK; △, 2810NH.

The left-hand-side of Eq. (8) was plotted *versus* pH, as shown in Fig. 3. For all of the CuO particles, the plots were nearly linear, and the slopes of the plots were between 2 and 3. These slope values imply that two kinds of complexes, $(>\text{S-O})_2\text{Be}$ and $(>\text{S-O})_3\text{Be}^-$, are the dominant Be(II) species formed on the surface of the CuO nanoparticles. Herein, Eq. (8) can be simplified as follows:

$$\log D + \log (1 + K_1/[\text{H}^+] + \beta_2/[\text{H}^+]^2) = \log (\beta_{(s,2)} [>\text{S-OH}]_s^2 / [\text{H}^+]^2 + \beta_{(s,3)} [>\text{S-OH}]_s^3 / [\text{H}^+]^3). \quad (9)$$

The surface $-\text{OH}$ concentrations, $[>\text{S-OH}]_s$, shown in Table 1 were substituted into Eq. (9). Thus, the values of $\beta_{(s,2)}$ and $\beta_{(s,3)}$ were obtained by nonlinear least-squares fittings of the plots in Fig. 3 according to Eq. (9), and are summarized in Table 2. As described in the Introduction section, both surface complexation properties of CuO for metal ions and the adsorption of the Be(II) ion on various metal oxides have not been studied extensively. Furthermore, the definitions and units of the surface complexation constants defined in Eq. (5) are not the same as

Table 2 Surface complexation constants for the adsorption of Be(II) on CuO nanoparticles in water ($\beta_{(s,2)}$, $\beta_{(s,3)}$)

	CIK	2810NH	US3063	US3065
$\log \beta_{(s,2)}$	-4.03 ± 0.12	-3.82 ± 0.09	-4.36 ± 0.22	-4.03 ± 0.06
$\log \beta_{(s,3)}$	-5.17 ± 0.05	-5.44 ± 0.11	-6.13 ± 0.13	-6.10 ± 0.08

those of other literature values.^{13–17} Thus, the values of the surface complexation constants in Table 2 cannot be directly compared with the appropriate reported equilibrium constants. Our recent experiments demonstrate that the surface complexation of CuO for Be(II) is smaller than those of other metal oxides: Al₂O₃, Fe₂O₃, and SiO₂.²³ The discussion in the view point of differences in metal oxides and metal ions will be reported in following reports.

In this section, the $\beta_{(s,2)}$ and $\beta_{(s,3)}$ values for four types of CuO nanoparticles are compared. The $\beta_{(s,2)}$ values are similar for all four kinds of CuO nanoparticles. As shown in Table 1, the particle sizes dispersed in water are different between four kinds of CuO nanoparticles; thus, the similarity in $\beta_{(s,2)}$ values for the different CuO particles imply that formation of ($>$ S-O)₂Be complexes are not largely affected by the particle size and the morphology of the particles. On the other hand, the $\beta_{(s,3)}$ values vary with the kind of CuO nanoparticle. The differences in $\beta_{(s,3)}$ may be related to the structure or the conformation of the -OH sites on the CuO nanoparticles, which may affect the formation equilibrium of ($>$ S-O)₃Be⁻ complexes. Herein, both the $\beta_{(s,2)}$ and $\beta_{(s,3)}$ values are similar for US3063 and US3065, which may have been synthesized by the same manufacturer using similar methods.

The dependence of the complexation constants on the kind of CuO nanoparticles is not clearly understood at present. More comprehensive investigations for characterizing the particles by physical/chemical analytical techniques and microscopic observations would be valuable for clarifying the characteristic surface-complexation properties for each CuO nanoparticle.

Colloid formation behavior of ⁷Be in the cooling water for electromagnets at a high-energy proton accelerator facility

In this section, the colloid formation behavior of ⁷Be in the cooling water for the electromagnets at a high-energy proton accelerator facility is discussed in the context of the surface complexation reactions between the CuO colloid and the Be(II) ions. Although other mechanisms, such as the co-precipitation of Be(II) with other metal ions, are also possible processes in the colloid formation of ⁷Be in the circulating cooling-water, we focus on evaluating the contribution of the surface complexation processes in colloid formation by ⁷Be in a typical cooling water system for electromagnets at high-energy accelerator facilities.

At the EP1 beamline in the 12 GeV proton accelerator facility of the High Energy Accelerator Research Organization, Japan (KEK-EP1), several tens of electromagnets were cooled by about 20 m³ of pure water circulating inside the copper coils of the magnets. The total flow rate of the circulating water was about 2.0 m³ min⁻¹, and the purity of the water was maintained by partially circulating the water through an ion-exchange unit filled with a mixture of cation-exchange resin and anion-exchange resin. The pH of the circulating water was around 6 and the temperature was maintained within the range of 20 – 35°C, depending on the accelerator operation conditions.

In the cooling-water system for electromagnets at KEK-EP1, the existing forms of ⁷Be in the water had been studied *via* ion-exchange column/ultrafiltration techniques, and were found

to be as follows: soluble cationic species: 38%; fine colloidal species (3 – 7 nm): 57%; relatively-large colloidal species (7 – 200 nm): <1%; particulate species (>200 nm): 5%.¹¹ Furthermore, elemental analysis of the cooling water sample showed that 120 µg dm⁻³ of Cu was contained in the water, most (~90%) of which was present as fine colloidal species (3 – 7 nm).¹¹ The colloidal Cu-related particles were assumed to be chemically comprised of CuO or Cu(OH)₂ in water at neutral pH. In the following discussion, the colloidal Cu-related particles were assumed to be in the form of CuO; thus, the concentration of CuO colloidal particles was estimated to be approximately 135 µg dm⁻³.

As a simplest model, spherical morphology and uniform diameter are assumed for CuO colloidal particles in the cooling water as well as the CuO nanoparticles used for the present adsorption experiments. When the radius and density of a spherical particle are represented by r and ρ , respectively, its surface area and mass are $4\pi r^2$ and $4\pi r^3 \rho / 3$. Thus, the specific surface area per unit mass, S_m , for the spherical particle can be expressed as follows:

$$S_m = (4\pi r^2) / (4\pi r^3 \rho / 3) = 3 / (r\rho). \quad (10)$$

The CuO nanoparticles from CIK have an average particle diameter of 600 nm in water (Table 1). These nanoparticles are considered to be formed by the aggregation of fine CuO spherical particles with average diameters of 48 nm, where the gaps between the CuO spheres are filled with water. Thus, the density of the CuO nanoparticles in water should be lower than the density of pure (dry state) CuO nanoparticles (6.3 g cm⁻³). Although the structure of the CuO nanoparticles formed in water has not been clarified, if we assume that CuO nanoparticles comprise a hexagonal close-packed (hcp) array of CuO spheres with uniform diameters (48 nm), the packing efficiency of the CuO spheres is 0.74; thus, the apparent density of CuO nanoparticles in water (600 nm) corresponds to 4.9 g cm⁻³. Therefore, if the values of r and ρ for the CIK CuO nanoparticles are assumed to be 300 nm and 4.9 g cm⁻³, respectively, the S_m value for the CIK CuO nanoparticles in water is calculated to be 2.0 m² g⁻¹. Considering the surface -OH concentration of 3.7×10^{-5} mol g⁻¹ (Table 1) and mass abundance of CuO in the aggregated nanoparticles (0.95), the surface -OH density per unit surface area (mol m⁻²) can be determined to be 1.8×10^{-5} mol m⁻².

On the other hand, the radius of the CuO colloidal particles formed in the present cooling water has been estimated to be 3.5 nm from the colloid size distribution profiles in the cooling water.¹¹ The density of the CuO colloidal particles in water is assumed to be of the same as that of pure CuO nanoparticles (6.3 g cm⁻³), consequently the S_m value for the CuO colloidal particles in the cooling water can be estimated to be 140 m² g⁻¹. Assuming that the surface -OH density per unit surface area is common to the CIK CuO nanoparticles in water and the CuO colloidal particles formed in the cooling water, the surface -OH concentration, [$>$ S-OH]_s (mol g⁻¹), for the colloidal particles in the cooling water is calculated to be 2.5×10^{-3} mol g⁻¹. By using the [$>$ S-OH]_s value as well as the values of $\beta_{(s,2)}$ ($10^{-4.03}$) and $\beta_{(s,3)}$ ($10^{-5.17}$) (Table 2), the distribution ratio, D , for partitioning of Be(II) between the CuO colloidal particle phase and water is calculated to be 2.1×10^4 dm³ g⁻¹ from Eq. (9).

The fraction of Be(II) adsorbed on the CuO colloidal particle, $Ads_{(col)}$ (%), in the cooling water can be evaluated by using

$$Ads_{(col)} (\%) = D [CuO]_{col} / (1 + D [CuO]_{col}) \times 100 (\%) \quad (11)$$

where $[CuO]_{col}$ denotes the concentration of CuO in the cooling water ($135 \mu g dm^{-3}$).¹¹ The $Ads_{(col)}$ (%) value is calculated to be 74%, which is not significantly different from the empirically determined fraction of ^7Be existing as a fine colloid (3 – 7 nm) (57%).¹¹ When other commercial CuO nanoparticles are selected as the model nanoparticles, similar results are obtained. Therefore, it can be concluded that the colloid formation of ^7Be in the cooling water for electromagnets can be explained in terms of the complexation between the surface –OH sites on the CuO colloidal particles and Be(II) ions.

Conclusions

The complexation equilibrium between the surface –OH sites of CuO nanoparticles and Be(II) in water was quantitatively analyzed. Detailed analyses of the experimental adsorption data demonstrated that $(>S-O)_2Be$ and $(>S-O)_3Be^-$ were the dominant Be(II) species formed at the CuO surface in water. The overall surface complexation constants, $\beta_{(s,2)}$ and $\beta_{(s,3)}$, were determined for four kinds of commercial CuO nanoparticles, and the results demonstrated that the $\beta_{(s,2)}$ value for $(>S-O)_2Be$ was almost constant, whereas the $\beta_{(s,3)}$ value for $(>S-O)_3Be$ was dependent on the type of CuO nanoparticle.

Based on the properties of surface complexation between CuO and Be(II), the colloid formation behavior of ^7Be in the cooling water for electromagnets at a high-energy proton accelerator facility was discussed. Calculations performed herein by invoking certain assumptions demonstrated that the colloid formation behavior of ^7Be in the cooling water for electromagnets can be explained by considering complex formation between the CuO colloidal particles and Be(II) ions in water.

More detailed analysis of the colloidal species, such as the chemical composition, colloid size distribution, and detailed morphological analysis of the colloids, may be required for clarifying the colloid formation behavior of radionuclides in the cooling water at various accelerator facilities. However, the approach proposed in this study should be useful for understanding the behavior of various radionuclides in the cooling water of accelerator facilities. A more comprehensive study concerning the adsorption of metal ions on metal oxide nanoparticles is now under way.

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