

pH Dependence of Cs(I) and Sr(II) Coprecipitation With Fe(III) in Aqueous Solutions

TaeJun Kim^{1,2}, Minsik Kim^{1,2}, and Jei-Won Yeon^{1*}

¹Korea Atomic Energy Research Institute, 111, Daedeok-daero 989beon-gil, Yuseong-gu, Daejeon, Rep. of Korea

²University of Science & Technology, 217, Gajeong-ro, Yuseong-gu, Daejeon, Rep. of Korea

*yeonysy@kaeri.re.kr

1. Introduction

A severe accident such as at Fukushima produces significant quantities of radioactive waste water. The water may contain non-volatile iodide (I^-) as a major fission product anion [1] and its oxidized form, molecular iodine (I_2), can be formed under irradiation. Although various techniques for the radioactive waste water treatment have been developed and used [2], a challenge still remains in keeping the volatility of radioactive iodine species low during the treatment. The water pH should be high enough for I_2 to decompose into non-volatile iodine species; such high pH conditions will also promote the formation of solid metal (hydr)oxides such as iron(III) (hydr)oxides. On the other hand, Jung et al. [3] demonstrated that one of water radiolysis products, hydrogen peroxide, can reduce I_2 to I^- at pH higher than 6. Thus, H_2O_2 addition to the waste water (pH > 6) may relieve the difficulty.

In this context, this study was conducted to validate that the optimum pH for treatment of radionuclides in the water is about 7. Only the formation of I_2 and the precipitation of Fe(III), Cs(I), and Sr(II) at room temperature were considered here.

2. Materials and methods

2.1 Gamma irradiation to iodide solution

To confirm that the concentration of I_2 significantly decreases under irradiation as pH increases above 6, an experiment was conducted using a cobalt-60 gamma irradiator (MDS Nordion). The aqueous solutions of 5 mol dm^{-3} sodium iodide at various initial pH were irradiated (8 kGy); their pH

were adjusted using hydroiodic acid and sodium hydroxide (NaOH) solutions. The triiodide concentration, which is linearly proportional to the concentration of I_2 with excess I^- , was measured, using a UV-Vis spectrophotometer (Biochrom).

2.2 Metal coprecipitation

The mass ratios of Cs and Sr to Fe in the precipitates formed in the aqueous solutions (pH 7 to pH 10; NaOH for pH adjustment) of ferric chloride ($FeCl_3$), and caesium chloride ($CsCl$) or/and strontium chloride ($SrCl_2$) were determined using inductively coupled plasma spectrometers. The molarity of Fe(III), Cs(I), and Sr(II) were (36, 18, 9) mmol dm^{-3} , respectively. The spectrometry samples were the residues obtained by filtration of the solutions using Whatman Grade 541 filter paper, dried in a drying oven at 103°C for three hours, and then dissolved in 1 mol dm^{-3} hydrochloric acid.

Sodium iodide (99.5%), $CsCl$ (99.9%), $SrCl_2$ (99.9%), $FeCl_3$ (97%), NaOH (97%), hydroiodic acid (57% in mass fraction), and hydrochloric acid (35% in mass fraction) used for this study were all purchased from Sigma-Aldrich.

3. Results and discussion

3.1 pH dependence of volatile iodine formation

The concentration of triiodide in the gamma-irradiated samples at pH (2.8, 6.1, 6.4, 6.5, 7.0) was (0.42, 0.37, 0.09, 0.02, 0.00) mmol dm^{-3} . The results confirm that the concentration of I_2 is drastically reduced under irradiation as the solution pH increases above 6.

3.2 pH dependence of metal coprecipitation

As Bull et al. [4] mentioned, linear relationships between the Sr–Fe mass ratio and the initial pH are found with coefficients of determination greater than 98% (see Fig. 1). The Cs–Fe mass ratios were only a few percent at most and were not linearly dependent on the initial pH. With Sr(II), Cs(I) precipitated less than without Sr(II). The smallest Sr–Fe mass ratio was found at the solution pH about 7, the lowest experimental pH.

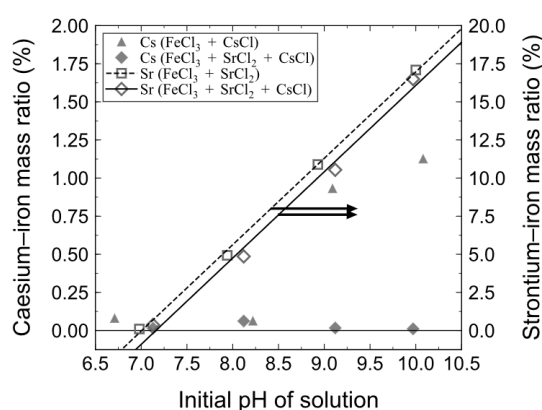


Fig. 1. Metal coprecipitation experiment results.

4. Conclusion

It appeared valid at least for the experimental conditions that the optimum pH for treatment of radionuclides in the waste water is about 7. Experiments on the aqueous solutions more alkaline and those containing other chemical species should be conducted to determine the relationship between the solution pH and the coprecipitation of Cs(I) and Sr(II) with Fe(III) under the conditions closer to the waste water.

ACKNOWLEDGEMENT

This work was supported by the National Research Foundation of Korea (NRF) grant funded by the Korean government (MSIT: the Ministry of Science and ICT) (No. 2017M2A8A4015281).

REFERENCES

- [1] C.-C. Lin, and J.-H. Chao, “Reassessment of Reactor Coolant and Iodine Chemistry under Accident Conditions.” *Journal of Nuclear Science and Technology*, 46, 1023–1031 (2009).
- [2] P. Sylvester, T. Milner, and J. Jensen, “Radioactive Liquid Waste Treatment at Fukushima Daiichi,” *Journal of Chemical Technology and Biotechnology*, 88, 1592–1596 (2013).
- [3] S.-H. Jung, J.-W. Yeon, S. Y. Hong, Y. Kang, and K. Song, “The Oxidation Behavior of Iodide Ion Under Gamma Irradiation Conditions,” *Nuclear Science and Engineering*, 181, 191–203 (2015).
- [4] P. S. Bull, J. V. Evans, and R. J. Knight, “Removal of Radioactive Strontium from Water by Coagulation–Flocculation with Ferric Hydroxide.” *Journal of Applied Chemistry and Biotechnology*, 25, 801–807 (2007).