



Removal of radioactive iodide from simulated liquid waste in an integrated precipitation reactor and membrane separator (PR-MS) system



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ABSTRACT

A novel integrated precipitation reactor (PR) and membrane separator (MS) system was developed for the removal of radioactive I^- from simulated liquid waste. The PR-MS system was evaluated at the lab-scale, the I^- removal efficiency reached approximately 97.0% and the concentration factor (CF) value was 1050 when the concentration of added Na_2SO_3 was 40 mg/L and the CuCl dosage was 260 mg/L. The addition of Na_2SO_3 played an important role in the removal of dissolved oxygen (DO) and the formation of CuI. These reactions, including the oxidation of SO_3^{2-} , the dissociation of CuCl, the disproportionation of Cu^+ , and the formation of CuI and Cu_2O , occurred in the PR. Furthermore, the influence of the concentration of added CO_3^{2-} on copper removal was investigated, and solid-liquid separation and copper removal were simultaneously attained in the MS. The I^- removal efficiency gradually decreased from 97.0% to 58.5% and the Cu^{2+} concentration in the effluent decreased from 18.71 mg/L to 0.719 mg/L with the addition of CO_3^{2-} at a concentration that ranged from 0 to 80 mg/L. Experimental results show the PR-MS system is of great potential for industrial application on treatment of liquid waste contaminated with radioactive I^- .

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1. Introduction

Radioactive element could pose a risk to the ecosystem and public health once it is released to the environment, its potential hazards are due to chronic health effect, carcinogenic and mutagenic characteristics. Radioactive iodine is one of toxic radioactivity elements because it can lead to an increase in thyroid cancer, leukemia and metabolic disorders, especially in children [1–3]. Iodine radioisotopes produced during uranium and plutonium fission reactions have been identified as one of the most dangerous radioactive substances in terms of the radiological effects of an accidental release [1,2,4–7]. For instance, radioiodine is a major risk of groundwater near the Hanford Site in the U.S. which is a former plutonium production site [8,9] and radioiodine contamination was detected in the surface water, drinking water, seawater and underground water in Japan after the Fukushima nuclear power plant accident [10–13]. In addition to the release from nuclear power plants, radioactive iodine is widely applied in medical diagnosis and treatment and is consequently one of the most often detected radioactive species in aquatic environments

receiving sewage effluent discharges [14–19]. Therefore, the removal of radioactive iodine from water is of great importance.

In general, radioactive iodine is released mainly in the form of cesium iodide from a light water reactor and iodide is relatively stable almost over the entire pH range [18,20,21]. Various methods have been proposed for the treatment of water contaminated with radioactive I^- [3,14,18,22–31], for example, ultrafiltration and reverse osmosis were applied for removing radioactive iodine from liquid wastes [14,29], powdered and silver-impregnated activated carbon, chalcogen-based aerogels and Ag_2O grafted titanate nanolamina as adsorbents were examined to adsorb radioactive iodine from water [3,22,23,31], the I^- removal from aqueous solutions was also investigated by polyethylenimine-epichlorohydrin resins [27]. All methods must minimize the volume of radioactive substances for stabilization and subsequent long-term storage or final disposal [29,32]. Compared to these methods, precipitation is an easy and efficient process that is suitable for treating large volumes of water with high salts [32]. One reported approach for the removal of I^- is the use some of compounds containing Hg^{2+} , Bi^{3+} , Ag^+ and Cu^+ , which interact with I^- to form precipitates [18,24–26,33,34]. These precipitates are then separated from the solutions and safely disposed. The Cu^+ ion is the most promising cation due to its relatively low toxicity and inexpensiveness [18,33,35], Lefèvre et al. and Kamensaya et al. reported that

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radioactive I^- could be removed by Cu_2O , Cu_2S , metallic copper and cupric compounds, Cu^+ -substituted NaX and NaA zeolites [25,33,36,37]. Our previous work based on a filter cup study suggested that at a low dosage and short reaction time, $CuCl$ could efficiently precipitate and remove radioactive I^- from water in the absence of DO [38]. In addition to our previous work, the removal of radioactive I^- by precipitation has also been studied using the jar test method [18,22,25,26,28,33,34,36,39]. However, experiments based on this method are not sufficient to address real radioactive water pollution such as the radioactive iodine that was detected in drinking water in 15 of 47 prefectures after the Fukushima nuclear accident [10,23]. Additionally, the removal efficiency of conventional precipitation process depends largely on the solid-liquid separation because precipitation process only transforms the target element from the liquid phase to solid one that may be in small size, and the combination of precipitation with another method (such as flocculation, flotation and membrane separation) can improve the removal efficiency. Furthermore, membrane technologies are increasingly being used to treat radioactive iodine, especially for the treatment of radioactive water, in order to improve effluent quality and reduce the radioactive waste volume for final disposal [14,16,29,32,40,41].

The purpose of this study was to (1) develop and evaluate a novel integrated precipitation reactor and membrane separator (PR-MS) system for the removal of radioactive I^- at the lab-scale to obtain high removal efficiency and reduce the volume of precipitates; (2) understand the I^- removal mechanism and possible chemical reactions occurred in the PR; (3) investigate the effect of added Na_2CO_3 concentrations on copper removal in the MS to reduce copper concentrations in the effluent.

2. Materials and methods

2.1. Experimental set-up and procedure

The flow chart of the PR-MS system used in this study is shown in Fig. 1. The entire system included two units: the precipitation (PR) unit and the membrane separation (MS) unit, the deoxygenation was performed in the PR unit. The PR-MS process was operated under closed conditions to avoid the dissolution of oxygen in air in the simulated liquid waste. The PR-MS system was operated in three steps: (1) the removal of DO from simulated liquid waste to minimize the interference of DO with the reaction system due to its strong oxidization (in other words, all reactions took place under N_2 protection); (2) the transfer I^- from the liquid phase to

the solid phase via the formation of precipitate particles and the subsequent removal of I^- by the sedimentation of large particles in the PR and filtration of fine particles in the MS; (3) the reduction of copper concentration in the effluent from the MS via the addition of Na_2CO_3 .

The continuous operation of the PR-MS system was controlled by a programmable logic controller (PLC). All pumps were interconnected via the PLC with liquid level probes. The PR-MS system was operated under a closed system, and the pressures of the PR and the MS were respectively maintained at 3–6 kPa and 1–3 kPa by pressure sensors in order to avoid oxygen in air dissolving into the raw water. When the raw water was pumped into the PR, N_2 was bubbled into the raw water at a given rate for 6 min and Na_2SO_3 was then added at a given dose, resulting in a DO concentration of less than 0.2 mg/L in the PR. After the deoxygenation process, a predetermined dose of $CuCl$ was added to the PR either in the form of a suspension prepared by dosing $CuCl$ into a dilute HCl solution in advance, or using solid powder dosing device (5812 DOSER powder doser, LAMAD laboratory instruments, Switzerland). The effects of the two modes of $CuCl$ dosing on I^- removal were compared. The reaction mixture in the PR was stirred for 15 min, the mixed liquor was allowed to settle for 8 min and a part of supernatant was pumped into the MS. The PR was operated as semi-batch mode, but the MS was operated continuously. In terms of the entire treatment process, the experiments were conducted by a continuous flow system. The MS was continuously aerated by a N_2 gas blower at a flow rate of 0.15 m^3/h , the off-gas from the MS was dried and reused as an inflow of the N_2 gas blower. The effluent was aspirated from the MS using an effluent pump (8 min filtration and 2 min pause). The effluent from the continuous operation process was collected every 24 h into a container and mixed homogeneously to obtain representative results. The sample was measured and reported as the result of the day. In the copper removal test, the PR effluent was collected at the end of the quiescent stage and the supernatant was then filtered with 0.22 μm membrane. The PR-MS system was operated for 2 days for each dosage of Na_2CO_3 , and the effluent sample in the MS was collected and measured on the second day of operation.

The simulated liquid waste containing I^- , which was the raw water, was prepared by dissolving the required concentration of KI in tap water. All isotopes of a given element have the same number and arrangement of electrons, and the chemical characteristics of a radionuclide are identical to those of its stable isotope [5,42]. Radioiodine level was extremely low in the radioactive water, adding the inactive iodine to the solution increases the total isotope concentration so that the solubility product of CuI , K_{sp} , can be exceeded. When using inactive I^- as the carrier to mix with radioactive I^- in an aqueous solution, the iodide isotopes may exhibit almost identical chemical behavior. Thus, the I^- removal efficiency should be the same for inactive and radioactive I^- . Initial I^- concentration in the raw water was set at 5 mg/L according to our previous study which indicated that the removal efficiency of I^- was almost not affected by initial I^- concentration ranged from 5 to 40 mg/L and the I^- concentration in the effluent was lowest when initial I^- concentration was 5 mg/L [38]. The main parameters of the tap water are listed in Table 1. The effective volumes of the PR and the MS were 9 L and 15 L, respectively, and the

Table 1
The main parameters of tap water.

Parameters	Value	Parameters	Value
Na^+	7.351–8.080	SO_4^{2-}	33.18–36.39
K^+	3.364–5.885	Cl^-	9.556–11.18
Ca^{2+}	37.18–37.28	NO_3^-	5.588–6.726
Mg^{2+}	9.139–9.415	pH	7.40–7.68

Note: the unit of ion concentration was mg/L.

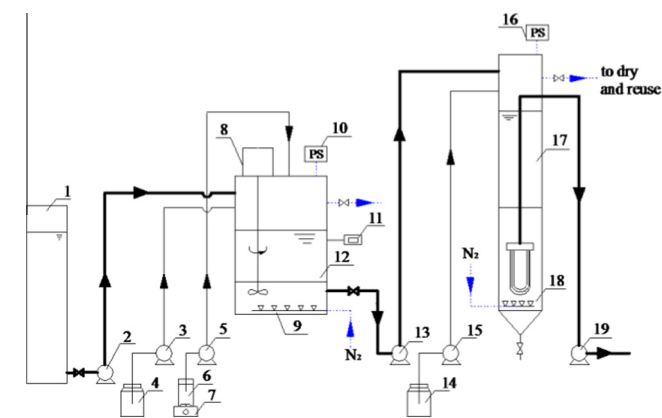


Fig. 1. Schematic illustration of the PR-MS system 1-raw water tank; 2, 3, 5, 13, 15 and 19-pump; 4-vessel of Na_2SO_3 solution; 6-vessel of $CuCl$ suspension; 7-magnetic stirrer; 8-electric stirring; 9 and 18-gas diffuser; 10 and 16-pressure sensor; 11-DO probe; 12-precipitation reactor; 14-vessel of Na_2CO_3 solution; 17-membrane separator.

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