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A New Technology Developed to Remove Aqueous Radioiodine

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1. Introduction

The global recycle and contamination of anthropogenic radioiodine is originated from human activities in nuclear, industrial, and medical fields [1]. Recently, the worldwide inventory of iodine isotope wastes continues to increase [2]. Radioiodine is considered as a critical radionuclide by the International Commission on Radiological Protection (ICRP) in terms of the radiation and radiotoxicity risks to nature and human health. In addition, there were reports about great increases of thyroid cancer in children in Belarus from 1990 [3]. In natural environments, there were several reports for the unusual increase of radioactive iodine ratios in most environmental samples (i.e. soils, vegetables, and seafoods) [4]. Iodine usually exists as iodide, iodate, and organic iodine in the environment. Iodide is considered as the most difficult form among the iodine species to treat by the conventional adsorption-based technology. This is why urgent new technology is needed to suppress such a progressive accumulation of radioiodine in natural environments.

2. Methods and Results

2.1 Methods

The strain of *Desulfosporosinus auripigmenti* isolated from natural bentonite in Korea was used in our experiment as a sulfate-reducing bacterium (SRB). To study a selective iodide removal from anions-rich soluton, we anaerobically prepared various anions-rich solutions including Cl^-, CO_3^{2-}, and SO_4^{2-} in serum bottles. From the iodide stock solution, 1 mM sodium iodide was aseptically added to the serum bottles containing the above anions. To perform a practical test using iodine radioisotope, a stock solution of radioactive ^{125}I (half-life of 60 days) was prepared in 185 MBq/l from a reagent of 5 mCi $Na^{125}I$ (Perkin Elmer, Inc.). From the radioactive stock solution, ^{125}I was injected to be 2.2 MBq/l radioactivity. Finally, cells were injected into the serum bottles providing ca. 0.75 mg/l cell protein, to determine the microbial effect on the removal of iodide. Reducible cationic copper was chosen and slightly provided as an electron acceptor and counterion in the iodide-containing media in less than 1.0 mM of $Cu(NO_3)_2 \cdot 3H_2O$. The overall solution pH was ~7.3. Then, the prepared serum bottles were put into a rotary-shaker (120 rpm at 30℃) in the dark for 10 days.

2.2 Removal of Aqueous Iodide

Aqueous iodine species normally exist as iodide (I^-) and iodate (IO_3^-) depending on redox conditions in solution. Under reducing environment, aqueous iodine generally exists as I^-. An experiment for a removal of soluble iodide (I^-) from anions-rich solutions was conducted at near-neutral pH. We have attempted to determine how background anions (Cl^-, CO_3^{2-} and SO_4^{2-}) influence the behavior of iodide under anaerobic conditions. There was little decline of soluble iodide concentrations in the absence of adsorbents. However, a particular decrease of iodide was observed in a solution with bacteria and trace copper even under highly competitive anion concentrations. The initial iodide concentration (1 mM) rapidly dropped to a very low level by just adding SRB and trace copper. Results indicated that equimolar mixture of natural common anions did not appreciably interfere with the iodide selectivity.

The decrease of iodide concentration contrasted with the increase of SRB population in the medium. Its microbial growth was considerable as compared to the other one. It is assumed that the trace copper would have positively functioned as an electron acceptor by being transformed from cupric ion (Cu^{2+}) to cuprous ion (Cu^{1+}) in the microbial redox system. Copper(II) is a transition elemental component that can switch to copper(I) by accepting free electrons in aqueous solution.

A large amount of radioiodine with high radioactivity in water that contains competitive anions was targeted to be reduced by our system, and consequential real test was nicely performed by dropping a strong ^{125}I radioactivity (2.2 MBq/L) to a near non-detectable level in several days. In general, the radioactivity of iodine does not easily decrease in the highly competitive anion conditions. The previous studies revealed that several aqueous anions, such as chloride and carbonate, could be strong competitors to I^-. In our system, however, the selective sequestration of radioactive $^{125}I^-$ under enzymatic Cu-reducing system was almost not influenced by Cl^- and CO_3^{2-} as well as OH^- species. More interestingly, water-soluble iodide (^{125}I) could be separable from the anions-rich water as solid phase.

Recently, Ag^+-doped materials (e.g., Ag-zeolite) have been attracting attention among getter materials. They have shown a good uptake of I^-, but they have several shortcomings to be applied to bulky wastewaters because of their high usage costs and limited capacities induced from the chemical adsorption.

Our method that is proposed here, however, is leading to a drastic cost-cutting way by using copper (Cu^{2+}) that is cheaper than silver (Ag^+). Furthermore, the selective sequestration of iodide can lower disposal costs by largely reducing radioactive waste volumes.

3. Conclusions

The conventional methods based on the anion-exchange and adsorption techniques have been difficult to manage the growing release of radioiodine for decades. Here we suggest a new solution that uses a biostimulant way to isolate radioiodine. We discovered that natural SRB can play an important intermediary role to get anionic iodide and cationic copper combined strongly as perfect counterions by enzymatic Cu-reduction. Through the process of biocatalysis, the halide nuclei gain a drive for growth with iodide selectivity and then precipitate in the solution.

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