

## **Radio-iodine biocrystallization based on microbial redox reactions**

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The releasing amounts of radionuclides and radioactive wastes from various nuclear and medical fields are causing public concern due to their short- and long-term radiotoxic impact on nature. In particular, the anthropogenic radio-iodine that has been released into nature for decades has become one of important issues due to its global and long-term recycling, which affects the worldwide ecosystem and even human health.

Aqueous iodine species in water commonly exist as iodide ( $I^-$ ) and iodate ( $IO_3^-$ ), depending on the redox conditions. We report a new biostimulant way to effectively remove radio-iodine that exhibits remarkable selectivity for the highly difficult-to-capture radio-iodine of >500-fold over other anions ( $CO_3^{2-}$ ,  $SO_4^{2-}$ ,  $OH^-$ ,  $Cl^-$ ), even under circumneutral pH. We discovered an interesting mechanism by which microbially reducible copper (i.e.,  $Cu^{2+}$  to  $Cu^+$ ) acts as a strong binder for iodide-iodide anions to form a crystalline halide salt of CuI that is highly insoluble in wastewater. During the microbially induced Cu reduction (redox reaction), characteristic halide crystals of CuI were generated as solid phase grown to micrometer-sizes, even in the low iodine concentrations (i.e.,  $I^- < 1$  mM).

The merit of the crystallization of iodine with copper is to fix the iodine in a structurally rigid framework, preventing  $I^-$  remobilization later on. For example, malachite ( $Cu_2(OH)_2CO_3$ ), which is a native mineral that is naturally synthesized by iodine and copper, is found to be a stable solid phase that has been preserved for long geologic times in the field. This strong natural evidence indicates that the biocrystallization of radio-iodine is a promising isolation method because the immobile solid phase is secure and stable in natural environments.