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Coupled reducion-amalgamation processes on silica-supported gold nanoparticles for mercury sequestration

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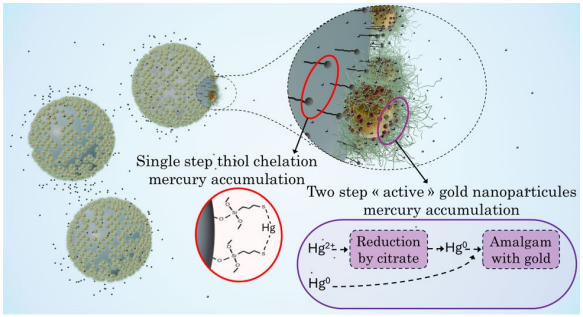

Mercury remains a priority contaminant due to its toxicity, redox complexity, and instability during environmental sampling and storage. Current monitoring approaches typically require large-volume collection, external reduction steps, and laboratory-based amalgamation procedures, increasing logistical constraints and introducing potential biases. Here, we report a redox-active silica core-gold nanoparticle satellite architecture (Si@AuNP) designed for direct, in situ capture and stabilization of both Hg(0) and Hg(II) in aqueous systems.

The hybrid material consists of thiol-functionalized silica microspheres decorated with 13 nm citrate-stabilized Au nanoparticles, subsequently reinforced with PEG diamine to enhance colloidal stability under high ionic strength conditions. This architecture enables three simultaneous and synergistic mechanisms: (i) direct amalgamation of elemental Hg(0) on gold surfaces, (ii) citrate-mediated in situ reduction of Hg(II) to Hg(0), and (iii) residual thiol-Hg(II) complexation. Thermogravimetric analysis quantified the surface citrate content (0.59 wt%), corresponding to a theoretical Hg(II) reduction capacity of 6.3 mg $Hg.g^{-1}$ material, consistent with experimentally observed uptake plateaus.

Across concentrations ranging from 1 $\mu g.L^{-1}$ to 10 $mg.L^{-1}$, Si@AuNPs exhibited 3-7 fold higher mercury accumulation compared to thiolated silica controls. Under strongly acidic conditions (pH 0.2), Hg(II) uptake decreased significantly, confirming the central role of citrate-mediated reduction. Validation in certified seawater (16.8 ± 1.1 $ng.L^{-1}$ Hg) yielded quantitative recovery (16.0 ± 3.6 $ng.L^{-1}$), demonstrating robustness in complex saline matrices.

Importantly, storage experiments revealed severe losses of dissolved Hg(0) in conventional containers (22% recovery after 30 h), whereas preconcentration on Si@AuNPs retained 73%, substantially improving sampling reliability. Owing to their micron-scale architecture, these structures are compatible with membrane filtration platforms, facilitating integration into field-deployable solid-phase extraction systems.

This work demonstrates how engineered hybrid nano-interfaces can couple redox transformation, amalgamation, and complexation to address both speciation control and practical deployment challenges in environmental mercury monitoring and remediation.