Field-Deployable Nanosensing Approach for Real-time Detection of Free Mercury Speciation and Quantification in Surface Stream Waters and Groundwater Samples at the U.S. DOE Contaminated Sites

University Lead Research

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Our proposition targets a critical element of the Environmental Remediation Science Program (ERSP) research portfolio, which is the development of enabling scientific tools for characterizing the spatial and temporal evolution of complex subsurface systems. We are developing a field-portable, on-site sensing device for real-time speciation of elemental and inorganic mercury in surface stream waters, groundwater, and sediment samples. The sensing device relies on the combination of two well-known phenomena, i.e. the amalgamation of metallic mercury (Hg(0)) and gold (Au) and the Surface Plasmon Resonance (SPR) of gold nanorods (Au NR). When Au NR are exposed to the presence of Hg(0), its amalgamation to Au causes a reduction of the effective aspect ratio of NR and a blue shift of the maximum absorption wavelength of the longitudinal mode band. The linear correlation that exists between the aspect ratio of Au NR and the position of the maximum wavelength of the longitudinal mode of the SPR makes quantitative analysis possible. Efforts have been primarily directed towards the following: (a) Development of an immobilization procedure for Au NRs to produce Hg (0) detection substrates; (b) Analytical figures of merit for Hg (0) detection with various types of Au NRs when in solution and immobilized on the detection substrate under static conditions; (c) Interference studies with synthetic interference mixtures; (d) Evaluation a flow injection analysis (FIA) system for Au nanoparticles detection; and (e) Development of Hg (II) and Hg (I) reduction techniques and speciation strategy.

Scanning electron microscopy (SEM) and absorption spectroscopy confirm a well-dispersed deposition of NRs on the surface of the solid substrates. Calibration curves with NRs in aqueous solutions were built for six NRs samples with different maximum absorption wavelengths (610, 615, 630, 690, 730 and 740). The 630nm NRs exhibited the greatest slope and were the most sensitive to the concentration of Hg (0). Their limit of detection was approximately 10^{-20} M/NR. The immobilized NRs showed a limit of detection (10^{-19} M/NR) approximately one order of magnitude better than their counterparts suspended in liquid solutions. Exposure of Au NRs substrates to several inorganic ions showed no interference. A one-line and a two-line FIA system were implemented in our lab to monitor Au nanoparticles in water flows. On-going studies are focusing on the following: (a) extending FIA measurements to monitor Au NRs substrates exposed to ORNL contaminated water flows; (b) reducing mercury species via a cyclic voltammetry (CV) speciation procedure; and (c) combining CV with SPR-Au NRs spectroscopy for the detection of mercury species.