

研 究 主 論 文 抄 録

論文題目 : 水素化物発生分析を用いた水中におけるヒ酸、亜ヒ酸の解析
(Investigation of arsenic behavior in aqueous solution by means of hydride generation
from arsenite and arsenate)

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主論文要旨

Worldwide arsenic occurrence to groundwater and its toxic effect on human health is a serious environmental problem. It is released into the environment from a variety of natural and anthropogenic sources. Chemically, there are two types of arsenic compounds: inorganic and organic. Inorganic arsenic is more toxic than organic arsenic. Arsenite is considered 60 times more toxic than arsenate and binds to sulphhydryl (-SH) groups which impairs function of many proteins. Chronic ingestion of inorganic arsenic causes cancer of the skin, bladder, and lung, as well as neurological and cardiovascular problems. The most severely groundwater contaminated countries including Bangladesh, West Bengal (India), Taiwan, Vietnam, Cambodia. It has become a challenge to researchers, reasons of arsenic leaching to groundwater and simpler removal technique obtaining safe drinking water. In the present research, I have investigated the behavior of hydride generation from arsenite and arsenate applied to monitoring arsenic removal, to investigate arsenic leaching from sediments, and development field instrument based on liquid-reagent-free hydride generation chemiluminescence detection for water analysis.

Arsenic removal

To obtain safe drinking water removal of arsenic is very important. Metal base adsorbents, including aluminum, iron, titanium, zirconium, and manganese, have been extensively studied to remove arsenic from water. Many oxidants like ozone, chlorine, hydrogen peroxide use to oxidize As(III) to As(V) and finally arsenic is removed by

adsorption process. To monitoring arsenic concentration of water treatment technologies many laboratory base analytical methods used to measure total arsenic. Speciation measurement is very important because arsenite is much more toxic than arsenate. An automated arsenic measurement system developed based on complete vaporization of arsenite and arsenate at pH 0 and vaporization of arsenite at pH 7 by a sequential procedure and collection/preconcentration of the vaporized arsine, which was subsequently measured by a flow analysis. Interference from the heavy metals was less because arsine was generated more effectively by mild and long vaporization. The method was applied to monitoring As(III) and As(V) concentrations for arsenic removal in batch wise. Arsenic removal was effective by coprecipitation when hypochlorite (oxidant) was added at a tap water to oxidize As(III) to As(V). A small amount of steel wool is very effective in arsenic removal, adsorbing As quantitatively within 2 h.

Leaching of arsenic from sediments

Leaching of arsenic poses a potential risk to groundwater quality. Various industrial and mining wastes remained left in the nearby area after manufacturing which contained extremely high concentration of arsenic from where soil is polluted, possible mobilization and subsequent leaching into groundwater or surface water or enter the human food chain through various chemical and biological reactions. Sequential hydride generation flow analysis (SHGFA) was applied to water analysis and leaching investigation from the contaminated sediments near an arsenic mine river. The SHGFA system showed the excellent performance for leaching behavior of arsenic analysis by discriminating As(III) and As(V). It was observed that rate of As(III) leaching was faster than As(V) while As(V) leached more in amount compared to As(III).

Arsenate is a phosphate analog and both have the similar electron configuration and form triprotic acids with similar dissociation constants. Phosphate plays an important role to groundwater arsenic contamination leaching from soil/sediments. Application of phosphate fertilizer in agricultural activities to soil surfaces, released arsenic ions sorbed to aquifer minerals by competitive ion exchange with phosphate ion that migrates into aquifer. Simultaneously monitoring arsenite and arsenate by SHGFA applied to investigate arsenic leaching from contaminated sediment by the effect of phosphate ion. Leaching of arsenate was significantly evaluated by the phosphate ion, as phosphate ion has more charge density than arsenate resulting replaced from sediment minerals surface.

Field Instrument Development

A manually hydride generation chemiluminescence instrument developed for

on-site arsenic measurement in the aqueous sample. A hydride generation method based on liquid-reagent-free by avoiding concentrated inorganic acid because transportation and handling of inorganic acids for field measurements is difficult and dangerous for the users without any formal chemistry training. In addition, in the arsenic affected areas it can be difficult to obtain pure water for reagents preparation. The optimized system was evaluated for analysis of natural water samples, and obtained data agreed well with those from ICP-MS and SHGFA. The sample throughput of 60 times h^{-1} and the limit of detection was $0.4 \mu\text{g L}^{-1}$. The measurable arsenic concentration was up to 1mg L^{-1} for 2 mL samples. The instrument is small and light with low power consumption, and could measure $\mu\text{g L}^{-1}$ concentrations of arsenic in a short time.