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| 氏名 | KRITSANA JITMANEE |
| 授与した学位 | 博士 |
| 専攻分野の名称 | 学術 |
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| 学位授与の要件 | 自然科学研究科物質分子科学専攻 (学位規則第4条第1項該当) |
| 学位論文の題目 | Speciation of Toxic Oxoacids by Inductively-Coupled-Plasma Spectrometry Coupled with On-Line Solid-Phase Collection/Flow-Based Method (オンライン固相捕集/フロー法と結合した誘導結合プラズマスペクトロメトリーによる毒性オキソ酸のスペシエーション) |
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学位論文内容の要旨

The speciation analysis of toxic elements, such as oxoacids and heavy metals, is currently of interest and becoming an important research area. Information on the behavior of individual species present in the specimen (environmental, biological) may assess the availability, mobility and toxicity of particular elements. In this study, speciation of oxoacids was studied. The currently interesting species such as Cr(III)/ Cr(VI), As(III)/ As(V) were targeted due to their toxicity and their mobility in the environment. The on-line collection/concentration system with solid phase adsorbent was proposed for the separation and concentration of target species prior to their detection. A bead injection with flow-based colorimetric system was proposed for the speciation of Fe(II) and Fe(III). The chelating resin was used for the collection of Fe species. Fe(III) can be determined from the difference between total Fe content and Fe(II) previously determined. The renewable process of the adsorbent in the detection cell takes advantages over the elution of strongly adsorbed Fe chelate. The other type of solid phase adsorbent was proposed, namely membrane-type extraction media. This type of adsorbent allows the target analytes to be collected in a short zone of extraction media, which results in higher detection sensitivity. The filter-type anion exchange resin was examined for the collection/concentration of oxoacids by using miniaturized collection/concentration apparatus prior to the elution and detection by ICP-MS. Alternatively, the bead-type of anion exchange resin was examined with an automatic on-line collection/concentration system coupled with ICP-AES for the determination of oxoacids.

Interesting and novel results were obtained from the speciation of Cr(III) and Cr(VI). The on-line collection/concentration strategy for ICP-AES measurement with both a cationic and an anionic exchange resin (filter-type) was demonstrated. The selective retention of Cr(III) and Cr(VI) based on their opposite charge and sequentially on-line elution of the collected species resulted in the simultaneous collection and determination of Cr(VI) and Cr(III). The concept was extended to the speciation of As(III) and As(V). The bead-type anion exchange resin was utilized for the collection of anionic species of arsenic. Arsenate could be collected on this adsorbent, while arsenite required on-line oxidation to arsenate prior to be collected. Two kinds of oxidizing agents were examined for the effective oxidation and collection of arsenite. Arsenite and arsenate were successfully determined in freshwater samples. The concept of the utilization of miniature collection/concentration system for the speciation of toxic elements was clarified and its usefulness in chemical analysis demonstrated.

論文審査結果の要旨

In this study, the author emphasizes the development of novel analytical methodology for the speciation analysis of toxic oxoacids, as well as heavy metals. Generally, the speciation analysis of toxic elements has often employed some chromatographic methods coupled with element-selective detection systems. Concentrations of target species, however, are usually very low, and therefore any preconcentration of the target species is required before analysis, because it is difficult to perform the collection/concentration and the separation of the target species in chromatographic methods. Therefore, in the speciation analyses for oxoacids at low concentration levels, such chromatographic separation techniques are not suitable for real samples.

In this study, the author developed interesting on-line collection/concentration strategies for the speciation of toxic elements. The solid-phase adsorbents were examined for the selective collection of the target species. The transparent chelating resin (bead type) was packed in the detection flow cell, where analytes were collected; Fe(II) and Fe(III) were demonstrated for examining the usefulness of this technique. The bead injection technique with an LED-based detection device was applied for this purpose. The system developed in this study was very compact and can be used for on-site analysis.

Two kinds of disk-type solid-phase ion exchange resins, a cationic and an anionic exchanger were utilized for simple collection/concentration and speciation of Cr(III) and Cr(VI). The disk-type solid phase provides high concentration factors along with its compactness. Also, interesting results were shown in the speciation of As(III) and As(V). An anion exchange resin (bead type) packed in a PTFE tubing could act as a collection/concentration device for the anionic arsenate species; the nonionic species, As(III) as H_3AsO_3 , could be oxidized rapidly and effectively with potassium permanganate; then the ionized species of As(V) was collected on the solid phase.

The sensitive and selective methods based on the on-line solid-phase collection/concentration resulted in an interesting strategy for the speciation of toxic oxoacid-forming elements.

In view of original contents and creative results obtained in this study, the committee evaluated this dissertation as PhD degree's worth of research.