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Application of micro-PIXE to quantitative analysis of heavy elements sorbed on minerals

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Abstract

Heavy element-sorbing montmorillonite and apatite were analyzed by micro-PIXE to examine its applicability to quantitative analysis of the heavy elements sorbed on these minerals. It was found that the sorbed heavy elements can be evaluated quantitatively using the linear correlation between the following two factors: (a) the concentration of the sorbed element in the mineral and (b) the ratio of the net peak area of characteristic X-ray from the sorbed element and to that from the matrix cation of the mineral. Mineralogical features of the tested minerals including heterogeneity of elemental distribution in the depth direction of the mineral hardly affected the results.

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1. Introduction

Intense efforts have been paid to clarify sorption behavior of heavy elements in soil environment. Because soil is composed of various mineral particles such as clays, other silicates, iron (hydro) oxides, and phosphate minerals, it is

needed to investigate sorption onto individual minerals in a mixed phase of these mineral particles. To investigate such sorption under laboratory conditions, we have used the micro-PIXE analyzing system developed in the TIARA facility, JAERI. The high spatial resolution of the micro-PIXE analyzing system (less than 1 μm in diameter) and the use of a high-energy proton beam (2.6 MeV) [1] allow us to obtain highly sensitive two-dimensional elemental mapping of the sorbed heavy elements and host minerals.

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Generally, studies on heavy element sorption under laboratory conditions use mineral particles having irregular morphology and size. Distribution of the sorbed element in mineral may vary depending on minerals and chemical conditions of solution. To quantify the elements sorbed on minerals, it is needed to evaluate influence of those mineralogical features on detection of characteristic X-rays. However, to the authors' knowledge, no papers have been published on this subject. Related studies on quantification by micro-PIXE have been limited to analysis of thin specimens of natural rock [2,3]. In this paper, applicability of micro-PIXE analysis to quantitative evaluation of the heavy elements sorbed on minerals was examined using standard materials, which are heavy element-sorbing minerals of practically uniform shape and size. This paper tested Cd, Pb and Eu as heavy elements and montmorillonite and apatite as minerals. Cd and Pb are representative toxic elements and Eu is used as simulant of trivalent actinides. Montmorillonite is a representative swelling clay mineral commonly found in soil [4]. This clay has a high capacity to accommodate cations by cation exchange. Apatite is a representative phosphate mineral. Apatite is not abundant in soil but has attracted intense attentions due to high capability to immobilize heavy elements [5,6].

2. Experimental

2.1. Materials

Natural montmorillonite powder, $(\text{Na}, \text{K}, \text{Mg}_{0.5}, \text{Ca}_{0.5})_{0.49}(\text{Si}_{3.85}\text{Al}_{0.15})(\text{Al}_{1.57}\text{Fe}_{0.09}^{3+}\text{Mg}_{0.34})\text{O}_{10}(\text{OH})_2$ [7], purchased from Kunimine Industries Co., Ltd., was used. Montmorillonite has small, thin plate-like crystals of 1 μm or less in size and about 1 nm in thickness [4]. Before use, the montmorillonite was purified by water elutriation to remove minor contaminant minerals and the subsequent conversion to homoionic Na^+ -type form by the procedure described elsewhere [8]. Two kinds of synthetic apatite, $\text{Ca}_5(\text{PO}_4)_3\text{OH}$, were used. One apatite purchased from Wako Pure Chemical Industries, Ltd., is fine powder of indefinite-shaped single crystals of about 0.1 μm or less

in size. The other one purchased from Bio-Rad Laboratories, Inc. is a sphere particle of about 20 μm in diameter. The latter has many pores of 60–80 nm in diameter.

2.2. Preparation of standard materials

Heavy element solutions were prepared by dissolving CdCl_2 , PbCl_2 and $\text{Eu}(\text{NO}_3)_3$ in 10 mM NaClO_4 aqueous solution. Each of the solutions contained only one kind of these heavy elements up to 10 mM (Pb solutions for montmorillonite contained Pb up to 20 mM). For most of the sample preparation, the starting pH of 10 cm^3 of the heavy element solution in a centrifuge tube was adjusted to 3.0 using dilute NaOH and HCl solutions to allow chemical species of those heavy elements to be cations (Cd^{2+} , Pb^{2+} , Eu^{3+}) [9]. For a part of the sample preparation using Eu, the starting pH of the Eu solution in a centrifuge tube was adjusted to 9.0 and then left for 30 min to allow Eu to form hydroxides. A 0.1 g of a mineral was then introduced to the centrifuge tube. The tube was sealed and shaken gently at 25 °C. After 24 h, solid phase was separated from liquid phase by centrifuge. The liquid phase was further filtrated using a membrane filter with a pore size of 0.2 μm . The solid phase was washed three times by ethanol.

2.3. Analysis methods

Concentrations of elements in liquid phase were determined by ICP-AES and ICP-MS. The concentrations of the sorbed heavy elements in mineral, C_m , were determined by $(C_0 - C)V/W$, where C_0 and C are respectively the initial and final concentrations of heavy element in solution, V is the volume of solution and W is the weight of mineral. For W , the initial weight of mineral was used though the true value of W would slightly change by the sorption of heavy elements.

For micro-PIXE analysis, the heavy element-sorbing minerals were spread on a graphite plate using water and dried in air. For montmorillonite, this method produced a thin film-like sample of a large area (about 1–2 mm in diameter) where crystals of montmorillonite were oriented and piled on top of the other crystals in the same basal plane.

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