

New preparation method of silica-gel activator for lead isotope measurement by hydrolysis of tetraethylorthosilicate (TEOS)

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The lead isotope ratio is one of the important forms of geochemical information for earth science. In the same manner as are Sr and Nd isotope ratios, it is powerful and fundamental tool for revealing dynamic processes such as material circulation during the earth's history. Isotope analyses using the thermal ionization mass spectrometer (TIMS) are performed at many laboratories. However, the analytical process required for lead isotope measurement is much more complicated relative to those of Sr and Nd isotope measurements.

The distinct difficulty in lead isotope measurement comes from the inability to use an internal standard for the correction of mass fractionation (normalization). This difficulty can be solved to a certain extent by determining a mass fractionation factor by analyzing a lead standard with a well-known isotopic composition (e.g. NIST SRM 981) and applying this same correction factor to all unknowns. However, another distinct difficulty also exists in the preparation of the silica-gel activator, which is usually used in order to enhance the ion beam and to limit instrumental mass fractionation. Silicon tetrachloride has usually been used for the preparation of the silica-gel activator thus far. Besides silicon tetrachloride, silicic acid colloidal solution has also recently been used. However, silicon tetrachloride has intense reactivity, making its handling very difficult. Moreover, it is difficult to make the each silica-gel activator with consistently high efficiency. On the other hand, the silicic acid colloidal solution is often difficult to obtain because of its short supply. Therefore, preparation of a silica-gel activator is problematic even at present. In order to solve these problems in the preparation of a silica-gel activator, we have developed a new preparation method using tetraethylorthosilicate (TEOS), a technique that mitigates the difficulty resulting from intense reactivity and avoids the problem of the reagent scarcity.

TEOS is alkoxide known which has been in existence for many years, and which is synthesized silicon tetrachloride with ethyl alcohol. This reagent is commonly used in various fields such as ceramics, chromatographic absorbents, etc., in order to produce silica particles. Thus, this reagent is easily obtainable. The most widely used methods to obtain silica particles are the so-called sol-gel processes based on the hydrolysis and polycondensation of alkoxy silanes. The hydrolysis of TEOS advances very slowly unlike that of silicon tetrachloride. Therefore, the quality of the silica-gel activator can be easily controlled. We succeeded in producing the optimal silica-gel activator for lead isotope measurement utilizing the TIMS. By using this new silica-gel activator, it is possible to consistently obtain a strong and stable ion beam. For example, when 200ng of lead samples (NIST SRM 981) are measured, a beam intensity (208Pb) of 4000mV or more can be obtained in almost all measurements. In addition, the beam intensity decayed less than 40% over the course of about 30 minutes.

The improvement in the reproducibility of the isotope ratio is very important for improving the reliability of analysis. Repetition measurement of lead reference material (NIST SRM 981) using the new silica-gel activator indicates an improvement in reproducibility. The measurement using new silica-gel activator was able to decrease the error no less than 37% in comparison with the measurement using old silica-gel activator made from silicon tetrachloride. Although there is still room for improvement, we can presently measure 25ng or more of lead samples with about same reproducibility. By adopting the new preparation method for the silica-gel activator, we can more simply and accurately measure lead isotope ratios.