10–1 Development of a New Method for Age Determination

— Reduction of Analytical Time Using the Secular Equilibrium State in a Sample —

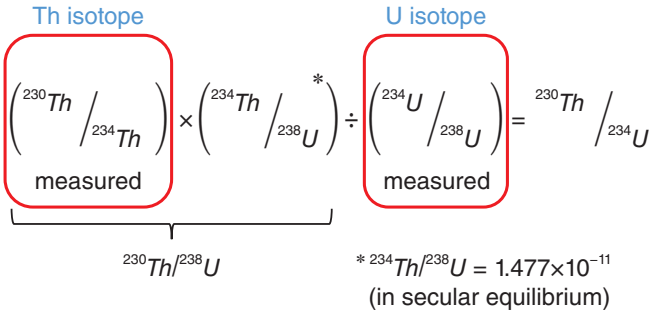

Fig.10-2 Equation for age determination without isotopic certified reference materials (spike solution)
The isotopic ratio of $^{234}Th/^{238}U$ in secular equilibrium can be calculated from the half lives of these isotopes. Using the measured isotopic ratios of $^{230}Th/^{234}Th$ and $^{234}U/^{238}U$ and this equation, the $^{230}Th/^{234}U$ isotopic ratio for age determination can be calculated.

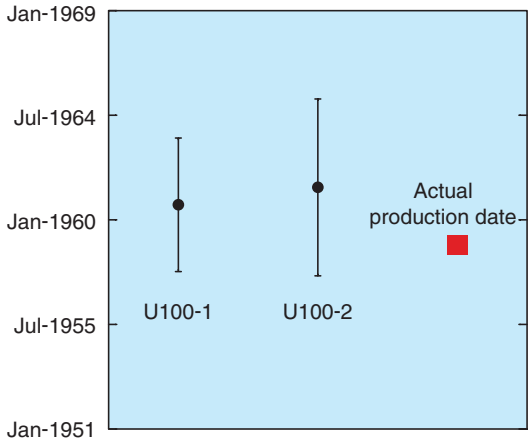

Fig.10-3 Age determination results for the uranium certified reference material (U100)
Applying the proposed method for age determination to the uranium certified reference material (U100), model-production dates that agree with the actual production date are obtained.

Each state is considered to have a responsibility to reinforce nuclear security to combat terrorism using nuclear and radioactive materials. Nuclear forensics is a technical measure for analyzing and assaying the origin, history, route of transportation, and objective of nuclear and radioactive materials. Each state is developing techniques and a framework for nuclear forensics as an important component of nuclear security. In particular, age determination is used to demonstrate the production date of nuclear material and is therefore believed to be essential information in nuclear forensics.

The principle of uranium (U) age determination can be described as follows. In U production processes, other elements are eliminated and the produced material does not contain its daughter nuclide of thorium (Th). As time progresses, Th is produced again at a constant rate in purified U materials. Consequently, the production date can be estimated to measure the ratio of U and Th in materials. To measure the amount of U and Th in the sample, the isotope-dilution mass-spectrometry (ID-MS) method is widely used; this method requires the addition of certified reference materials (spike solution). Accurate measurement of U and Th isotopes using the ID-MS method requires precise weighing of a sample and spike solution, as well as strictly controlled preparation of the spike solution at its concentration. In this study, to establish a concise method for U age determination, a new method is developed using the $^{230}Th/^{234}Th$ and $^{234}U/^{238}U$ ratios in the sample and the calculated $^{234}Th/^{238}U$ ratio in secular equilibrium (Fig.10-2). According to this new method, U and Th in the U sample solution are separated by two steps of anion exchange. The isotope ratios of U and Th are then determined by a thermal-ionization mass spectrometry. The ID-MS method requires three days for performing the analytical procedure; the new method reduced this time to six hours, thereby completing the process rapidly. Applying the new method for age determination to the uranium certified reference material (U100), we obtained model-production dates that agree with the actual production date (Fig.10-3). Joint research between JAEA and the European Commission, Joint Research Center is underway for the comparison between the ID-MS method and our new method, which is applied for radioactive measurements.

Reference

Okubo, A. et al., Uranium Age-Dating Using In-Situ Isotope Ratios by Thermal Ionization Mass Spectrometry for Nuclear Forensics, Journal of Radioanalytical and Nuclear Chemistry, vol.314, issue 1, 2017, p.231–234.