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Uranium Age Dating by Gamma Spectrometry

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A new method for uranium age dating was developed using gamma-spectrometry based on the daughter/parent 214Bi/234U by direct measurement. The daughter/parent ratio as a function of decay time is widely used for determining the age of radioactive samples. In the case of uranium, age dating is somewhat difficult because the relevant isotopes (234U, 235U, 238U) have very long half-lives, so only small amounts of daughter nuclides grow in. In contrast, the age of nuclear materials is, at most, merely a few decades, which is very short compared to the long half-lives of the parent isotopes. Therefore, one would expect that the daughter nuclides could only be quantified after destructive chemical separation, followed by mass-spectrometric or alpha-spectrometric techniques. However, it has been demonstrated that the daughter/parent activity ratio 214Bi/234U can be obtained by directly measuring the count rates of the relevant gamma peaks of 214Bi and 234U by low-background gamma spectrometry. The method is non-destructive and does not require the use of reference materials of known ages. The least enriched uranium sample dated by HRGS was a 5% enriched oxide material, the age of which was determined as 54 ± 7 yr. The youngest sample was a 6.7 ± 0.7 yr old metallic U of 90.8% enrichment.

234U decays through 230Th to 226Ra, which in turn decays to 214Bi through three short-lived nuclides. The time needed for secular equilibrium between 226Ra and 214Bi is about 2 weeks, so it can be assumed that the activities of 226Ra and 214Bi are equal at the time of the measurement. The activity ratio 214Bi/234U at time T after purification/enrichment of the material can be calculated with a good approximation.

The activity ratio 214Bi/234U can be determined in several ways from the gamma spectra of a sample. A reliable method uses a reference material of approximately like age, enrichment, and form, as the investigated sample. Another approach does not require any reference materials but uses the absolute efficiency of the detector determined by "point-like"standard sources. In an alternate approach, a relative efficiency calibration is used to determine the activity ratio 214Bi/234U, without the use of any reference materials. Measurements of HEU metal lump and oxide powder, HEU and LEU reactor fuel rods, LEU pellet and powder were made in the enrichment range from 4.4 to 90%.

For lower 235U abundances, the amount of 234U (and therefore of 214Bi) is lower as well, so the corresponding activity is more difficult to measure and the uncertainty caused by the variation of the natural background becomes greater. In addition, a Compton background caused by the peaks of 238U daughters is also present in the spectrum, disturbing the evaluation of the 214Bi peaks. A lower limit on the 235U abundance of the material exists that allows the age to be determined by gamma-spectrometry, depending on the amount and the age of the material, detector efficiency and background level. For example, the 609 keV peak of 214Bi (and the peaks of 238U) are measured by a 150 cm3 coaxial Ge detector in a low-background iron chamber with a wall thickness of 20 cm. Low energy peaks of 234U (and 235U) are recorded by applying planar Ge detectors under normal laboratory conditions.

The "difficult"samples are the same as in the case of mass spectrometry: low-enriched and/or "young"uranium. According to our estimate, with our current equipment, 10 g of natural uranium can only be dated, if it is more than 35 years old, whereas the lower bound for age determination of 90 % enriched HEU was estimated to be around 2 years. The sensitivity and the range of applicability of the methods may be improved by using e. g. an average size well-type Ge detector, so that the lower bound for the applicability of the method would be approximately 1.5 year for 90% enriched uranium and 20 years for natural uranium, under the present background conditions.

Gamma-spectrometric age dating of uranium is, within the described limits, a reliable tool for determining the age of uranium samples encountered in combating illicit trafficking of nuclear materials and in nuclear

safeguards.

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Author: Prof. LAKOSI, L. (Hungary)

Presenter: Prof. LAKOSI, L. (Hungary)

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