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Feasibility study for the preparation of plutonium reference materials for nuclear age dating

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Plutonium is an antropogenic element which is formed by multiple neutron capture reactions starting with uranium. Beyond this textbook knowledge, the element plutonium only existed in minute traces in (fairly rich) uranium ores. Nowadays, traces of plutonium are more widely spread in the environment due to the numerous atmospheric and underwater nuclear weapons tests in the late 1950's and early 1960's as well as due to the few nuclear accidents. The total amount of plutonium is growing continuously due to the production of plutonium in nuclear reactors. Although the nuclear fuel cycle is subject to strict regulatory control schemes (nuclear safeguards), nuclear material (uranium, plutonium) is occasionally discovered outside this regulatory control system. In these cases nuclear forensic investigations are applied in addition to traditional forensics in order to provide clues on the history of the material and to possibly identify the last legal owner of the material. The isotopic and elemental composition, the physical appearance of the material (dimensions of larger objects; particle form and size of powders) and its "age" are determined.

In order to determine the age of nuclear material, the relationship between amounts of the originating so-called mother isotopes and amounts of daughter isotopes continually being formed by radioactive decay is used. This principle has been applied for the age determination of minerals and rocks by geologists for many years; the isotopic composition of rocks changes with time after their formation, thus the age can be derived from measuring the ratio of mother to daughter nuclide. Similarly, the "age" of nuclear material is defined as the time that has passed since the last chemical separation of the daughter nuclides from the mother nuclides (e.g. in-grown uranium and americium from plutonium). The ratio of mother to daughter nuclides depends on the half-lives of the radio-nuclides involved and the time that has passed since the purification of the mother isotopes. For the determination of the age of a plutonium material different pairs of mother and daughter radio-nuclides can be used: ^{241}Pu/^{241}Am, ^{238}Pu/^{234}U, ^{239}Pu/^{235}U, ^{240}Pu/^{236}U, and possibly ^{242}Pu/^{238}U.

At the moment no uranium or plutonium reference materials certified for "age" are available, although they are needed in order to provide a more solid metrological basis for nuclear forensics and environmental measurements. Addressing these present needs the Institute for Reference Materials and Measurements (EC-JRC-IRMM) is closely cooperating with the Institute for Transuranium Elements (EC-JRC-ITU). As part of the presented feasibility study for the development of plutonium reference materials for age dating the well known and widely distributed isotopic abundance reference materials NBS SRM 946, 947 and 948 (NBL CRM 136, 137 and 138) are investigated. The "ages" of these materials (with known separation dates) of differing ages and isotopic compositions will be verified by measuring amount concentrations of mother (^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu) and daughter (^{234}U, ^{235}U, ^{236}U, ^{238}U and ^{241}Am) isotopes. Proper sample preparation and elemental separation is a prerequisite for accurate isotope ratio measurements. Thermal ionisation mass spectrometry (TIMS) is applied for the measurement of the plutonium and uranium isotope amount concentrations via isotope abundance measurements and isotope dilution mass spectrometric (IDMS) measurements while ^{241}Am is measured by gamma-ray spectrometry.