RIMS, A Suitable Method for Nuclear Forensic Investigations

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Efforts were made in Germany during the Second World War for attaining nuclear energy by a self sustained nuclear chain reaction. In Haigerloch, a little village in the south-west part of Germany, a team of scientists headed by W. Heisenberg tried to reach criticality with a heavy water reactor, the B-8 pile, using 664 metallic uranium cubes suspended from the lid by 76 chains arranged in honey-comb geometry (1.58 t $^{nat.}$U) and immersed in 1.71 m^3 of heavy water. It was reported that, at the pile's mantle, a neutron multiplication of 6.7 was measured when the B-8 was seeded with neutrons from a (Ra-Be)-source. It is known that the fuel was subject to purification prior to introduction in the reactor, with the intention to eliminate thorium from the matrix. It can be assumed that plutonium would have been also removed by this procedure, provided it was present as Pu(IV).

It is not clear whether the scientists were able to reach criticality with this design. This could be determined, however, by analyzing the nuclear fuel material, by determining the amount of ^{239}Pu formed as a result of the elevated neutron flux.

Unfortunately, most of the material used for the experiments in the B-8 pile was lost. However, in the 1960's, several cubes of the above mentioned uranium were found. One of them was sent to the Institute for Transuranium Elements, European Commission (ITU). A comprehensive analysis was performed on this material in order to determine its age, the level of impurities, and its origin by the colleagues from ITU. It was found by the uranium/thorium dating technique that the purification took place around September 1943. This indicates that the material could be from the batch used in the Haigerloch reactor.

The isotopic composition of uranium was determined also in Mainz by ICP-MS and indicated just like at ITU the natural composition of uranium (99.27 % ^{238}U, 0.72 % ^{235}U, and 0.0055 % ^{234}U).

Resonance Ionization Mass Spectrometry (RIMS) is an element and isotope selective method and has a very low detection limit for plutonium (LOD ~ 10^6 atoms). This makes it suitable for analysis of ultra trace amount of plutonium where element and isotopic selectivity is needed.

Therefore, subsamples of the Haigerloch fuel material were analysed by RIMS. For comparison, a sample of uranium ore originating from Joachimsthal and a sample of metallic uranium from the same batch that, however, had not been seeded with neutrons, were investigated. In the first place, the surfaces were etched with 8 M HNO_3 in order to eliminate any possible surface contamination. The samples were then divided into 3 subsamples of ~ 1 g and RIMS filaments were prepared as described in [1]. The content of ^{239}Pu in the etched material was also determined.

Prior to the RIMS analyses, the efficiency and potential cross contamination of the apparatuses was checked in order to eliminate any suspicions on the accuracy of the measurements. A typical analysis result is shown in Fig. 1.

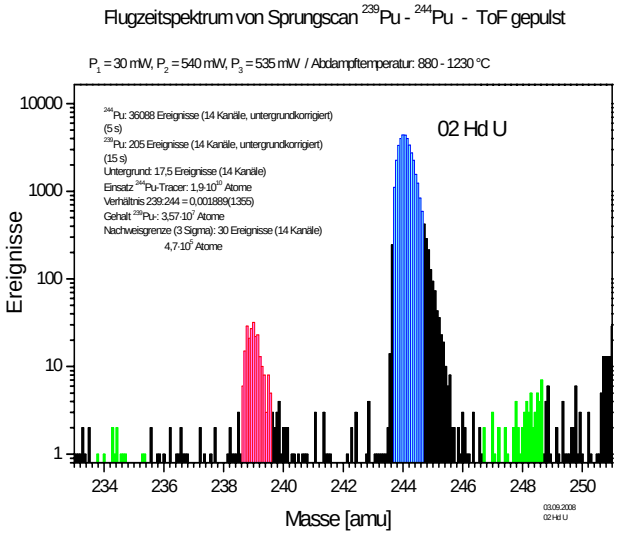

Figure 1. ^{239}Pu analysis by RIMS. 3*10^{10} Atoms ^{244}Pu as tracer

It was found that the amount of ^{239}Pu in uranium was comparable for all three samples examined at a level of ~ 1.5*10^{-14} g ^{239}Pu/ g ^{238}U. Also, the amount of plutonium found in the material etched from the surface was comparable with the value stated above.

An estimation of the amount of plutonium formed as a result of irradiation by cosmic rays and neutrons resulting from the spontaneous fission of uranium as well as a simulation of the level expected after a potential criticality of the reactor were performed at ITU [2].

After comparing all the experimental results obtained for plutonium with the theoretical simulations, one can conclude that the reactor was not critical.

It can also be stated that RIMS is a suitable method for analyses at ultra trace amounts where high accuracy is needed.

References

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