Primordial ^{244}Pu on Earth?

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The actinides Thorium and Uranium are produced during explosive stellar nucleosynthesis, for example a supernova. Such an event had presumably taken place just before the formation of the Earth. With its half-life of 81.2 million years [1] ^{244}Pu could also still be present on Earth in smallest traces and thus would be the third primordial actinide.

With the time-of-flight setup at the MLL tandem accelerator the first search for primordial ^{244}Pu with AMS was possible. Hereby the previous results on this topic by Hoffman *et al.* [2], who detected a concentration of about 2500 atoms ^{244}Pu per gram in the mineral Bastnaesite, could not be confirmed. Bastnaesite was chosen as target for the search for primordial ^{244}Pu, because it is highly enriched in rare earth elements, which are chemical homologues of the actinides.

We derived an upper limit of about 550 atoms ^{244}Pu per gram Bastnaesite. This limit is consistent with calculations of the production of this isotope as a primordial nuclide during a supernova explosion just before the formation of the Solar system 4.566 billion years ago and the assumption of a similar enrichment of Plutonium in this mineral as the homologeous rare earth elements. These computations indicate a concentration of few 100 atoms ^{244}Pu per gram Bastnaesite.

The chemical separation of ^{244}Pu from the matrix after the dissolution of the mineral in aqua regia in portions of 150 grams is performed by use of the liquid-liquid-extractant HDEHP and the anion-exchange-resin Amberlite-IRA-402 (Cl). During the liquid-liquid-extraction from the aqua regia solution Plutonium is separated from the main part of the rare earth elements and is extracted from the HDEHP back into HCl after several washing steps and after transformation into its III-valent state [2,4]. This HCl solution is directly cleaned on the anion exchange resin, because a good separation from Uranium left over from the sample matrix is necessary for clean spectra during the AMS measurement. In this step Uranium is kept on the resin, while III-valent Plutonium stays in the eluent. Further treatment of the Plutonium solution according to conventional anion exchange techniques [5] reduce the sample to few milligrams. After drying on the heating plate the material is mixed with iron powder, redissolved in HCl, again dried and put in a muffle furnace. From this material the AMS samples are fabricated.

The way of Plutonium during the chemical separation was traced by liquid scintillation measurements in aliquots of the processed sample. For this reason 50 kBq of the short-lived α-emitter ^{236}Pu ($T_{1/2}$=2.86 years) were produced by an irradiation of an Uranium target [3]. A precisely determined quantity of usually 80 Bq of this ^{236}Pu in an HNO$_3$ solution was given to each portion of the dissolved mineral initially.

During the AMS measurements this ^{236}Pu in the samples served as reference nuclide, wherefrom the detection efficiency for Plutonium ions could be derived. Yields of 1 detected event per 10^4 ^{236}Pu atoms in the target were achieved for the cleanest samples [6].

No event could be attributed to ^{244}Pu. The range of time-of-flight and residual energy, in which signals of Plutonium were expected, was calculated from the signals of ^{232}Th and is shown in figure 1:
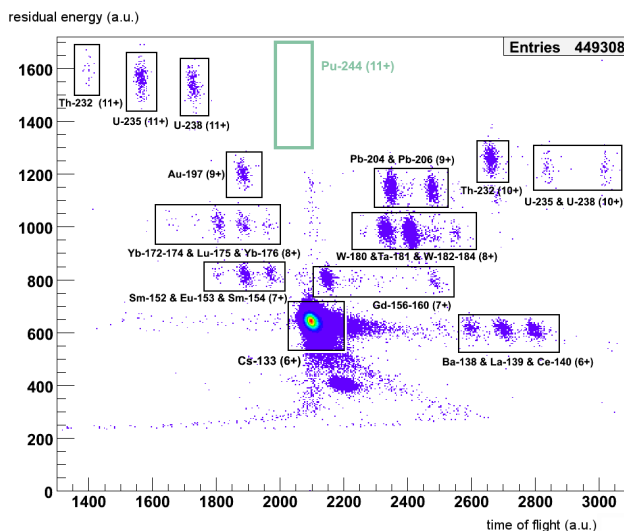

Fig. 1: Spectrum of the residual energy signal against the time-of-flight signal during measurements of the mineral Bastnaesite with settings for ^{244}Pu.

A measurement of a ^{244}Cm sample confirmed these calculated ranges for ^{244}Pu events. Masses and charge states of scattered background events in the various spectra can be identified and originate from the sample matrix (actinides, rare earth elements, Pb) and from the ion source (Cs and Ta).

References

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