Production of a ^{236}Pu Tracer via a (p,3nβ^-)-Reaction on ^{238}U

J. Lachner, A. Alke [a], T. Faestermann, F. Klein [a], G. Korschinek, C. Lierse von Gostomski [a], M. Maiti, M. Poutivtsev, G. Rugel, and A. Türler [a]

[a] Institut für Radiochemie, Technische Universität München, Walther-Meissner-Straße 3, 85748 Garching

The detection of primordial ^{244}Pu ($T_{1/2} = 81.2\,Ma$) on earth might reveal information on the formation of our solar system. This requires accurate monitoring of Pu chemistry of natural samples which can be performed by means of shortlived anthropogenic Pu isotopes.
The main advantages of ^{236}Pu as chemical tracer are its halflife of 2.86 years and the possibility to produce it with rather small contaminations from other Plutonium isotopes, here only the ^{238}Pu ($T_{1/2} = 87.7\,a$).
The chosen reaction was ^{238}U(p,3n)^{236m}Np with succeding β^--decay from ^{236m}Np ($T_{1/2}$=22.5 h) to ^{236}Pu [1]. This decay has the same yield as the competing β^+-decay to ^{236}U.
An irradiation of Uranium with protons at energies in the MeV range not only induces Neptunium isotopes [1] but predominantly fission products. This complicated the handling of the irradiated target and its chemical processing.

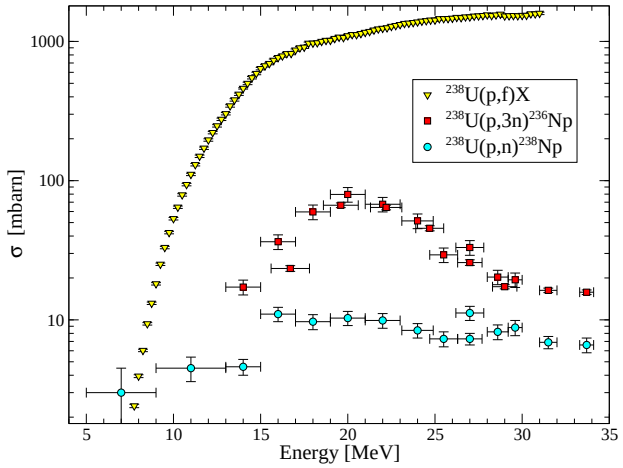

Fig. 1: Cross sections for proton reactions on ^{238}U

The irradiation was made with thick target geometry, therefore the energy was not at about 20 MeV, the maximum of the direct cross section for the production of ^{236m}Np, but at a proton energy of 24 MeV. In a thick target one achieves at this energy the best ratio of ^{236}Pu related to ^{238}Pu.
The target consisted of 0.66 g $^{238}U_3O_8$ - i.e. $1.4{\cdot}10^{21}$ atoms of ^{238}U - in an aluminium holder mounted on copper for thermal conductivity to the beam line.

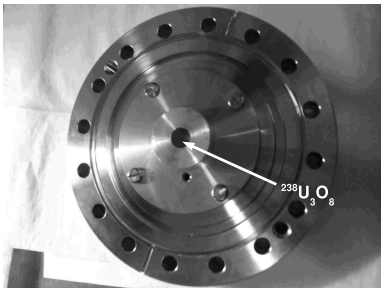

Fig. 2: Sample of $^{238}U_3O_8$ pressed in aluminium

In November 2007 the irradiation with 24 MeV protons took place on the beamline I-40 ° of the MLL Tandem Accelerator. During the 26 hours of irradiation a mean current of 200nA produced an activity of approximately 40 kBq of ^{236}Pu and 520 Bq of ^{238}Pu.
The high cross section for fission in this experiment made it necessary to check the activity of the target with a Germanium γ-detector. Besides the fission fragments - as one example the line of ^{115}Cd - we also measured activation in the beamline (^{55}Co, $T_{1/2} = 17.5\,h$) and the production of ^{238}Np. The γ-lines of ^{236}Np have too low intensities and were therefore not visible.

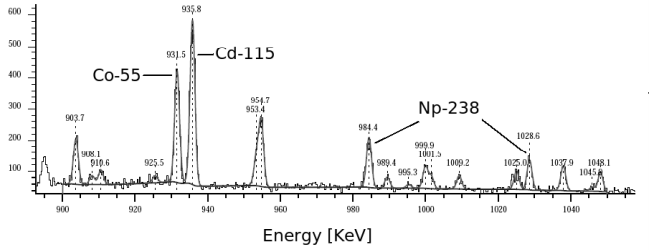

Fig. 3: γ-spectrum 60 hours after the irradiation

The α-detection showed ^{236}Pu with α-energies at 5.72 and 5.77 MeV, ^{238}Pu and ^{238}U and its daughter ^{234}U in the irradiated target, no significant amounts of other α-emitting actinoides were produced.
Further the target was dissolved and the Plutonium was separated from the Uranium and the fission products by the anion-exchange resin Amberlite IRA-402 Cl. The dominant fission products still present after this purification are ^{95}Nb, ^{95}Zr and ^{103}Ru. They have to be separated in an additional chemical step.
Up to this the efficiency for the chemical extraction of the Plutonium from the target was determined as 57±4%.
The ^{236}Pu and ^{238}Pu will be used as a tracer to monitor the efficiency of the chemical extraction of longlived ^{244}Pu from the rare-earth-mineral Bastnaesite. ^{244}Pu should be present in an enriched abundance related to its mean occurence in the earth crust, similar as its chemical homologous Cerium which is enriched by a factor of 10.000 in this mineral. Because of this increased abundance the ^{244}Pu could be detectable with mass spectrometrical methods.

References

[1] S.-J. Heselius *et al.* Radiochimica Acta **91** (2003) 557