## Search for Neutron-deficient Th Isotopes

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For several neutron-deficient isotopes of Thorium  $(^{211,213,217,218}$ Th) the discovery of naturally occurring long-lived high-spin K-isomers was announced by Marinov et al. [1] after detecting several counts at the respective masses in a chemical solution of Th by use of an ICP-MS. These findings were tested using the time-of-flight setup for accelerator mass spectrometry (AMS) at the Maier-Leibnitz-Laboratory. Although with this method we reach a higher sensitivity, we cannot confirm the discoveries of new primordial Th isotopes and thus provide upper limits [2].

The natural existence of  $^{211,213,217,218}$ Th would require several new physical processes and phenomena: As these isotopes cannot be produced during stellar burning, s- or rprocess, because of their position on the neutron-deficient side of stability among short-lived  $\alpha$ -emitters, a new and previously unidentified production mechanism would be necessary, e.g. heavy ion reactions. Even if a comparable yield for the mentioned isotopes is assumed in this unknown nucleosynthesis process as for  $^{232}$ Th in the rprocess, still half lives above  $10^8$  years would be required to explain their existence today.

The ground states of these isotopes are all  $\alpha$  emitters and have half-lives below seconds. According to [1] isomeric states in the mentioned nuclides shall gain additional stability against electromagnetic decay from K-hindrance, but also an angular momentum hindrance is necessary to slow down the  $\alpha$ -decay by at least 16 orders of magnitude to realize the long half lives.

Besides, prolate deformations of the ground states are essential for the existence of K isomers, whereas for <sup>211,213,217,218</sup>Th oblate or spherical shapes are calculated.

ThO<sub>2</sub> from Merck was mixed with clean Carbon powder and was pressed in a graphite cathode for the sputter ion source, from which ThC<sup>-</sup>-molecules were extracted. Beam currents of several nA were achieved for <sup>232</sup>ThC<sup>-</sup>molecules on the low-energy-side. The terminal voltage of the MP tandem accelerator was set at voltages around 11 MV, so in total the Th<sup>10+</sup> ions typically had an energy of around 120 MeV. To adjust the beam transport system we used <sup>232</sup>Th<sup>10+</sup> from <sup>232</sup>ThC<sup>-</sup> and <sup>209</sup>Bi<sup>10+</sup> ions from <sup>209</sup>BiO<sup>-</sup>. The transmission was additionally checked by measuring <sup>228</sup>Th in the same sample originating from the natural <sup>232</sup>Th decay line. Because we used more than 15 years old ThO<sub>2</sub> for the sample preparation, the radioactive equilibrium between <sup>228</sup>Th and <sup>232</sup>Th was assured.

The ions are identified by a time-of-flight spectrometer, where the start signal is delivered by a microchannel plate detector. An ionization chamber determines the energy loss of the ions and a silicon surface barrier counter measures the residual energy and simultaneously acts as the stop detector. In alternation with the search for the rare isotopes the macroscopic beam of  $^{232}$ Th, attenuated to count rates below 1 kHz by transmission grids, was sent to the detector in order to normalize the flux of the Th isotopes. Each isotope was measured for several hours.

None of the four neutron-deficient Th isotopes could be detected with an abundance of 1 to  $10 \cdot 10^{-11}$  as suggested in [1]. No events appeared in the calculated range of time-of-flight and energy signal during the search for the nuclides <sup>211</sup>Th and <sup>217</sup>Th. A single event was measured for <sup>213</sup>Th and <sup>218</sup>Th respectively, which would correspond to an abundance of these isotopes on the level of several  $10^{-13}$ . But these signals can be attributed to the outspread background. Generally, background events are detected at a wide range of different time-of-flights and energies due to ions of  $^{232}$ Th which by scattering processes during the beam transport may obtain the time-of-flight and energy assigned to the rare isotope in question. With the installation of two more Wien-filters after the 90° analyzing magnet the occurence of such events in the spectra should be reduced.



Fig. 1: Spectrum of the residual energy signal against the time-of-flight energy signal during the search for  $^{217}$ Th in ThO<sub>2</sub>.

Measurements lasted for several hours for each isotope and were stopped as soon as an abundance limit of the neutron-deficient Th isotopes in our sample of about an order of magnitude below the values from [1] had been reached. Limitations for our sensitivity are therefore not dominated by the background events from scattered <sup>232</sup>Th ions, but the upper limits could have been reduced further by longer run times.

The final results are given in the following table:

Isotope	events	upper limit for $^{A}Th/^{232}Th$
$^{211}$ Th	0	$9.6 \cdot 10^{-13}$
$^{213}$ Th	1	$1.2 \cdot 10^{-12}$
$^{217}$ Th	0	$6.6 \cdot 10^{-13}$
$^{218}\mathrm{Th}$	1	$2.4 \cdot 10^{-12}$

Table 1: AMS-results for the abundance of  $^{211,213,217,218}$ Th, upper limits given at the 68% confidence level.

## References

[1] A. Marinov et al., Phys. Rev. C 76 (2007) 021303(R)

[2] J. Lachner et al., Phys. Rev. C 78 (2008) 064313