Search for A=60 Fragments from Neutron Induced Fission

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The mass yields of fission fragments from nuclei with A$\approx$ 240 have the well known asymmetric distribution. For neutron induced fission of ^{235}U yields are known down to Y$\approx 10^{-8}$ per fission for fragments with A=69 [1] (see fig. 1). Very light nuclei have also been observed, when a nucleus fissions into three fragments, ^{21}O being the heaviest ternary particle for ^{235}U(n$_{th}$,f) with a yield of 10^{-8} [2]. We investigated the possibility to detect even smaller fission yields with AMS. The longlived ^{60}Fe seemed well suited for this purpose, because it should collect all A=60 fragments after β decay, its detection threshold is very low (^{60}Fe/Fe $\leq 10^{-15}$, see also [3]), and because the possibility to be produced by other reactions than fission seemed small enough.

In order to get samples from irradiated uranium, where the radioactivities had time to cool down, we approached the former reprocessing plant Wiederaufbereitungsanlage Karlsruhe (WAK). This plant was in operation until 1990 and stores - dissolved in nitric acid - the waste from reprocessing spent fuel elements of nuclear power reactors. This solution still has 1.4×10^{11} Bq/l α-activity and 1.1×10^{13} Bq/l β-activity, but in the hot labs of the WAK one can handle such activities.

In collaboration with the Institut für Radiochemie der TU München a separation scheme was developed with a high efficiency for iron and a good suppression of the main sources of radioactivity. The first step, performed at the WAK, was a solid phase extraction using the organic agent CMPO. The second purification then served to further reduce the activity of the samples and also to reduce the content of nickel, the disturbing isobar in AMS. With this procedure the main β activities were reduced by a factor of 2×10^7, the α emitters by 2×10^6. The 20 ml of waste concentrate, which were used for the chemical separation, contained 100 mg of iron. 40 mg were recovered after the separation. Part of this material in the form of FeO was analyzed for ^{60}Fe with AMS at the gas filled magnet GAMS. The result was a surprisingly high concentration of ^{60}Fe/Fe $= (2.0 \pm 0.5) \times 10^{-10}$.

Since the only disturbing production of ^{60}Fe in a reactor could be via successive double neutron capture on ^{58}Fe we have also measured the unknown ^{59}Fe(n,γ) cross section. We irradiated 1.07 mg of iron enriched to 58.4% in ^{58}Fe for 24 hours in the FRM. The neutron flux of 1.9×10^{13} cm^{-2}s^{-1} was determined from the activity of ^{59}Fe. With the measured concentration of ^{60}Fe/^{58}Fe $= (1.0 \pm 0.2) \times 10^{-11}$ we get a capture cross section $\sigma_{59} = (7.0 \pm 1.4)$ barn for neutrons with a reactor spectrum.

The amount of iron irradiated by neutrons could be estimated from: a) the activity of ^{55}Fe measured in the WAK sample, b) the knowledge that its container was filled during the years 1987 to 1990, and the average values of c) neutron flux $\Phi = 2.3 \times 10^{13}$ cm^{-2}s^{-1}, d) irradiation time of 2.5 years and e) cooling down time of 4 years after removal of the fuel rods. These latter values were adjusted to reproduce the measured activities of a number of fission fragments and neutron capture products with a wide range of half lives.

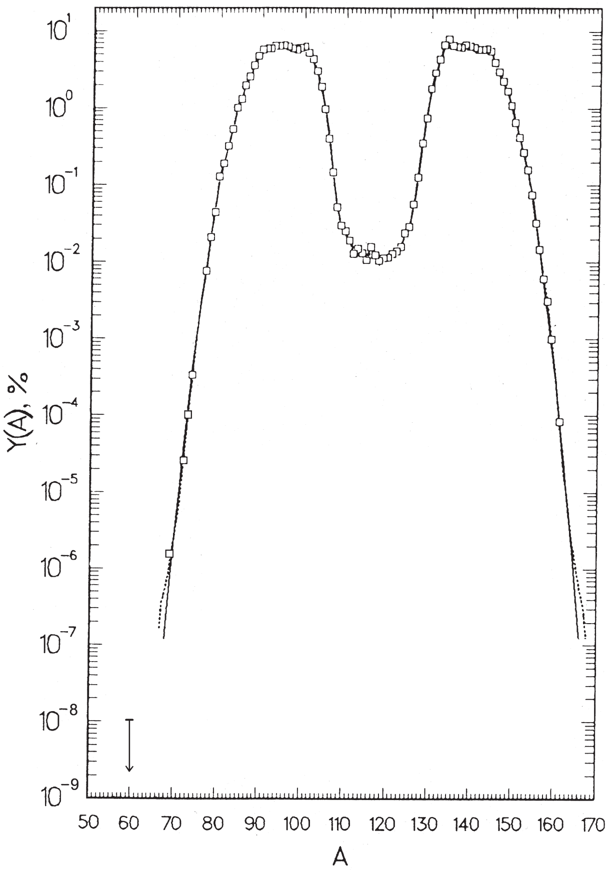

Fig. 1: Mass distribution (in %) [4] of fission fragments for ^{235}U(n,f). The data point at A=60 is our upper limit.

With these values we expect an ^{60}Fe concentration in our sample from double capture of ^{60}Fe/Fe $\approx 0.6 \times 10^{-10}$ with a rather large uncertainty. We therefore cannot rule out that all ^{60}Fe measured in the WAK sample was produced by n-capture. But we still can use the measured concentration to deduce an upper limit for the A=60 yield in neutron induced fission of $Y_{60} \leq 1 \times 10^{-10}$, two orders of magnitude more sensitive than the value for A=69 (see fig. 1). By irradiating ^{235}U with a small iron content in the FRM we will be able, after a cooling down period of at least a year, to measure this yield with a sensitivity being four orders of magnitude higher.

References

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