

Cross sections for the formation of long-lived radionuclides ^{10}Be , ^{26}Al and ^{36}Cl in 14.6 MeV neutron induced reactions determined via accelerator mass spectrometry (AMS)

By F. Sudbrock¹, U. Herpers^{1,*}, S. M. Qaim^{1,2}, J. Csikai³, P. W. Kubik⁴, H.-A. Synal⁵ and M. Suter⁵

¹ Abteilung Nuklearchemie am Institut für Biochemie, Universität zu Köln, D-50674 Köln, Germany

² Institut für Nuklearchemie, Forschungszentrum Jülich GmbH, D-52425 Jülich, Germany

³ Institute of Experimental Physics, Debrecen University, H-4001 Debrecen, Hungary

⁴ Paul Scherrer Institut, CH-5232 Villigen, Switzerland

⁵ Institut für Teilchenphysik, ETH Hönggerberg, CH-8093 Zürich, Switzerland

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Summary. Cross sections for the neutron induced production of the long-lived radionuclides ^{10}Be (from carbon), ^{26}Al (from aluminium) and ^{36}Cl (from potassium) were determined, and upper limits for the formation of ^{10}Be from nitrogen and oxygen were estimated. Target samples were irradiated with 14.6 ± 0.2 MeV neutrons in Debrecen, Hungary. After radiochemical separations and preparation of suitable samples at Köln, Germany, the above mentioned long-lived radionuclides were measured via ultrasensitive accelerator mass spectrometry (AMS) in Zürich, Switzerland. Our cross section value for the $^{27}\text{Al}(n,2n)^{26}\text{Al}$ reaction is within the range of the reported AMS data. For the other two reactions, viz. $^{13}\text{C}(n,\alpha)^{10}\text{Be}$ and $^{39}\text{K}(n,\alpha)^{36}\text{Cl}$, our work describes the first results obtained via measurement of the product nuclides. The experimental results for all the three investigated reactions fall within the range of values obtained from semi-empirical systematics described in the literature.

Introduction

Long-lived activation products represent a basic problem in accelerator development, fusion reactor technology (FRT) and the presently debated nuclear waste transmutation concepts, not to mention their general applicability in testing nuclear models. In the FRT, considerable activation may be caused by neutrons in the energy region around 14 MeV [cf. 1–3]. Under the auspices of an IAEA Coordinated Research Project (CRP) several long-lived products have been recently characterized [4]. There are, however, a few other products for which the formation cross section data are either sparse, contradictory or not available at all. Simple counting techniques have turned out to be insufficient for many long-lived nuclides, e.g. ^{10}Be , ^{36}Cl etc., especially when they are produced only in small amounts. For those nuclides the ultrasensitive accelerator mass spectrometry

(AMS) now provides a versatile tool to determine the formation cross sections. It allows to measure isotopic ratios down to 10^{-14} (N^*/N : unstable isotope/stable isotope) for at least six radionuclides routinely [cf. 5]. We studied the formation of ^{10}Be ($T_{1/2} = 1.5 \times 10^6$ a) in 14 MeV neutron induced reactions on the target elements carbon, nitrogen and oxygen. Furthermore, the formation of ^{26}Al ($T_{1/2} = 7.16 \times 10^5$ a) from aluminium and of ^{36}Cl ($T_{1/2} = 3.0 \times 10^5$ a) from potassium was also investigated.

Experimental

Samples and irradiations

Five samples used as targets are listed in Table 1. They consisted of high purity elemental foil or disc (e.g. Al and graphite), or chemical substances of well-defined stoichiometry. Quartz and silicon nitride were available as discs. The potassium bicarbonate had to be pressed into pellets before irradiation.

Irradiations with monoenergetic neutrons of energy 14.6 ± 0.2 MeV were carried out intermittently between June 1992 and February 1993 using the intense D-T neutron generator at the Institute of Experimental Physics, Debrecen University, Hungary. The long irradiation durations of about 30 h *in toto* ensured relatively high fluences between 3.9×10^{12} and 1.3×10^{13} n cm^{-2} which were essential for obtaining sufficient amounts of the long-lived isotope under investigation.

Aluminium monitor foils were attached to each sample on both sides. Sets of three foils were prepared and irradiated in the 0° direction with respect to the 180 ± 5 keV magnetically analyzed deuteron beam. A thick TiT target of 49 mm diameter connected to a wobbling holder in a scatter-free arrangement assured a constant neutron fluence over a long period at 1 mA beam current. The neutron energy was determined via the $^{93}\text{Nb}(n,2n)^{92\text{m}}\text{Nb}$ and $^{90}\text{Zr}(n,2n)^{89}\text{Zr}$ activity ratio method. For details of the irradiation conditions see e.g. Ref. [6]. Neutron fluences were determined via the $^{27}\text{Al}(n,\alpha)^{24}\text{Na}$ reaction using the

* Author for correspondence
(E-mail: ulrich.herpers@uni-koeln.de).

Target material	Supplier, purity and chemical composition (%)	Mass of the target material (mg)	Neutron fluence (10^{12} n cm ⁻²)
Graphite	Union Carbide C: 99.9	91.75	8.65 ± 0.26
Quartz	Heraeus Si: 46.7 O: 53.3	1190.70	3.91 ± 0.12
Silicon nitride	Goodfellow Si: 60.1 N: 39.9	407.15	4.28 ± 0.14
Aluminium	Goodfellow Al: 99.999	20.85	8.69 ± 0.26
Potassium bicarbonate	Pure crystal*	480.90	12.93 ± 0.38

Table 1. Target materials irradiated with neutrons.

*: The pure crystal of KHCO_3 was kindly supplied by the Institut für Kristallographie, Universität zu Köln.

cross section data given in the literature [7]. Additionally, minor changes in the neutron flux densities during the irradiations were recorded with BF_3 long counters.

Radiochemical separations

The preparation of a sample suitable for AMS measurement is a crucial step: radiochemical separation techniques are inevitable. In the present work all the separations were done at the University of Köln, Germany. For the target materials graphite, quartz, silicon nitride and aluminium, separation methods have been reported earlier [cf. 8–12]. We give here only some necessary details. For the separation of ^{36}Cl from potassium bicarbonate, the method used was somewhat different from that employed within the AMS community [cf. 13].

Graphite

The target was dissolved in 10 mL HClO_4 containing 0.5 mg CrO_3 and 1 mg Be-carrier in a teflon vessel while heating to 100 °C for three hours. This catalytic oxidation was obligatory in order to avoid change in the isotopic ratio during this step which could be caused by accidental loss of material before equilibration of isotope and carrier. Teflon remarkably assuages the amount of the isobar ^{10}B – interfering during AMS measurements – which is contained in glass.

After complete dissolution of the target, the mixture was evaporated to dryness and then dissolved in 1 M HCl. Beryllium hydroxide was precipitated with dilute ammonia solution and the remaining Cr(III) was removed by dissolving again in HCl and reprecipitating several times with a mixture of $\text{NH}_3/\text{H}_2\text{O}_2$. The purified hydroxide was then reprecipitated with dilute ammonia and washed five times with distilled and bi-distilled water. In order to remove traces of boron the precipitate was washed with bi-distilled water by heating to 80 °C for half an hour on a water bath. It was then dried at 100 °C and converted to BeO at 800 °C in an oven.

Quartz

After addition of 1 mg Be and 2 mg Al as carriers, the target was dissolved in a mixture of HNO_3/HF by heating to 200 °C in a teflon vessel for 12 hours. The Al-carrier was added to ensure a clean separation of Be from Al. On

evaporation to dryness, silicon was fumed off as SiF_4 with 20 mL of a mixture of HClO_4/HF (1:1). This procedure was repeated three times to assure the removal of all silicon. The residue was then taken up in 1 M HCl. On addition of dilute ammonia, hydroxides were precipitated and separated from the liquid by centrifugation. The hydroxides were then dissolved in 1 M HCl and separated via cation-exchange chromatography (column filled with DOWEX 50 WX8, 25 cm long, 1 cm in diameter). The beryllium fraction was selectively eluted with 120 mL of 1 M HCl and the aluminium fraction with 50 mL of 7.1 M HCl. After reducing the volume of the beryllium fraction to 5–10 mL, the beryllium was reprecipitated and treated further, as described above in the case of graphite.

Silicon nitride

The target was dissolved after addition of 1 mg Be and 2 mg Al as carriers in HNO_3/HF by heating to 150 °C for 24 h using a PARR bomb. In the next step silicon was removed as described above. A very small amount of iron contained in the target-foil had to be separated via anion-exchange chromatography (column filled with DOWEX 1X8, 25 cm long, 1 cm in diameter). The hydroxides were then precipitated with dilute ammonia and the separation of beryllium was done the same way as described above.

Aluminium

The target was dissolved in 4.5 M HCl. Addition of aluminium carrier was not necessary and Be-carrier was omitted. Aluminium hydroxide was precipitated with dilute ammonia, reprecipitated twice, washed three times, then dried and glown to Al_2O_3 at 800 °C.

Potassium bicarbonate

The separation of ^{36}Cl is generally performed via a “high temperature pyrolysis technique” [cf. 13]. We used a different procedure. To the irradiated target 10 mg Cl-carrier was added as well as 1 mL HNO_3 to dissolve it. The chlorine fraction was separated by adding dilute silver nitrate dropwise till in excess. The silver chloride precipitate was kept overnight in darkness. It was then dissolved again in ammonia and reprecipitated with highly diluted nitric acid. This step was repeated three times in order to reduce the small amount of possibly adsorbed sulphur. After centrifugation

Table 2. Isotopic ratios (blank corrected) in AMS measurements.

Target element	$^{10}\text{Be}/^9\text{Be}$ ($\times 10^{-12}$)	$^{26}\text{Al}/^{27}\text{Al}$ ($\times 10^{-12}$)	$^{36}\text{Cl}/\text{Cl}$ ($\times 10^{-12}$)	Mass of carrier (mg)
Graphite	0.228 ± 0.019			0.99
Nitrogen	$< 0.068^*$			2.0
Oxygen	$< 0.068^*$			2.0
Aluminium		19.0 ± 13.4		20.85
Potassium			33.31 ± 0.80	10.3

*: This is the blank value. It was used for calculating the upper limit of cross section.

gation the silver chloride was washed seven times with distilled and bi-distilled water for removing adherent ammonium nitrate. The precipitate was then dried at 80°C .

For every target element the naturally occurring amount of the investigated radionuclide was checked by preparing a blank-sample using non-irradiated material. Possible contaminations or cross-contaminations could thereby be recorded.

Accelerator mass spectrometry

The oxides were mixed with copper (1:4) and pressed into target holders designed for AMS measurements at the ETH/PSI, Zürich, Switzerland (for details, cf. Refs. [14, 15]). The final step of the preparation of silver chloride for the AMS measurement was done in Zürich. The results of the AMS measurements are given in Table 2.

The following AMS standards were used:

^{10}Be : S 555 $N^*/N = (9.55 \pm 0.23) \times 10^{-11}$

^{26}Al : ZAL 94 $N^*/N = (5.26 \pm 0.37) \times 10^{-10}$

^{36}Cl : K 380/4 $N^*/N = (1.53 \pm 0.15) \times 10^{-11}$

In actual experimental runs, measurements were done for the pairs $^{10}\text{Be}/^9\text{Be}$, $^{26}\text{Al}/^{27}\text{Al}$ and $^{36}\text{Cl}/\text{Cl}$, i.e. the ratio of the atoms of the radioactive product to those of the inactive carrier was determined.

Calculation of cross sections and error estimates

With the isotopic ratios (N^*/N) derived from AMS measurements, cross sections were calculated by applying the well-known activation formula for the special case of very long-lived activation products.

$$\sigma [\text{mbarn}] = \frac{N^*}{N} \cdot \frac{m_{\text{carrier}} \cdot M_A}{m_A \cdot M_{\text{carrier}} \cdot \Phi \cdot t \cdot 10^{-27}}$$

m_{carrier} : mass of carrier; m_A : mass of target A; M_A : molar mass of the target; M_{carrier} : molar mass of carrier; Φ : neutron flux density ($\text{n cm}^{-2} \text{s}^{-1}$); t : time of irradiation (s).

For error estimation one has principally to consider the error involved in the determination of the neutron flux density and hence the fluence (cf. Table 1). Furthermore, every individual AMS measurement contributes to the total error which can turn out to be the main error for the case of low isotopic ratios (below 10^{-13}). In the case of ^{26}Al determination, the current was exceptionally low; the number of events recorded was small and hence the statistical error was rather large. Finally, the chemical separation leads to an error of about 3%. Combining the individual errors in

Table 3. Experimental cross sections for the formation of long-lived products ^{10}Be , ^{26}Al and ^{36}Cl in the interactions of 14.6 MeV neutrons with various materials.

Target element	Nuclear reaction	Cross section (mb)
Graphite	$^{13}\text{C}(\text{n},\alpha)^{10}\text{Be}$	0.24 ± 0.02
	$^{12}\text{C}(\text{n},\alpha)^{10}\text{Be}$	$22 \pm 2^*$
Nitrogen	$^{14}\text{N}(\text{n},\alpha)^{10}\text{Be}$	< 0.3
Oxygen	$^{16}\text{O}(\text{n},\alpha)^{10}\text{Be}$	< 0.1
Aluminium	$^{27}\text{Al}(\text{n},2\text{n})^{26}\text{Al}$	22 ± 16
Potassium	$^{39}\text{K}(\text{n},\alpha)^{36}\text{Cl}$	159 ± 8

*: Considering that in graphite only the $^{13}\text{C}(\text{n},\alpha)$ -reaction leads to ^{10}Be .

quadrature we obtained the total error of each cross section value.

Results and discussion

The isotopic ratio $^{10}\text{Be}/^9\text{Be}$ for irradiated graphite, $^{26}\text{Al}/^{27}\text{Al}$ for aluminium and $^{36}\text{Cl}/\text{Cl}$ for potassium exceeded the respective blank-ratio by at least one order of magnitude. On the other hand the $^{10}\text{Be}/^9\text{Be}$ ratio for nitrogen and for oxygen was comparable to the respective blank ratio so that for those two elements only an upper limit of the corresponding cross section could be given.

A summary of the results is given in Table 3. The formation of ^{10}Be in the interaction of graphite with 14.6 MeV neutrons occurs only via the reaction channel $^{13}\text{C}(\text{n},\alpha)^{10}\text{Be}$. Similarly, in the case of the target element potassium, ^{36}Cl can be produced only via the $^{39}\text{K}(\text{n},\alpha)^{36}\text{Cl}$ reaction. For the first reaction no cross section data in this energy region are available. For the latter, only a few rather old data [cf. 16–18], determined via α -counting, exist. Those data give a sum of all α -emitting processes, i.e. (n,α) , $(\text{n},\text{n}'\alpha)$, $(\text{n},2\alpha)$ etc. At $E_n \approx 14$ MeV, total α -emission cross sections of about 110 mb [cf. 16, 17] and 160 mb [18] have been reported. Our value of 159 mb for the pure (n,α) process on ^{39}K , obtained via the AMS technique, is within the range of values reported via α -counting.

It should be mentioned that detailed empirical systematics for (n,α) reactions exist [cf. 19–26]. In general, for medium and heavy mass targets the (n,α) cross section decreases exponentially with the increasing asymmetry parameter $[(N-Z)/A]$. Within this frame a few semi-empirical formulae have been worked out. A comparison of the experimental results for the two reactions studied here with the predictions from the systematics shows good agree-

ment. This holds even for the reaction $^{13}\text{C}(n,\alpha)^{10}\text{Be}$, though it must be mentioned that the validity of most of the semi-empirical descriptions is definitely restricted to medium and heavy mass targets. The cross section for the $^{39}\text{K}(n,\alpha)^{36}\text{Cl}$ reaction is the highest as compared to any other (n,α) reaction with 14.6 MeV neutrons. This is possibly due to the very high binding energy of the neutron, favouring thereby the emission of charged particles. However, further investigations are needed to clarify the mechanism of the α -particle emission from this nucleus.

The excitation function of the $^{27}\text{Al}(n,2n)^{26}\text{Al}$ reaction has been experimentally determined by several groups [cf. 27–32], using both radiometric [28, 29, 31] and AMS [27, 30, 32] methods. The reason for relatively extensive studies from threshold up to about 40 MeV is its relevance to FRT. Al is potentially interesting as structural material provided its activation via the $(n,2n)$ reaction is not high. Out of all the studies, the AMS work of Wallner *et al.* [32] is very comprehensive. Our value at 14.6 MeV is within the range of reported AMS data, though the error in our work is rather large. The cross section also falls within the range of values predicted from the systematics of $(n,2n)$ reaction cross sections at 14.6 MeV [cf. 33].

The formation of ^{10}Be in the interactions of 14.6 MeV neutrons with ^{14}N and ^{17}O targets can occur only via $^{14}\text{N}(n,\alpha p)^{10}\text{Be}$ and $^{17}\text{O}(n,2\alpha)^{10}\text{Be}$ processes, respectively. However, as only upper limits of cross sections (and not concrete values) for the two elements of natural composition could be determined (cf. Table 3), we did not attempt to deduce the real cross sections of the two above mentioned contributing processes.

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