Excitation Function for the ^{24}Mg(^{15}N, αn)^{34m}Cl Reaction

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Abstract

In order to determine traces of magnesium in alumina by Heavy Ion Activation Analysis (HIAA), the yield curve and the total cross sections for the ^{24}Mg(^{15}N, αn)^{34m}Cl reaction were determined in the 19 to 31 MeV energy range of incident ^{15}N ions. The yields of characteristic γ-rays (E_γ = 511 keV) were detected by coincidence measurements using two 3″ × 3″ NaI(Tl) scintillation detectors.

Introduction

The trace determination of light elements by HIAA is often sensitive and selective [1]. Knowledge of the excitation functions of the nuclear reactions involved in the procedure is useful to find out the best experimental conditions for an analytical application.

In this work, the excitation function for the ^{24}Mg(^{15}N, αn)^{34m}Cl reaction has been determined for incident ^{15}N ion energies ranging from 19 to 31 MeV, in order to study the possibility for a selective and sensitive determination of magnesium in an alumina matrix. This reaction is of special interest (i) because of the influence on the kinetics of grain growth of a small amount of magnesium introduced as dopant in Al_2O_3 [2] and (ii) because other analytical techniques for magnesium determination are hampered by interferences due principally to aluminum [3, 4, 5].

Experimental

Beams of ^{15}N ions at various energies and charge states were obtained with the HV EC-EN Tandem Van de Graaff accelerator of the Institute for Medium Energy Physics at the Swiss Federal Institute of Technology in Zürich, using 98% enriched ammonia gas produced by the US Department of Energy.

MgO was used for the cross section measurements to avoid the oxidation problem of pure magnesium, which could introduce an uncertainty in the estimation of the ion range. Targets were prepared by pelleting this powder as small discs of 13 mm diameter and 1–2 mm thickness. Nuclear interferences were checked with pelleted compounds or with pure metallic foils. All targets were thicker than the range of the incident ion beam.

The experimental irradiation set-up has been described in an earlier paper [6]. The vacuum level in the irradiation chamber was 10^{-5} mm Hg. The diameter of the ^{15}N ion beam on the target was restricted to 7 mm by two tantalum collimators. The number of ^{15}N ions striking the target was determined from the integrated charge of a wire mesh (57% transmission) placed 15 cm in front of the target. Prior to the target irradiation, the current ratio was determined with a Faraday cup mounted in place of the target. In a typical experiment, the targets were irradiated for 10 minutes in a 150 to 250 nA · cm^{-2} beam. Under these conditions, no target deterioration was observed.

Irradiated targets were mounted on a CEA source holder between two thin polyethylene foils and placed in an aluminum box whose wall thickness had been calculated to increase the yield of the 511 keV γ-ray from the positron annihilation. An automatic sample changer for coincidence counting equipped with two 3″ × 3″ NaI(Tl) scintillation detectors was used to measure the activity and to follow the decay curves. These curves were then deconvoluted using the CLSQ computer program [7]. The overall counting efficiency of the system, determined with a standard ^{22}Na source, was 8.5%.

Cross section calculation

The activity resulting from the production of a given radionuclide following irradiation of a thick target with heavy ions can be expressed as:

$$A = n\phi(1 - e^{-\lambda t}) \int_E^0 \sigma(E) \left(\frac{dx}{dE}\right) dE \tag{1}$$

A activity of the radionuclide of interest (Bq)
n concentration of atoms from which the radionuclide is formed (atom g^{-1})
ϕ heavy ion flow (ions s^{-1})
$\sigma(E)$ cross section for incident energy E (cm^2)
$\dfrac{dE}{dx}$ differential energy loss of the incident ions (MeV g^{-1} cm^2)
λ decay constant (s^{-1})
t irradiation time (s)

The yield of the nuclear reaction can then be defined as:

$$Y = \frac{A}{\phi(1 - e^{-\lambda t})} \tag{2}$$

which can be determined experimentally for each inci-

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dent energy using the relations:

$$A = \frac{a_0}{\epsilon u \, \xi} \tag{3}$$

and

$$\phi = \frac{6.24 \times 10^{18} \, (I \, t)}{z \, t} \tag{4}$$

a_0 counting rate at the end of irradiation (s^{-1})
ϵ detector efficiency for the γ-ray of interest
u abundance of the γ-ray of interest [8]
ξ isotopic abundance of the target nucleus
$(I \, t)$ integrated charge collected on target (C)
6.24×10^{18} beam current (I) and ion flow (ϕ) interrelation factor
z charge of incident heavy ion

The experimental yield curve thus obtained, given in Bq ion^{-1} s vs. MeV, was smoothed using a computer program [9], which took into account the errors on each separate value of $Y(E)$.

By combining equations (1) and (2), the yield expression becomes:

$$Y = n \int_E^0 \sigma(E) \left(\frac{dx}{dE}\right) dE \tag{5}$$

The derivative of this latter equation at a definite energy leads to the cross section

$$\sigma(E) = \frac{1}{n} \left(\frac{dY}{dE}\right)_E \left(\frac{dE}{dx}\right)_E \tag{6}$$

where the second term is the derivative of the experimental reaction yield at the energy E and the third term represents the differential energy loss of the incident heavy ion as given by NORTHCLIFFE et al. [10].

Results and discussion

The ^{34m}Cl yield curve for the nuclear reaction studied was obtained by irradiating natural magnesium (MgO), which is composed of 79% ^{24}Mg, 10% ^{25}Mg and 11% ^{26}Mg. The ^{24}Mg(^{15}N, αn)^{34m}Cl reaction, whose threshold energy is 1.2 MeV, is probably the one that leads to the production of almost all ^{34m}Cl activity. It is however possible that ^{25}Mg also produces ^{34m}Cl via the ^{25}Mg(^{15}N, α2n)^{34m}Cl reaction (E_{th} = 10.0 MeV). On the other hand the production of ^{34m}Cl via the ^{26}Mg(^{15}N, α3n)^{34m}Cl reaction is unlikely owing to its high threshold energy (E_{th} = 28.0 MeV).

A typical decay curve is presented in Fig. 1. Beside the expected ^{34m}Cl activity, ^{38}K (T = 7.7 min) was also produced. Furthermore, a complete γ-spectrum measured with a Ge(Li) detector showed the formation of ^{28}Al (E_γ = 1779 keV, T = 2.25 min) and of ^{29}Al (E_γ = 1273 keV, T = 6.6 min) for all ^{15}N beam energies ranging from 19 to 31 MeV.

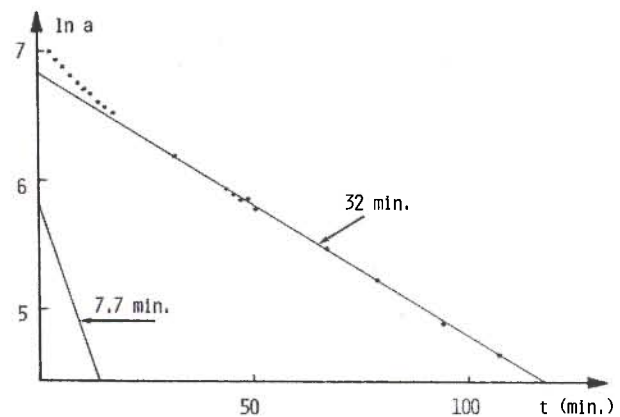
Fig. 1. Typical decay curve (E^{15}N = 30 MeV, irradiation time = 10 min, counting time = 100 s.).

The ^{34m}Cl production yield vs. incident energy is presented in Fig. 2. The curve rapidly increases for energies up to 27 MeV before bending slowly without reaching a plateau at 31 MeV. The Tandem Van de Graaff does not provide sufficient beam current for investigation of higher energies.
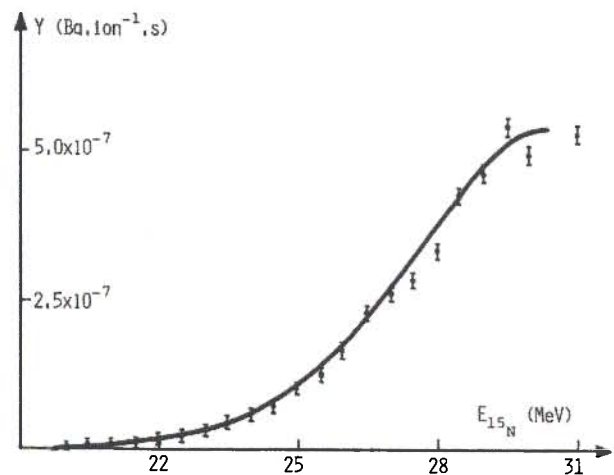

Fig. 2. ^{34m}Cl yield curve for the reaction ^{24}Mg(^{15}N, αn).

The derivative of the smoothed yield curve was divided by the number of Mg atoms contained in 1 mg of MgO and combined with the differential energy loss of the ^{15}N ions in the MgO matrix. The cross sections thus obtained for the reaction of interest lead to the excitation function shown in Fig. 3. The errors for the cross section values are ± 15%. The maximum cross section was found to be 40 ± 6 mb corresponding to a 28 MeV ^{15}N beam. This value is large enough to be utilized in an analytical application. With this purpose in mind, we tested a series of elements which would likely produce ^{34m}Cl through a nuclear reaction with ^{15}N ions. With the aid of the mass excess table of WAPSTRA et al. [11], these potential nuclear interferences were selected with a computer. The program [12] calculates the Q value, the threshold energy and the Coulomb barrier of any possible reaction with ^{15}N ions on target nuclei. Among the elements tested, only Na and Al produced ^{34m}Cl with the ratios of specific activities given in Table 1. It shows that due to the interference
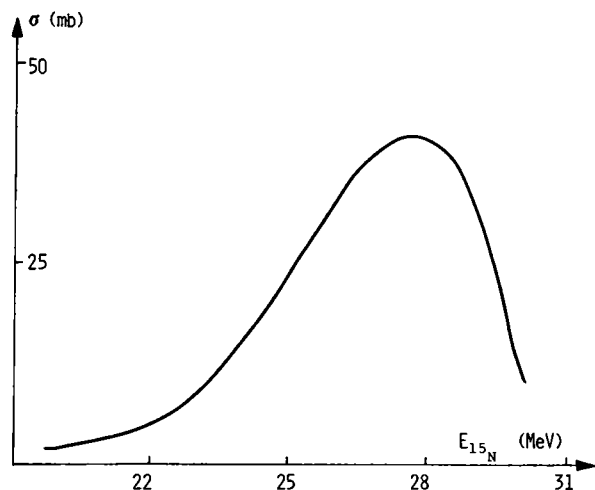
Fig. 3. Total cross sections for the reaction ^{24}Mg(^{15}N, αn).

Table 1. Relative extent of nuclear interferences for magnesium determination

Element	Ratio of specific activities at			
	30 MeV	27 MeV	25 MeV	20 MeV
Na	0.06	0.025	0.01	0
Al	0.001	0.0003	0	0

arising from aluminum, a trace determination of magnesium in Al_2O_3 is only possible if the incident beam energy is kept below 25 MeV ^{15}N. For different types of matrices, on the other hand, a sensitive magnesium determination can be undertaken with beam energies above this latter energy providing that the sodium content of the sample is not higher than that of magnesium. The detection limit for a non-destructive magnesium determination in Al_2O_3 was found to be 100 ppm; this relatively poor value is due to instrumental interferences occurring with this matrix. However, for heavier metals such as gold, the detection limit at 30 MeV ^{15}N was found to be 5 ppm.

Moreover, owing to the low ion flow, this technique can be applied to non-destructive determinations of magnesium in biological material.

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