Excitation Functions for the Reactions ${}^7\text{Li}({}^{15}\text{N}, {}^1\text{H}){}^{21}\text{F}$, ${}^{6,7}\text{Li}({}^{15}\text{N}, {}^{1,2}\text{H}){}^{20}\text{F}$ and ${}^9\text{Be}({}^{15}\text{N}, 2\alpha){}^{16}\text{N}$

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${}^{15}\text{N}$ reactions / Lithium and beryllium target / Excitation function / Yield

Abstract

The yield curves and the excitation functions for the ${}^7\text{Li}({}^{15}\text{N}, {}^1\text{H}){}^{21}\text{F}$, ${}^{6,7}\text{Li}({}^{15}\text{N}, {}^{1,2}\text{H}){}^{20}\text{F}$ and ${}^9\text{Be}({}^{15}\text{N}, 2\alpha){}^{16}\text{N}$ reactions were determined in the 10 to 30 MeV energy range. The highest yield was obtained with the reaction ${}^9\text{Be}({}^{15}\text{N}, 2\alpha){}^{16}\text{N}$. The maximum cross-section for this reaction is 370 ± 70 mb at 24 MeV.

Introduction

During the past years, charged particle activation has proved to be an outstanding analytical technique for determining light elements [1]. The recent development of heavy ion beams has further increased the number of nuclear reactions available for that purpose. The potential applicability of ${}^7\text{Li}$, ${}^9\text{Be}$, ${}^{10}\text{B}$, ${}^{11}\text{B}$, and ${}^{18}\text{O}$ ion beams for the determination of light elements have been described in preceding papers [2–6]. The main advantages of the technique consist firstly in the linear relation between the signal intensity and the concentration of the element to be determined, and secondly in the independence of the signal from the chemical nature of the matrix. Contrarily to most of the other direct analytical methods applicable to the solid phase, this method allows the direct comparison of the activity produced in a standard with that of the sample to be analyzed. A very precise calibration technique, described by ISHII *et al.* [7], is next used to calculate the concentration of the trace element. To apply ISHII's technique one needs, however, to know the activation curve of the nuclear reaction of interest. For this reason and also to define the best experimental conditions for the determination of lithium and beryllium, the activation curves and the excitation functions for the ${}^7\text{Li}({}^{15}\text{N}, {}^1\text{H}){}^{21}\text{F}$, ${}^{6,7}\text{Li}({}^{15}\text{N}, {}^{1,2}\text{H}){}^{20}\text{F}$ and ${}^9\text{Be}({}^{15}\text{N}, 2\alpha){}^{16}\text{N}$ reactions have been established.

Experimental

Irradiation

Beams of ${}^{15}\text{N}$ ions at various energies and in various charge states were obtained from the HV EC-EN Tandem Van de Graaff accelerator of the Institute for Medium Energy Physics at the Swiss Federal Institute of Tech-

nology in Zürich. Enriched ammonia (98% ${}^{15}\text{NH}_3$, US Department of Energy) was used to produce the ${}^{15}\text{N}$ ion beam. The gas, mixed with hydrogen, was ionized using a tungsten filament at high temperature to produce ${}^{15}\text{NH}_3^+$ ions.

The irradiation chamber, especially designed for determining the yields of short lived radionuclides, has been described in a preceding paper [8]. A mobile target holder permits cyclic irradiations and activity measurements without interruption of the vacuum (10^{-5} mm Hg). The beam current was monitored immediately after the measurement of the sample activity with a Faraday cup placed directly behind the target. The whole cycle (irradiation – activity measurement – beam monitoring) was electronically controlled and repeated for at least 10 cycles.

Detection

A coaxial Ge(Li) detector (ORTEC 8001 – 1020 V, 2.0 keV resolution at 1.33 MeV) was used to detect the γ radiation emitted by ${}^{16}\text{N}$, ${}^{20}\text{F}$ and ${}^{21}\text{F}$, through a thin plexiglass window attached to the irradiation chamber. The detector was connected to a multichannel analyzer (CANBERRA 80). At the end of the last counting, the total spectrum, obtained by summing all the individual countings, was transferred to a floppy disc recorder (SCINETIFIC MICRO SYSTEM, D222) for further analysis with a PDP 11/23 computer. The absolute detection efficiency for the 350 keV γ -rays of ${}^{21}\text{F}$ was determined to be $(1.65 \pm 0.05) \times 10^{-2}$ in the counting conditions, using a ${}^{152}\text{Eu}$ standard source. For the 1634 keV γ -rays of ${}^{20}\text{F}$, this value was found to be $(2.6 \pm 0.1) \times 10^{-3}$. In the case of ${}^{16}\text{N}$, the detector's efficiency was estimated to be $(3.5 \pm 0.5) \times 10^{-4}$ at 6128 keV, as suggested by SEYFARTH *et al.* [9].

Targets

Beryllium was irradiated as pure metallic foils (Be, 99.8%, GOODFELLOW METALS Ltd.). The lithium targets consisted of p. a. LiF (99.9%, MERCK AG); the powder was dried and pressed in small discs of 13 mm diameter and 1–2 mm thickness. All targets were thicker than the range of the incident ions.

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Quantitation

Due to the cyclic mode of activation and the summation of the consecutive spectra, the primary results were obtained as the total number of counts, N_t , detected in the γ -line of the radionuclide of interest during k cycles. The results are related to the yield of the nuclear reaction by the equation given by BATCHELOR *et al.* [10]:

$$\eta = \frac{N_t \lambda (1 - e^{-\lambda t_i}) (1 - e^{-\lambda t_c})}{\phi \epsilon \xi \mu (1 - e^{-\lambda \Delta t}) e^{-\lambda t_D}} \left[k + \frac{1 - e^{-\lambda k t_c}}{1 - e^{-\lambda t_c}} \right]^{-1}$$

with η = yield of the nuclear reaction (Bq s ion⁻¹)

λ = decay constant (s⁻¹)

t_i = single irradiation time (s)

t_c = time separating two consecutive irradiations (s)

ϕ = incident particle intensity (ion s⁻¹)

ϵ = absolute efficiency of the detector for the corresponding γ energy

ξ = abundance of the detected γ -rays (11)

μ = natural abundance of the target nucleus

Δt = single counting time (s)

t_D = delay between the end of irradiation and the beginning of counting (s)

The activation curve $\eta(E)$ for a given reaction could then be obtained by reporting the yield as a function of the beam energy. This curve was used to determine the average activation energy according to ISHII [7]. Its derivative allowed the calculation of the excitation function applying the relation:

$$\sigma(E) = \frac{1}{n} \left(\frac{d\eta}{dE} \right)_E \left(\frac{dE}{dx} \right)_E$$

with σ = cross-section of the reaction (mb)

n = concentration of the target nuclei (at g⁻¹)

$\frac{dE}{dx}$ = stopping power of the target material [12].

Results and discussion

The nuclear reactions investigated are listed in Table 1, together with their Q -value and Coulomb barrier (E_c). All the reactions are exoenergetic and thus their threshold energy is zero. Beryllium has one single natural isotope and therefore the ¹⁶N activities measured at various incident beam energies are directly related to the reaction cross-sections. Lithium occurs naturally as ⁶Li (7.5%) and ⁷Li (92.5%). While ²¹F can only be produced by the reaction with ⁷Li, ²⁰F might be formed by the ¹⁵N bombardment of both ⁶Li and ⁷Li. In this latter case, the ²⁰F activity measurements yield the determination of an overall activation curve and cross-section values.

The characteristics of the radionuclides produced are also given in Table 1. The cyclic activation set-up allowed the accurate measurements of the production yields of ²⁰F, ²¹F, and ¹⁶N in spite of their short half-lives [13].

The experimental yield curves of the nuclear reactions investigated, obtained by reporting the yield as a func-

Table 1. Kinematic parameters of the reactions and characteristics of the products

Reaction	Radionuclide produced				
	Q (MeV)	E_c (MeV)	$T_{1/2}$ (s)	E_γ (keV)	γ -Rays intensities
⁷ Li(¹⁵ N, ¹ H) ²¹ F	7.7	14.5	4.35	350	0.70
⁶ Li(¹⁵ N, ¹ H) ²⁰ F	6.9	16.5	11.4	1634	1.00
⁷ Li(¹⁵ N, ² H) ²⁰ F	1.8	14.0		1122 611	P P
⁹ Be(¹⁵ N, 2 ⁴ He) ¹⁶ N	0.8	15.2	7.13	6128 5617 5106	0.69 P P

Fig. 1. Yield curves of the reactions a) ⁹Be(¹⁵N, 2 α)¹⁶N, b) ^{6,7}Li(¹⁵N, ^{1,2}H)²⁰F, c) ⁷Li(¹⁵N, ¹H)²¹F.

tion of the incident beam energy, are presented in Figure 1. In order to facilitate their comparison, the yield scale is logarithmic. The error on individual yields has been estimated to be 5%, taking into account the counting statistics and the error on the various parameters involved in the cyclic mode of irradiation and activity measurement. The ⁹Be(¹⁵N, 2 α)¹⁶N reaction is characterized by relatively high yields whatever the beam energy. Hence the sensitivity could be excellent for analytical applications. The yield curves of both ⁹Be(¹⁵N, 2 α)¹⁶N and ⁷Li(¹⁵N, p)²¹F reactions reach a plateau at 30 MeV ¹⁵N while the ²⁰F yield continues to increase beyond that limit.

For each reaction, the experimental activation curves were refined using a computer program [14]. They were expressed as a polynomial function of the 4th order by a least square fitting. The derivative of these expressions, $d\eta/dE$, at a given beam energy led to the determination

Fig. 2. Excitation functions of the reactions a) ${}^9\text{Be}({}^{15}\text{N}, 2\alpha){}^{16}\text{N}$, b) ${}^6,{}^7\text{Li}({}^{15}\text{N}, {}^{1,2}\text{H}){}^{20}\text{F}$, c) ${}^7\text{Li}({}^{15}\text{N}, {}^1\text{H}){}^{21}\text{F}$.

of the corresponding excitation function presented in Figure 2. The errors on the cross-sections were estimated to be about 20% due to the fitting and the derivative calculations.

The examination of Figure 2 and Table 2, which records the maximum cross-sections with the corresponding beam energies, confirms the excellent yield of the ${}^9\text{Be}({}^{15}\text{N}, 2\alpha){}^{16}\text{N}$ reaction. At this point, it is worth noticing the similitude of this reaction with ${}^9\text{Be}({}^{18}\text{O}, 2\alpha){}^{19}\text{O}$ (owing the same target nucleus and the same nature and number of particles ejected) which has also relatively high cross-sections [15]. On the other hand, the cross-section of the reaction ${}^7\text{Li}({}^{15}\text{N}, p){}^{21}\text{F}$ remains low for the whole range of energy studied; the maximum value barely reaches 30 mb at 20 MeV ${}^{15}\text{N}$, while the excitation functions of the two other reactions show maximum values for higher beam energies.

Table 2. Maximum cross-sections

Reaction	Energy (MeV)	Cross-section (mb)
${}^7\text{Li}({}^{15}\text{N}, {}^1\text{H}){}^{21}\text{F}$	20	30 ± 6
${}^6,{}^7\text{Li}({}^{15}\text{N}, {}^{1,2}\text{H}){}^{20}\text{F}$	25	160 ± 30
${}^9\text{Be}({}^{15}\text{N}, 2\alpha){}^{16}\text{N}$	24	340 ± 70

Conclusion

The activation curves and the excitation functions for the nuclear reactions induced by bombarding lithium and

beryllium with energetic ${}^{15}\text{N}$ ions have been determined. The short half-life of the radionuclides produced and the relatively high cross-sections should allow the fast, sensitive, nondestructive and simultaneous determination of traces of lithium and beryllium [16].

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Literature

- GIRARDI, F.: Radioactivation Analysis. Past Achievements, Present Trends and Perspective for the Future. *J. Radioanal. Chem.* **69**, 15 (1982).
- FRIEDLI, C., SCHWEIKERT, E. A., LERCH, P.: Studies in Heavy Ion Activation Analysis. VI. Trace Determination Possibilities with ${}^7\text{Li}$ Ion Bombardment. *J. Radioanal. Nucl. Chem.* **88**, 369 (1985).
- FRIEDLI, C., SCHWEIKERT, E. A., LERCH, P.: Studies in Heavy Ion Activation Analysis. IX. Trace Determination Possibilities with ${}^9\text{Be}$ Ion Bombardment. *J. Radioanal. Nucl. Chem.* **116**, 401 (1987).
- FRIEDLI, C., SCHWEIKERT, E. A., LERCH, P.: Studies in Heavy Ion Activation Analysis. VII. Trace Determination Possibilities with ${}^{10}\text{B}$ Ion Bombardment. *J. Radioanal. Nucl. Chem.* **90**, 341 (1985).
- FRIEDLI, C., COLIN, M. E., LERCH, P.: Studies in Heavy Ion Activation Analysis. X. Trace Determination Possibilities with ${}^{11}\text{B}$ Ion Bombardment. *J. Radioanal. Nucl. Chem.* **120**, 253 (1988).
- FRIEDLI, C., LASS, B. D., SCHWEIKERT, E. A.: Studies in Heavy Ion Activation Analysis. II. Trace Determination Possibilities with ${}^{16}\text{O}$ Ion Bombardment. *J. Radioanal. Chem.* **54**, 281 (1979).
- ISHII, K., VALLADON, M., SASTRI, C. S., DEBRUN, J. L.: Accurate Charged Particle Activation Analysis: Calculation of the Average Energy in the Average Stopping Power Method. *Nucl. Instrum. Methods* **153**, 503 (1978).
- FRIEDLI, C., ROUSSEAU, M., DIACO, T., LERCH, P.: Dosage de traces de soufre et de béryllium par activation dans un faisceau d'oxygène-18. *Analusis* **13**, 176 (1985).
- SEYFARTH, H., HASSAN, A. M., HRASTNIK, B., GOETTEL, P., DALANG, W.: Efficiency Determination for some Standard Type Ge(Li) Detectors for Gamma-Rays in the Energy Range from 0.04 to 11 MeV. *Nucl. Instrum. Methods* **105**, 301 (1972).
- BATCHELOR, R., McK. HYDER, H. R.: The Energy of Delayed Neutrons from Fission. *J. Nucl. Energy* **3**, 7 (1956).
- ERDTMANN, G., SOYKA, W.: *The Gamma-Rays of Radionuclides. Topical Presentation in Nuclear Chemistry*, Vol. 7, Verlag Chemie, Weinheim 1979.
- ZIEGLER, J. F.: *Handbook of Stopping Cross-Sections for Energetic Ions in all Elements*, Vol. 5, Pergamon Press, New York 1980.
- SPIROU, N. M.: Cyclic Activation Analysis – A Review. *J. Radioanal. Chem.* **61**, 211 (1981).
- GOUMAZ, J. Y., DURAND, C., OTTIN, T.: Programme FIDES, LTA-GRH Report 14B, EPF-Lausanne 1983.
- DIACO, T., FRIEDLI, C., LERCH, P.: Excitation Functions for the ${}^9\text{Be}({}^{18}\text{O}, 2\alpha){}^{19}\text{O}$ and ${}^9\text{Be}({}^{18}\text{O}, d){}^{25}\text{Na}$ Reactions. *Radiochim. Acta* **38**, 185 (1985).
- FRIEDLI, C., COLIN, M. E., LERCH, P.: Studies in Heavy Ion Activation Analysis. XI. Trace Determination Possibilities with ${}^{15}\text{N}$ Ion Bombardment. *J. Radioanal. Nucl. Chem.* **122**, 115 (1988).