Submitted to Journal
of Radioanal. Chem.

LBL-682
Preprint

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June 1972

AEC Contract No. W-7405-eng-48

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June 9, 1972

RESUMÉ

Three easily-detected radionuclides, ^{38}Cl, ^{34m}Cl, and ^{38}K, produced from the interactions of sulfur, chlorine, potassium, and calcium, were used as "signals" for the determination of the latter elements by He^3 activation analysis. A number of samples were analyzed non-destructively by using differing nuclear reactions to examine the accuracy of the method and the extent of interferences. Absolute excitation functions for the He^3 reactions are given.

INTRODUCTION

The unique advantages that the ^3He ion offers as an incident particle for activation analysis were first suggested by Markowitz and Mahony.[1] Since then considerable efforts have been devoted to developing the practicality of this system for solving various analytical problems.[2-7] Although ^3He activation analyses of oxygen, carbon, and fluorine have been investigated extensively in recent years,[8-12] there is much less information available about the ^3He reactions with sulfur, chlorine, potassium, and calcium. Activation of S, Cl, K, and Ca by thermal neutrons, fast neutrons, protons, deuterons, and alpha particles has been reported;[13-19] most of these analyses were applied to a

*Work was performed under the auspices of the U. S. Atomic Energy Commission.

specific matrix with various degrees of success. The use of ^{3}He ions may provide alternative method for the determination of the above elements with high sensitivity and a more convenient one in some cases, particularly at low energy.

In order to utilize the charged-particle activation technique to its optimum, a measurement of activation cross sections for a specific nuclear reaction is essential. This knowledge of excitation functions is also valuable for the production of radionuclides and for studies of nuclear reaction mechanisms.

EXPERIMENTAL PROCEDURES

Target preparation and assembly

For the measurements of the excitation functions the targets used were prepared by vacuum-depositing CaF_2, KI, $PbCl_2$ and pure sulfur onto tantalum backing-foils. Sulfur targets were protected by depositing a thin film of aluminum over the sulfur to promote heat transfer during the irradiations. The thickness of the targets were determined by weighing; they ranged from 1.5 to 4.5 mg/cm^2. For the subsequent analyses, samples were synthesized as thin targets or as thick ones. Thin targets were prepared either by vacuum evaporation or by settling the finely-ground powder onto a tantalum backing-foil from an ether suspension; the powder was fixed to the foil with a drop of dilute polystyrene in dichloroethylene. The thick targets were prepared by homogenizing a known, small amount of material to be analyzed, with pure lead powder (approx. 200 mg/cm^2) and then pressing the mixture into a 1-inch (2.54 cm) diameter thin disk. Targets were mounted on an aluminum ring and were covered with thin gold or aluminum foils to form one target assembly. This target assembly was constructed to facilitate the handling of a fragile target, to promote heat transfer, to avoid recoil loss, and to obtain a proper, reproducible geometry.

Irradiation

Irradiations were performed at the Berkeley 88-inch cyclotron. Aluminum foils were placed in front of the sample assembly to degrade the beam from an initial energy of 30-MeV ^{3}He to an appropriate energy for each irradiation. The length of bombardment varied between 8 to 15 minutes. The average beam currents

were about 0.1 μA to 0.4 μA of ^{3}He (++). Thin targets were irradiated simultaneously at different energies by stacked-foil technique; thick targets were irradiated one-at-a-time with a thin standard in the same stack, or with a thick standard from two successive irradiation. The total charge received from the Faraday cup was measured by a calibrated integrating electrometer.

Radioactivity measurements and data acquisition

After irradiation, samples were analyzed by gamma-ray spectrometry using Ge(Li) detectors, coupled to a 1024-channel analyzer with a magnetic tape recording unit.

Two Ge(Li) detectors, having active volumes of 16 cm^3 and 30 cm^3, manufactured by the nuclear detector group at LBL, were used for this work. The gamma-ray photo-peak efficiencies were calibrated using a set of eight absolute gamma-ray standards, obtained from the International Atomic Energy Authority, Vienna.

Half-lives and gamma rays used for the measurements are given in Table I.

RESULTS AND DISCUSSION

Excitation functions

Absolute cross sections were calculated based on the assumption that the reactions in Table I were attributed entirely to a single reaction of a specific target nuclide. A summary of experimental cross-section data obtained in this work is presented in Table II, and the excitation functions for each element are shown in Fig. 1 through 4. Each of the cross sections shown in Table II represents an average value of 2-3 replicates; the precision (average deviation) was about 5%. Additional uncertainties include: (1) the determination of total beam current, (2) the half-lives of nuclides, (3) the determination of disintegration rates, (4) the uniformity of thickness of targets, (5) possibility of recoil gas losses during irradiation. With all the above considerations, the estimated absolute errors in the cross sections are 10% for S data, 10-15% for Cl, 20% for K, and 10% for Ca.

Interferences

There are two types of interferences in non-destructive activation analysis using gamma-ray spectrometry. The first type of interference is due to the closely-spaced gamma-rays, or gamma-rays having the same energies; the second type is caused by the production of the same nuclide from more than one element. Interference of the first type may be minimized by using special counting techniques such as coincidence or anti-coincidence counting, or using a high resolution Ge(Li) gamma-ray detector system. (One can resort to chemical separations of course, destroying the sample.) Often, there is more than one gamma ray emitted from the same nuclide; the choice of using a particular gamma ray depends on two important factors: the relative intensity and the presence of other gamma-rays. In view of these interference possibilities, gamma rays of high energy were chosen for the measurements of ^{38}Cl, ^{34m}Cl, and ^{38}K in this work.

Relative activities of the same reaction product-nuclide formed from more than one element in the activations of S, Cl, K and Ca are shown in Tables III and IV. Chlorine can be determined unambiguously by detection of ^{38}Cl from $^{37}Cl(^{3}He, 2p)^{38}Cl$ reaction, because no other element will produce ^{38}Cl under our conditions. However, for the determinations of Ca, K, and S, the interfering activities, if present, cannot be completely eliminated. It is possible to minimize these interferences by the adjustment of bombarding energies. The extent of interference can be assessed by knowing the ratio of production from the interfering element. For example, to determine Ca in the presence of K and Cl at 20-MeV ^{3}He bombarding energy, the ^{38}K acitivty produced from K and Cl can be estimated by the activity of ^{34m}Cl and ^{38}Cl.

Sensitivity of the method

The sensitivity of the method may be predicted by considering the systematic parameters which govern the production of activities from sought elements in an easily obtainable condition. The parameters are the incident beam energy, the beam current, the counting efficiency, the counting interval, and the duration

of irradiation. The detailed mathematical and statistical treatment of the limits for qualitative detection and quantitative determination in application to radiochemistry has been discussed by Currie.[20] Applying Currie's "working expression" for the interference-free determinations of S, Cl, K, and Ca by activation with ^{3}He at the energy of the maximum cross section, using 1 μA current for one half-life irradiation, starting counting immediately after irradiation for one half-life, and employing a photo-peak counting efficiency of only about, 0.5%, we estimate the determination limits to be 0.025 μg for S, 0.05 μg for Cl, 0.2 μg for K, and 0.025 μg for Ca, in a 1-gram sample.

Samples analyses

Samples containing compounds with known amount of S, Cl, K and Ca were analyzed non-destructively. The A_o values (activity at the end of bombardment) were determined by following the gamma-ray photo-peaks for 5 to 6 half-lives; the intensity of the the photo-peaks were analyzed by computer code SAMPO[21] and the A_o values were obtained by decay-curve analysis using CLSQ[22] code. The amount of elements present was calculated by comparing the ratio of the A_o values of the samples to the appropriate standards. The standards used for this work were pure S, $PbCl_2$, KI, and CaF_2, having thicknesses similar to the samples. The results of these sample analyses are shown in Table V; the average deviation from the known concentrations of all samples is about 6%.

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TABLE I. Nuclear data, Q value for the reactions investigated in the present work.

Nuclear Reaction	$T_{1/2}$ of Product Nuclide(min)	Gamma-ray Energies (% Intensity)	Gamma-rays Used for Measurement (MeV)	Q Value (MeV)
$^{32}S(^3He,p)^{34m}Cl$	32.5	0.145(45%), 0.511(100%), 1.17(12%), 2.12(38%), 3.30(12%)	2.12	+6.08
$^{35}Cl(^3He,\alpha)^{34m}Cl$	32.5	"	"	+7.94
$^{39}K(^3He,2\alpha)^{34m}Cl$	32.5	"	"	-5.70
$^{37}Cl(^3He,2p)^{38}Cl$	37.1	1.60(38%), 2.17(47%)	2.17	-1.61
$^{37}Cl(^3He,2n)^{38}K$	7.7	0.511(200%), 2.170(100%)	"	-4.19
$^{39}K(^3He,\alpha)^{38}K$	"	"	"	+7.49
$^{40}Ca(^3He,\alpha p)^{38}K$	"	"	"	+0.84

TABLE II. Cross sections (in mb) for ^3He nuclear reactions on S, Cl, K, and Ca.

Average Beam Energy (MeV)	Energy Interval (MeV)	^{32}S $(^3He,p)$ ^{34m}Cl	^{35}Cl $(^3He,\alpha)$ ^{34m}Cl	^{37}Cl $(^3He,2p)$ ^{38}Cl	^{37}Cl $(^3He,2n)$ ^{38}K	^{39}K $(^3He,\alpha)$ ^{38}K	^{39}K $(^3He,2\alpha)$ ^{34m}Cl	^{40}Ca $(^3He,\alpha p)$ ^{38}K
4.2	4.6-3.8	0.25						
4.6	5.0-4.3	1.1						
5.2	5.6-4.9	3.1						
6.3	6.6-6.1	11.1						
7.5	7.8-7.3	28.4	7.9	0	0.5	6.4		
8.2	8.4-8.1			0.1	3.8	8.2		0.36
8.7	9.0-8.4	35.2				9.8	0	
8.9	9.2-8.6		10.3	2.8				
10.1	10.3-9.4	34.2			9.1	10.2		5.9
10.6	10.9-10.4		12.1	13.4	9.3	9.9		
11.2	11.5-11.0		11.6	21.8	12.9			
11.9	12.1-11.6	31.0						
12.4	12.6-12.3				18.3	7.7	0	22.4
13.8	14.0-13.6	23.1	9.5	48.0	22.8			39.1
14.8	15.0-14.7		8.5	53.1	23.0	6.9	0.5	50.5
15.4	15.6-15.3	16.2	7.4	60.5				
15.9	16.0-15.7		7.8	62.2	22.6			
16.2	16.4-16.0					6.3	3.6	62.6
17.1	17.3-17.0	12.4	7.7	70.0				
18.0	18.1-17.8	10.3	6.7	75.0	20.1	6.1	7.1	72.1
18.8	19.0-18.6	9.7						
20.0	20.2-19.9	7.2	8.1	77.7	17.2	5.8	8.2	76.6
22.0	22.1-21.8	6.7						

TABLE III. Percent ^{38}K activity produced from K, Cl, and Ca.

^3He Beam Energy (MeV)	From Elements[a]		
	K	Cl	Ca
8	90	9	1
10	51	16	33
15	10	10	80
20	7	5	88

[a]Equal weight of natural elements in a thin sample.

TABLE IV. Percent ^{34m}Cl activity produced from Cl, S, and K.

^3He Beam Energy (MeV)	From Elements[a]		
	Cl	S	K
8	16	84	0
10	19	81	0
15	24	73	3
20	30	36	34

[a]Equal weight of natural elements in a thin sample.

TABLE V. Results of ^3He activation analyses.

Sample	Element Sought	^3He Beam (MeV)	Nuclide Used	Element, mg/cm^2 Present	Found[a]	Dev. from Known (%)
S-1	S	10	^{34m}Cl	0.21	0.20	-4.7
S-2	S	10	^{34m}Cl	0.54	0.52	-3.7
S-3	S	10	^{34m}Cl	1.09	1.14	+4.5
S-4+Pb	S	15	^{34m}Cl	0.56	0.61	+8.9
S-5+Pb	S	15	^{34m}Cl	1.12	1.20	+7.0
Saran-1	Cl	15	^{38}Cl	1.20	1.13	-5.8
Saran-2	Cl	15	^{38}K	0.99	0.95	-4.0
AgCl-1	Cl	10	^{34m}Cl	0.75	0.80	+6.6
AgCl-2	Cl	15	^{38}K	0.75	0.78	+4.0
NaCl-1	Cl	15	^{38}Cl	0.21	0.20	-4.7
NaCl-2	Cl	15	^{38}K	1.08	1.06	-1.8
$PbCl_2$+Pb	Cl	15	^{38}Cl	0.10	0.09	-10
$PbCl_2$+Pb	Cl	15	^{38}K	0.10	0.09	-10
KCl-1	Cl	15	^{38}Cl	1.35	1.31	-2.9
KCl-1	K	8	^{38}K	1.48	1.41	-4.7
Mica-1	K	10	^{38}K	0.09	0.08	-11
Mica-2	K	10	^{38}K	0.07	0.06	-14
Mica-3	K	20	^{38}K	0.18	0.16	-11
CaF_2-1	Ca	20	^{38}K	0.54	0.55	+1.8
CaF_2-2	Ca	20	^{38}K	0.26	0.27	+3.8
CaF_2-3	Ca	20	^{38}K	0.13	0.14	+7.7

[a]Average of 2 measurements. Av. Dev. 6%

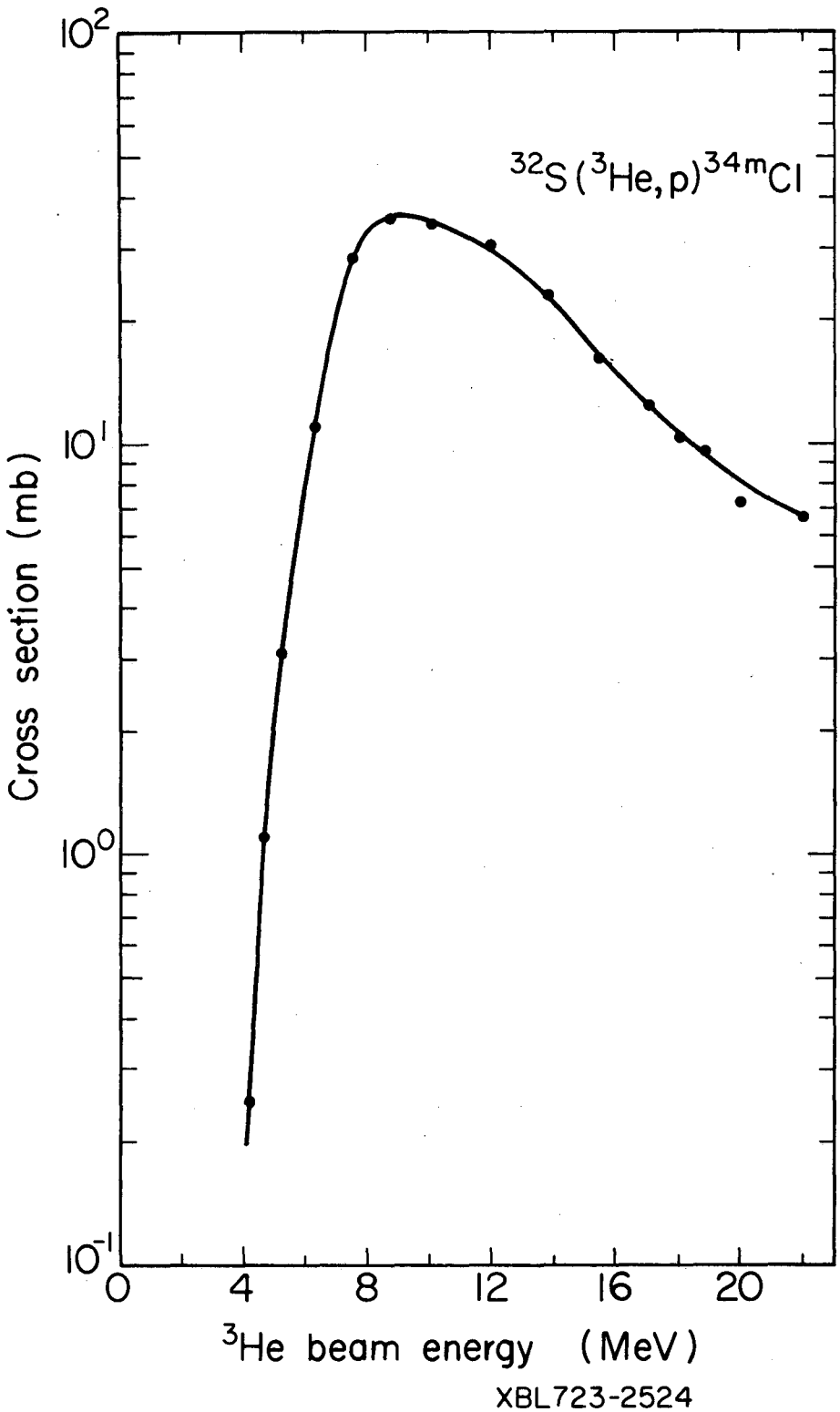

Fig. 1. Excitation function for the reaction $^{32}S(^3He,p)^{34m}Cl$.

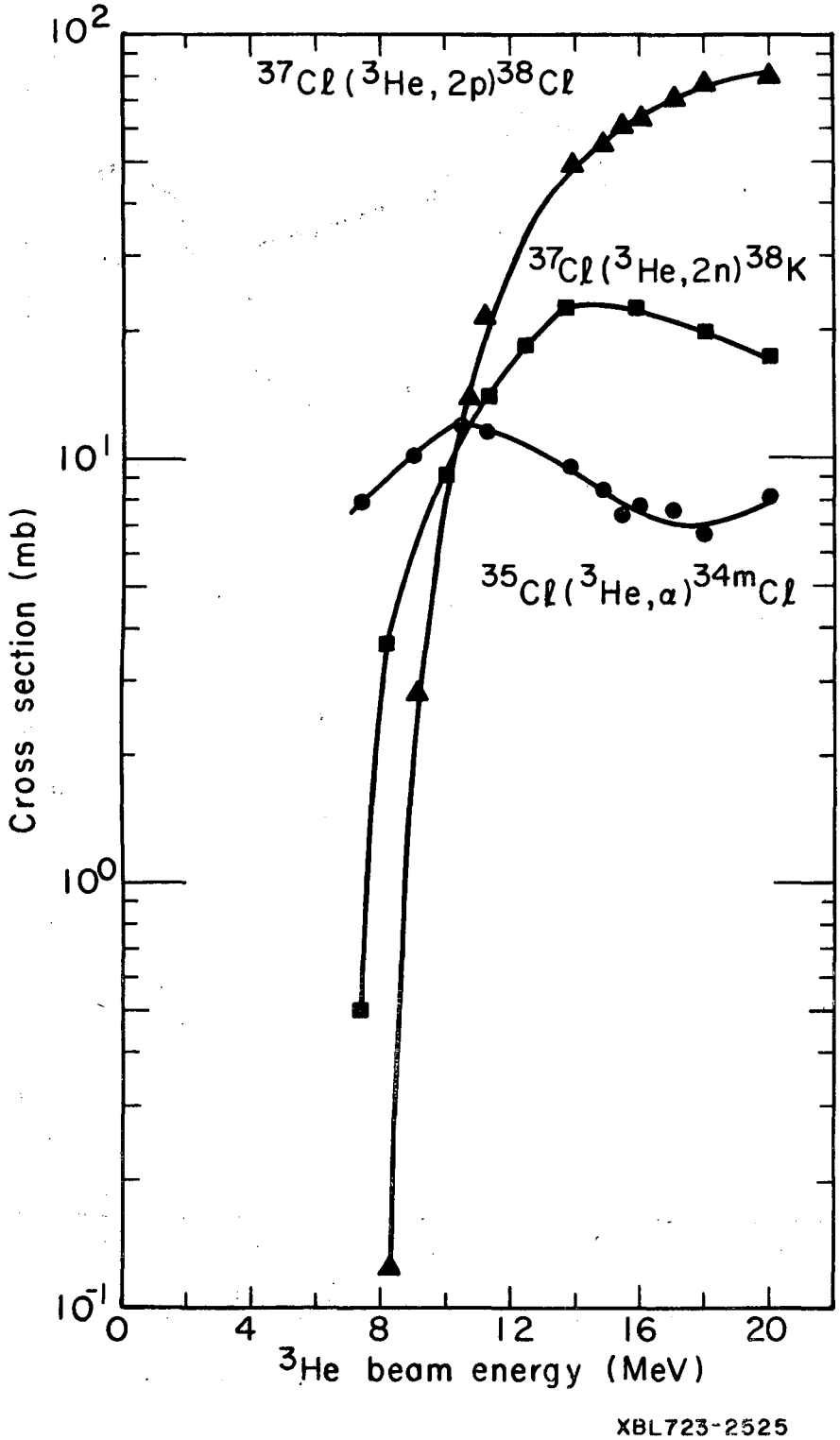

Fig. 2. Excitation functions for reactions ^{3}He + Cl.
▲ $^{37}Cl(^{3}He,2p)^{38}Cl$ $^{37}Cl(^{3}He,2n)^{38}K$
$^{35}Cl(^{3}He,\alpha)^{34m}Cl$.

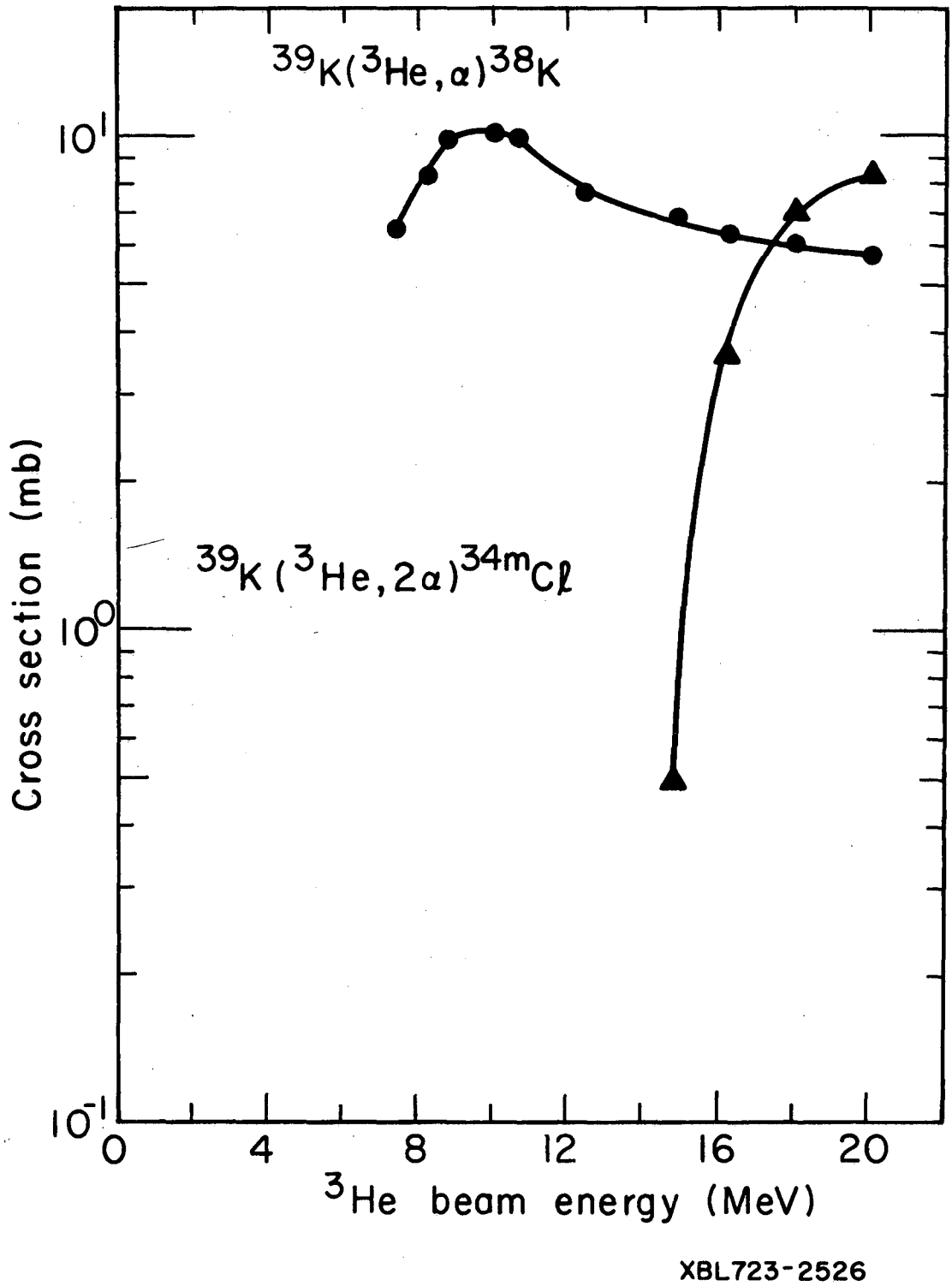

Fig. 3. Excitation functions for reactions ^{3}He + K.
$^{39}K(^{3}He,\alpha)^{38}K$ ▲ $^{39}K(^{3}He,2\alpha)^{34m}Cl$.

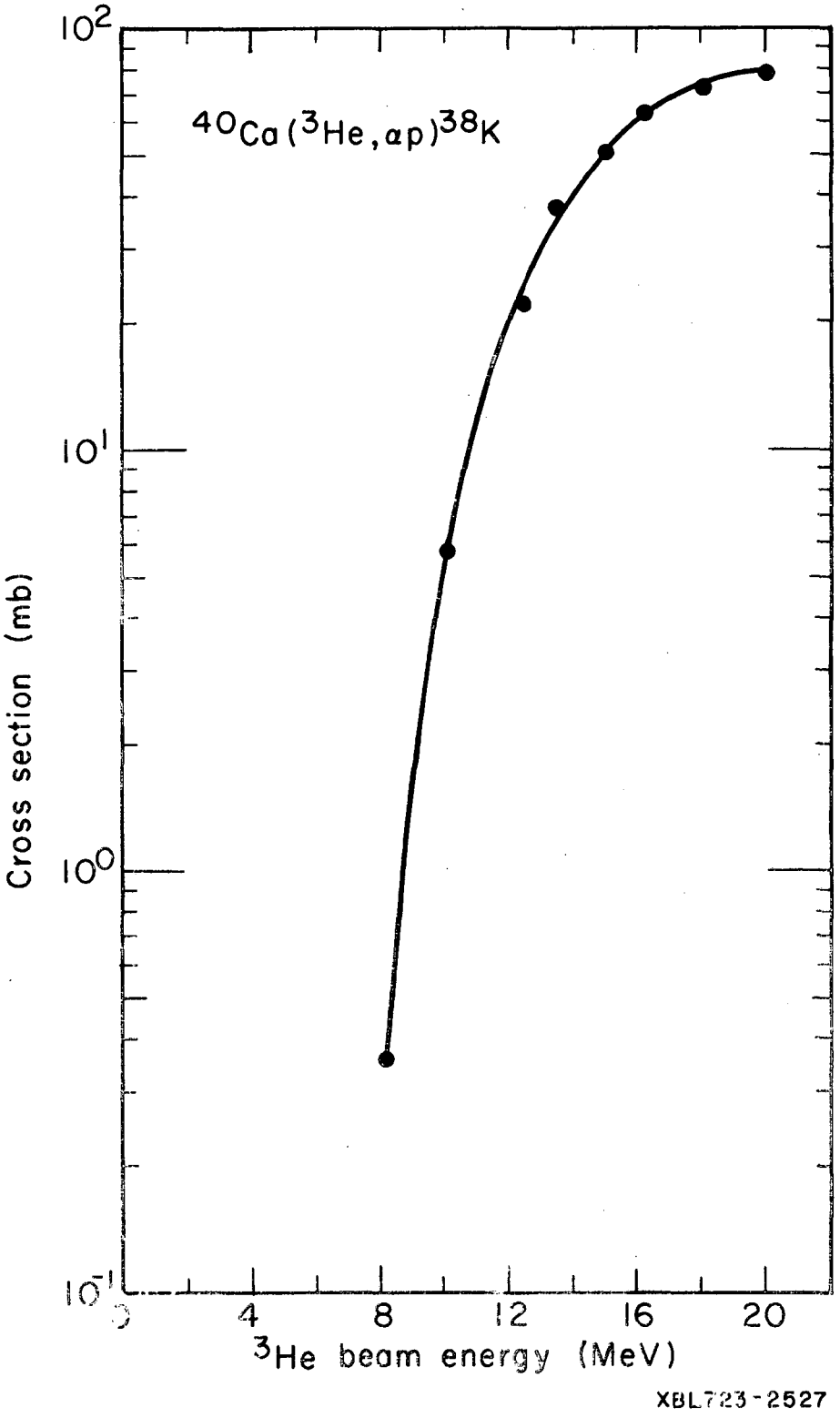

Fig. 4. Excitation function for the reaction $^{40}Ca(^3He,\alpha p)^{38}K$.

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