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SIMULTANEOUS DETERMINATION OF O^{18} AND O^{16} ISOTOPES BY He^3 ACTIVATION ANALYSIS *

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Charged particle activation analysis using accelerated $^3He^{++}$ ions provides a method for determining the isotopic content of oxygen in various materials. The method is most important for samples whose oxygen content is difficult to prepare in a form suitable for mass spectrometric analysis, or for samples which must be analyzed non-destructively. Among the former are a variety of high melting inorganic oxygen compounds and organic compounds which yield much too complicated mass spectra when analyzed directly.

The method reported here is based upon the nuclear reactions induced in oxygen during bombardment with $^3He^{++}$ ions accelerated to about 10 MeV. The reactions of interest are

(1) $^{16}O(^3He,p)^{18}F + {}^{16}O(^3He,n)^{18}Ne \xrightarrow{1.5\ sec} {}^{18}F$

(2) $^{18}O(^3He,p)^{20}F.$

The ^{18}F activity, produced from ^{16}O by the two reactions, has been reported previously.[1] ^{18}F decays with a 110-minute half life directly to the ground state of stable ^{18}O by positron emission. The cross section for total production of ^{18}F is about 260 mb at E_{He-3} = 10 MeV. ^{20}F is a negatron emitter with a half life of 11.4 sec.[2] The cross section for reaction 2 has not been previously investigated.

* This work was done under the auspices of the U.S. Atomic Energy Commission.

I. EXCITATION FUNCTION FOR THE REACTION $^{18}O(^{3}He,p)^{20}F$

The absolute cross section for the reaction $^{18}O(^{3}He,p)^{20}F$, measured at incident ^{3}He particle energies 2 to 20 MeV, is shown in Fig. 1. The beam intensity used was about 0.2 μA.

The short half life of ^{20}F precluded the use of the stacked foil technique so that each point in Fig. 1 represents a separate bombardment. The energy of the incident beam from the Berkeley HILAC was degraded from its initial value of 31.2 MeV by the appropriate thickness of aluminum foil beam degraders.[3] The target consisted of ^{18}O enriched Ta_2O_5 prepared by anodization of tantalum foil.[4] The same target was used throughout the experiment because the ^{20}F half life is sufficiently short.

The irradiation chamber is shown diagramatically in Fig. 2. The secondary electron monitor foil allowed measurement of beam intensity during sample irradiation even though the thick tantalum foil sample backing attenuated the beam current readings. This was done by obtaining a monitor foil versus Faraday cup calibration at each beam energy before introducing the sample into the chamber. The chamber was operated under a helium gas atmosphere rather than in the accelerator vacuum to minimize time loss during sample retrieval. The activated samples were transferred to the counting chamber by a gravity fall delivery track which introduced the sample for counting within 4 sec after the end of bombardment.

The ^{20}F activity was determined by following the decay of its associated 1.6-MeV γ ray using a 3 X 3 in diameter NaI(Tl) scintillator coupled to a RIDL 400-channel analyzer operated as a scalar, with scaling intervals of

2 sec. The resulting decay curves were resolved using a computer code. Fig. 3 is a typical spectrum showing the 1.6-MeV photopeak from ^{20}F decay.

II. ISOTOPIC OXYGEN ANALYSIS

The isotopic content of a number of samples was determined by measuring the ^{16}O:^{18}O ratio from the activity produced in a single bombardment. Samples consisted of H_2O, KH_2PO_4, and glycine of various atom percent ^{18}O, obtained, together with approximate analyses, from the Weizmann Institute of Science, Rehovoth, Israel.

Targets were prepared from the KH_2PO_4 and glycine by allowing the finely-ground materials to settle onto a backing foil from an ether suspension, drying, and fixing the powder with a drop of dilute polystyrene in dichloroethylene. Samples were prepared from the water by anodizing tantalum foil in a solution of the H_2O^{18} containing a few crystals of KCl.[4] Thin tantalum cover foils were placed over all targets to prevent recoil losses.

Each sample was irradiated for 10 sec at a beam energy of 10 MeV. Decay of the short-lived ^{20}F was followed for 300 sec, the samples allowed to cool for 4 hr, and then the decay of the ^{18}F followed by measuring the 511-keV γ-ray resulting from β^+ annihilation. The 4-hr cooling period was sufficient for complete decay of any activities resulting from any other reactions of C, N, and O which produce positron activity.

The ^{18}O atom percents were calculated by comparing the ratios of the A_o values of ^{18}F:^{20}F in the samples to the ratio measured for one of the Ta_2O_5 targets selected as a standard. The composition of the water from which the

standard was prepared was verified by mass spectrometric analysis. This method of treating the data removes beam-current integrator readings and sample weight determination from the calculations, leaving the counting statistical error as the major source of error. Results of several isotopic oxygen analyses are shown in Table 1; the errors shown represent maximum variations in triplicate analyses.

III. CONCLUSIONS

Helium-3 activation analysis provides a nondestructive method for determination of the isotopic content of oxygen in a wide variety of materials which, in principle, meets analytical requirements of accuracy, sensitivity, and relative freedom from interference.

The accuracy and sensitivity (at the concentrations studied in this paper) are both concerned primarily with counting statistics which, in turn, will be governed by available beam current. Both the $^{16}O(^{3}He,p)^{18}F$ and the $^{18}O(^{3}He,p)^{20}F$ reactions have maximum cross sections of hundreds of millibarns and they both produce radiation which may be detected without complication and with reasonable counting efficiency. No effort has been made to push either the sensitivity or the accuracy of the analysis to the limit, since available ^{3}He beam intensity at the Berkeley HILAC is only a fraction of that which may be produced in a small cyclotron. Limitations on sample sizes due to the small depth of penetration of the ^{3}He beam are not generally of much importance when the sample under consideration is of an isotopically-enriched material.

There is only one nuclide whose presence in a sample would produce significant interference. ^{19}F, if present, would produce both ^{18}F and ^{20}F by the reactions $^{19}F(^{3}He,\alpha)^{18}F$ and $^{19}F(^{3}He,2p)^{20}F$. The cross section for production of ^{18}F from ^{19}F is about 20 mb at E_{He-3} = 10 MeV.[5,6,7] The corresponding cross section for ^{20}F production is now being investigated and appears to be about 40 mb at E_{He-3} = 10 MeV. Should it be necessary to measure isotopic oxygen in the presence of fluorine, it might be possible to account for the ^{18}F and ^{20}F contribution from fluorine through the reaction $^{19}F(^{3}He,n)^{21}Na$. This reaction is also being currently studied at this laboratory. The ^{19}F cross sections are, however, considerably lower than the ^{16}O and the ^{18}O cross sections; furthermore, the ^{16}O may be determined via the $^{16}O(^{3}He,\alpha)^{15}O$ reaction.

A small, "desk-top", cyclotron has been devloped at the Lawrence Radiation Laboratory, Berkeley, for the purpose of acceleration of 18-MeV He^{3} ions for activation analysis and other nuclear reaction studies; details are given in Ref. 8.

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Table 1

NONDESTRUCTIVE ANALYSIS OF OXYGEN-18
BY HELIUM-3 ACTIVATION

SAMPLE	% ^{18}O Quoted*	% ^{18}O Found
Ta_2O_5	0.204	0.24 ± 0.01
Ta_2O_5	3.13	3.3 ± 0.1
Ta_2O_5	9.4	Standard
KH_2PO_4	40	44.8 ± 1.4
KH_2PO_4	8.7	7.7 ± 0.5
KH_2PO_4	1.6	1.9 ± 0.1
GLYCINE†	10	11.5 ± 1.7

*Weizmann Institute of Science

†NH_2CH_2COOH

FIGURE CAPTIONS

Fig. 1. Excitation function for the reaction $^{18}O(^{3}He,p)^{20}F$.

Fig. 2. Irradiation chamber for helium-3 activation analysis.

Fig. 3. Gamma-ray spectrum of ^{18}F and ^{20}F produced by bombardment of ^{18}O-enriched Ta_2O_5 target. ^{3}He energy = 10 MeV.

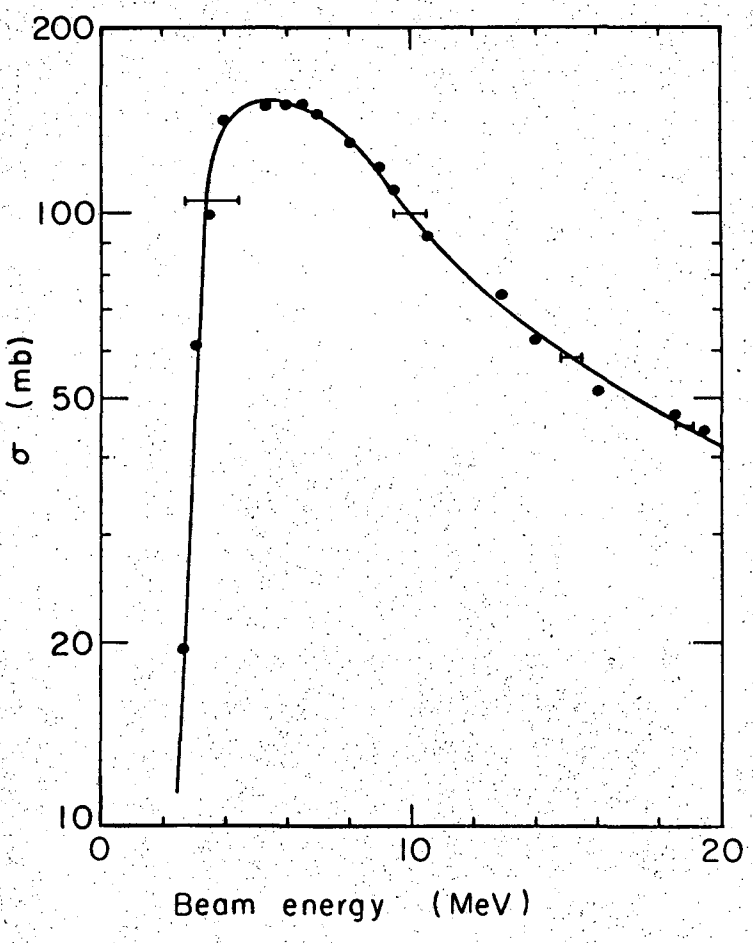

Fig. 1

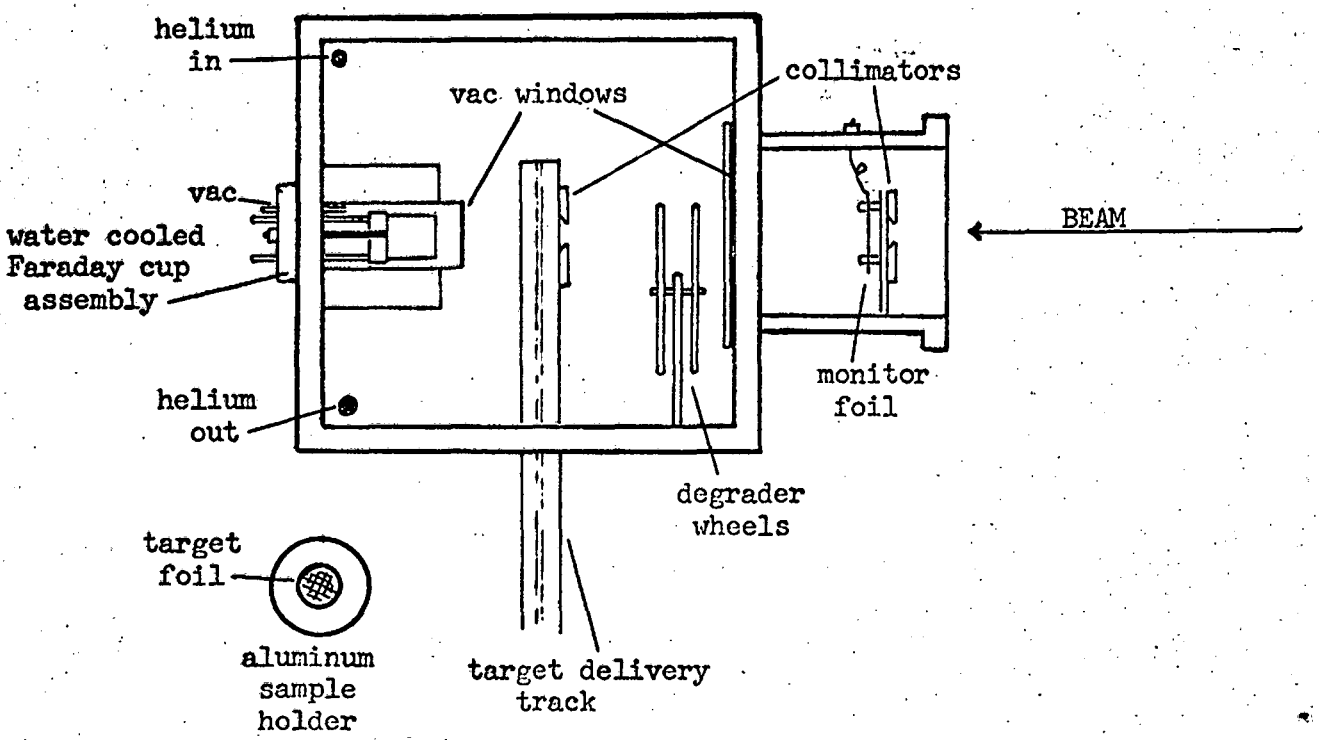
helium
in
vac windows
collimators
vac
water cooled
Faraday cup
assembly
BEAM
helium
out
monitor
foil
degrader
wheels
target
foil
aluminum
sample
holder
target delivery
track

Fig. 2

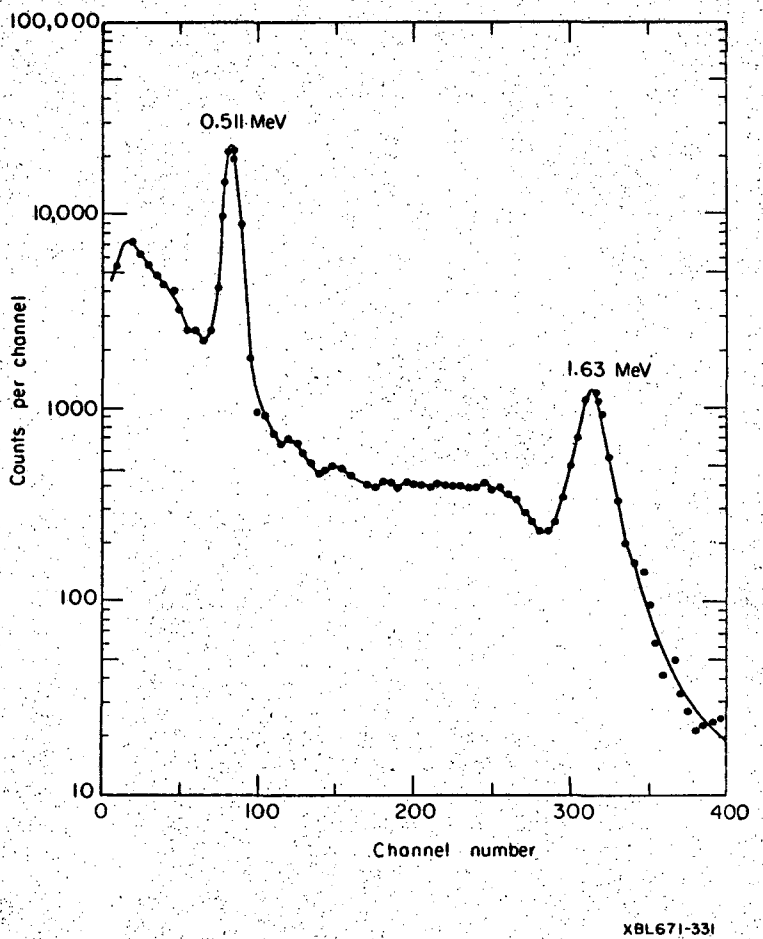
100,000
10,000
1000
100
10
Counts per channel
0.511 MeV
1.63 MeV
0
100
200
300
400
Channel number
XBL671-331

Fig. 3

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