The Determination of Carbon, Nitrogen, and Oxygen and their Stable Isotopic Tracers in Gases by Neutron Time-of-flight Spectroscopy

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ACTIVATION analysis methods have generally not been applied to some common gaseous compounds because the nuclear properties of ^{12}C, ^{1}H, ^{14}N, and ^{16}O do not readily lend themselves to thermal neutron activation, although some nuclear methods have been used for their heavier isotopes. With accelerated charged particles, nuclear reactions, particularly those leading to the emission of neutrons, occur at relatively low energies and can be used for analytical purposes. When the gaseous phase is irradiated, the major portion of the beam energy is taken up by the walls of the gas cell and damage to the gas is negligible if the gas does not decompose in contact with the heated cell wall.

After irradiation and analysis the sample may often be recovered.

The energy of an emitted neutron, E(Mev) is obtained from the time, t(nanoseconds), it takes to travel from the target cell to a detector d metres away and is given in the non-relativistic case by

$$t = 72{\cdot}3 \frac{d}{\sqrt{E}}$$

The number of neutrons emitted is proportional to the number of target nuclei presented to the beam, *i.e.*, to the path length of the beam through the gas, the partial pressure of the gas, and the number of atoms of the nuclide under investigation per molecule of the gas.

Beams of protons or deuterons in pulses of 4 nsec. duration and 400 nsec. apart were used to generate neutrons by (p,n) and (d,n) reactions in gases. Pulse-shape discrimination was used in the measuring circuit[1] to discriminate against γ-rays, but this discrimination limited detection to neutrons above 700 kev. Flight paths were about 3 metres.

With 3 Mev deuterons, ^{12}C and ^{16}O produced neutrons of 2·58 Mev and 1·22 Mev respectively measured at 30°. Within 3 minutes' irradiation at gas pressures of a few mm., sufficient counts were accumulated under each peak in the energy spectrum of the neutrons, to enable C and O to be separately determined. In analyses where the relative atomic concentrations of the elements are required, absolute calibration need not be carried out provided the relative counting efficiency is known for each peak. In the presence of nitrogen, which gave neutrons of 7·93, 2·77, 2·71, 1·76, 1·11, and 1·04 Mev from ^{14}N, the analysis was not as straightforward because the unresolved peaks at 2·77 and 2·71 Mev and at 1·11 and 1·04 Mev in the energy spectrum partly overlapped those from carbon and oxygen respectively. Correction for this interference is made from a measurement of the relative areas under each spectrum peak as obtained with natural N_2. When the sample also contained deuterium, its content was obtained from a lone peak corresponding to 5·74 Mev neutrons. Absolute sensitivities were of the order of 10^{-8} g.D and 10^{-7} g.C, O, and N, but sufficiently large samples had to be used to give a few mm. pressure in the cell; usually about 20 μg. were sufficient. The relative standard deviation was about 3%, but as the major error was in the statistics of counting, the error could be reduced by a longer irradiation.

Proton irradiation of gases labelled with the heavier stable isotopes of carbon, nitrogen, or oxygen results in neutron-energy spectra suitable for analysis, because no neutrons can be formed from the very abundant ^{12}C, ^{14}N, or ^{16}O, at proton energies of 4·5 Mev, whereas ^{13}C gives 1·38 Mev neutrons, ^{15}N 0·82 Mev, ^{17}O 0·86 Mev, and ^{18}O 1·9, 1·0, 0·86, and 0·81 Mev neutrons measured at 30°.

Neutron time-of-flight spectroscopy thus provides a nondestructive method for the determination of C, N, and O by deuteron irradiation and of the stable tracers ^{13}C, ^{15}N, ^{17}O, and ^{18}O by proton irradiation. As only a few mm. pressure is required, liquids with vapour pressure of this order can be analysed provided their vapours can withstand the high temperatures. Some typical results for C, N, and O are shown in the Table.

TABLE

Typical results for the determination of C, N, O, *and* D.

Gas	Element determined	Weight element (μg.) Present	Found	Relative error (%)
Air	N	98·43	98·01	−0·4
	O	41·78	40·07	−4·1
CO_2	C	61·28	60·67	−1·0
	O	163·4	163·7	+0·2
H_2/D_2	D	40·55	41·56	+2·5
H_2O	O	132·5	131·0	−1·1
D_2O	D	27·38	27·00	−1·4
	O	109·5	111·5	+1·8
CCl_4	C	308·4	313·2	+1·6
NH_3	N	86·3	86·8	+0·6

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[1] W. R. McMurray, P. van der Merwe, and I. J. van Heerden, *Phys. Letters*, 1965, **18**, 319.