Designation: E385 – 22

Standard Test Method for Oxygen Content Using a 14-MeV Neutron Activation and Direct-Counting Technique[1]

This standard is issued under the fixed designation E385; the number immediately following the designation indicates the year of original adoption or, in the case of revision, the year of last revision. A number in parentheses indicates the year of last reapproval. A superscript epsilon (ε) indicates an editorial change since the last revision or reapproval.

1. Scope

1.1 This test method covers the measurement of oxygen concentration in almost any matrix by using a 14-MeV neutron activation and direct-counting technique. Essentially, the same system may be used to determine oxygen concentrations ranging from under 10 µg/g to over 500 mg/g, depending on the sample size and available 14-MeV neutron fluence rates.

NOTE 1—The range of analysis may be extended by using higher neutron fluence rates, larger samples, and higher counting efficiency detectors.

1.2 This test method may be used on either solid or liquid samples, provided that they can be made to conform in size, shape, and macroscopic density during irradiation and counting to a standard sample of known oxygen content. Several variants of this method have been described in the technical literature. A monograph is available which provides a comprehensive description of the principles of activation analysis using a neutron generator (**1**).[2]

1.3 The values stated in SI units are to be regarded as standard. No other units of measurement are included in this standard.

1.4 *This standard does not purport to address all of the safety concerns, if any, associated with its use. It is the responsibility of the user of this standard to establish appropriate safety, health, and environmental practices and determine the applicability of regulatory limitations prior to use.*Specific precautions are given in Section 8.

1.5 *This international standard was developed in accordance with internationally recognized principles on standardization established in the Decision on Principles for the Development of International Standards, Guides and Recommendations issued by the World Trade Organization Technical Barriers to Trade (TBT) Committee.*

2. Referenced Documents

2.1 *ASTM Standards:*[3]

E170 Terminology Relating to Radiation Measurements and Dosimetry

E181 Test Methods for Detector Calibration and Analysis of Radionuclides

E496 Test Method for Measuring Neutron Fluence and Average Energy from $^{3}H(d,n)^{4}He$ Neutron Generators by Radioactivation Techniques

2.2 *U.S. Government Document:*

Code of Federal Regulations, Title 10, Part 20[4]

3. Terminology

3.1 *Definitions* (see also Terminology E170):

3.1.1 *accelerator*—a machine that ionizes a gas and electrically accelerates the ions onto a target. The accelerator may be based on the Cockroft-Walton, Van de Graaff, or other design types (**1**). Compact sealed-tube, mixed deuterium and tritium gas, Cockcroft-Walton neutron generators are most commonly used for 14-MeV neutron activation analysis. However, "pumped" drift-tube accelerators that use replaceable tritium-containing targets are also still in use. Reviews of operational characteristics, descriptions of accessory instrumentation, and applications of accelerators used as fast neutron generators for activation analysis are available (**2**, **3**).

3.1.2 *comparator standard*—a reference standard of known oxygen content whose specific counting rate (counts min^{-1} [mg of oxygen]$^{-1}$) may be used to quantify the oxygen content of a sample irradiated and counted under the same conditions. Often, a comparator standard is selected to have a matrix composition, physical size, density and shape very similar to the corresponding parameters of the sample to be analyzed.

3.1.3 *14-MeV neutron fluence rate*—the areal density of neutrons passing through a sample, measured in terms of neutrons cm^{-2} s^{-1}, that is produced by the fusion reaction of

[1] This test method is under the jurisdiction of ASTM Committee E10 on Nuclear Technology and Applications and is the direct responsibility of Subcommittee E10.05 on Nuclear Radiation Metrology.

Current edition approved Feb. 1, 2022. Published March 2022. Originally approved in 1969. Last previous edition approved in 2016 as E385 – 16. DOI: 10.1520/E0385-22.

[2] The boldface numbers in parentheses refer to a list of references at the end of the text.

[3] For referenced ASTM standards, visit the ASTM website, www.astm.org, or contact ASTM Customer Service at service@astm.org. For *Annual Book of ASTM Standards* volume information, refer to the standard's Document Summary page on the ASTM website.

[4] Available from the Superintendent of Documents, U.S. Government Printing Office, Washington, DC 20402.

deuterium and tritium ions accelerated to energies of typically 150 to 200 keV in a small accelerator. Fluence rate has been commonly referred to as "flux density." The total neutron fluence is the fluence rate integrated over time.

3.1.3.1 *Discussion*—The $^{3}H(d,n)^{4}He$ reaction is used to produce approximately 14.7-MeV neutrons. This reaction has a *Q*-value of + 17.586 MeV.

3.1.4 *monitor*—any type of detector or comparison reference material that can be used to produce a response proportional to the 14-MeV neutron fluence rate in the irradiation position, or to the radionuclide decay events recorded by the sample detector. A plastic pellet with a relatively high oxygen content is often used as a monitor reference in dual sample transfer systems. It is never removed from the system regardless of the characteristics of the sample to be analyzed. It is important to distinguish that the monitor, whether an independent detector or an activated reference material, is not a standard used to scale the oxygen content of the samples to be measured, but rather is used to normalize the analysis system among successive analytial passes within the procedure.

3.1.5 *multichannel pulse-height analyzer*—an instrument that receives, counts, separates, and stores, as a function of their energy, pulses from a scintillation or semi-conductor gamma-ray detector and amplifier. In the 14-MeV instrumental neutron activation analysis (INAA) determination of oxygen, the multichannel analyzer may also be used to receive and record both the BF_3 neutron detector monitor counts and the sample gamma-ray detector counts as a function of stepped time increments (**4-6**). In the latter case, operation of the analyzer in the multichannel scaler (MCS) mode, an electronic gating circuit is used to select only gamma rays within the energy range of interest.

3.1.6 *transfer system*—a system, normally pneumatic, used to transport the sample from an injection port (sometimes connected to an automatic sample changer) to the irradiation station, and then to the counting station where the activity of the sample is measured. The system may include components to ensure uniform positioning of the sample at the irradiation and counting stations.

4. Summary of Test Method

4.1 The weighed sample to be analyzed is placed in a container for automatic transfer from a sample-loading port to the 14-MeV neutron irradiation position of a particle accelerator. After irradiation for a pre-selected time, the sample is automatically returned to the counting area. A gamma-ray detector measures the high-energy gamma radiation from the radioactive decay of the ^{16}N produced by the (n,p) nuclear reaction on ^{16}O. The number of counts in a pre-selected counting interval is recorded by a gated scaler, or by a multichannel analyzer operating in either the pulse-height, or gated multiscaler modes. The number of events recorded for samples and monitor reference standard are corrected for background and normalized to identical irradiation and counting conditions. If the sample and a monitor reference sample are not irradiated simultaneously, the neutron dose received during each irradiation must be recorded, typically by use of a BF_3 neutron proportional counter. The amount of total oxygen (all chemical forms) in the sample is proportional to the corrected and normalized sample count and is quantified by use of the corrected and normalized specific activity of the comparator standard(s).

4.1.1 ^{16}N decays with a half-life of 7.13 (2)[5] s by β-emission (**7**), thus returning to ^{16}O. From Ref (**7**), 67.0 (6) % of the decays are accompanied by 6.12863 (4) MeV gamma rays, 4.9 (4) % by 7.11515 (14) MeV gamma rays, and 0.82 (6) % by 2.7415 (5) MeV gamma rays. Other lower intensity gamma rays are also observed. About 28 % of the beta transitions are directly to the ground state of ^{16}O. Useful elemental data including calculated sensitivities and reaction cross-sections for (14-MeV INAA) are provided in Refs (**3**) and (**8**). (See also Test Methods E181.)

5. Significance and Use

5.1 The conventional determination of oxygen content in liquid or solid samples is a relatively difficult chemical procedure. It is slow and usually of limited sensitivity. The 14-MeV neutron activation and direct counting technique provides a rapid, highly sensitive, nondestructive procedure for oxygen determination in a wide range of matrices. This test method is independent of the chemical form of the oxygen.

5.2 This test method can be used for quality and process control in the metals, coal, and petroleum industries, and for research purposes in a broad spectrum of applications.

6. Interferences

6.1 Because of the high energy of the gamma rays emitted in the decay of ^{16}N, there are very few elements that will produce interfering radiations; nevertheless, caution should be exercised. ^{19}F, for example, will undergo an (n,α) reaction to produce ^{16}N, the same indicator radionuclide produced from oxygen. Because the cross section for the $^{19}F(n,\alpha)^{16}N$ reaction is approximately one-half that of the $^{16}O(n,p)^{16}N$ reaction, a correction must be made if fluorine is present in an amount comparable to the statistical uncertainty in the oxygen determination. Another possible interfering reaction may arise from the presence of boron. ^{11}B will undergo an (n,p) reaction to produce ^{11}Be. This isotope decays with a half-life of 13.76 (7) s, and beta decays to populate excited states in ^{11}B which subsequently decay by emitting several high-energy gamma rays with energies up to 7.97473 MeV (**9**, **10**). In addition, there is Bremsstrahlung radiation produced by the high energy beta particles emitted by ^{11}Be. These radiations can interfere with the oxygen determination if the oxygen content does not exceed 1 % of the boron present.

6.2 Another possible elemental interference can arise from the presence of fissionable materials such as thorium, uranium, and plutonium. Many short-lived fission products emit high-energy gamma rays capable of interfering with those from ^{16}N.

NOTE 2—Argon produces an interferent, ^{40}Cl, by the $^{40}Ar(n,p)^{40}Cl$ reaction. Therefore, argon should not be used for the inert atmosphere during sample preparation for oxygen analysis. ^{40}Cl ($t_{½}$ = 1.35 (3) m) has

[5] The value of uncertainty, in parentheses, refers to the corresponding last digits, thus 14.958 (2) corresponds to 14.958 ± 0.002.