Designation: D4239 – 17

Standard Test Method for Sulfur in the Analysis Sample of Coal and Coke Using High-Temperature Tube Furnace Combustion[1]

This standard is issued under the fixed designation D4239; 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.

This standard has been approved for use by agencies of the U.S. Department of Defense.

1. Scope*

1.1 This test method covers the determination of sulfur in samples of coal or coke by high-temperature tube furnace combustion.

1.1.1 Two analysis methods are described.

1.2 When automated equipment is used, either method can be classified as an instrumental method.

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 All percentages are percent mass fractions unless otherwise noted.

1.5 *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 and health practices and determine the applicability of regulatory limitations prior to use.*

1.6 *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:*[2]

D346 Practice for Collection and Preparation of Coke Samples for Laboratory Analysis

D2013 Practice for Preparing Coal Samples for Analysis

D3173 Test Method for Moisture in the Analysis Sample of Coal and Coke

D3176 Practice for Ultimate Analysis of Coal and Coke

D3180 Practice for Calculating Coal and Coke Analyses from As-Determined to Different Bases

D7448 Practice for Establishing the Competence of Laboratories Using ASTM Procedures in the Sampling and Analysis of Coal and Coke

D7582 Test Methods for Proximate Analysis of Coal and Coke by Macro Thermogravimetric Analysis

E691 Practice for Conducting an Interlaboratory Study to Determine the Precision of a Test Method

2.2 *ISO Standard:*[3]

ISO 11722 Solid Mineral Fuels-Hard Coal – Determination of Moisture in the general analysis test sample by drying in nitrogen

3. Summary of Test Method

3.1 *Combustion Method A (1350°C)*—A weighed test portion of sample is burned in a tube furnace at a minimum combustion tube operating temperature of 1350°C in a stream of oxygen. During combustion at temperatures above 1350 °C, the sulfur and sulfur compounds contained in the sample are decomposed and oxidized almost exclusively to gaseous sulfur dioxide, SO_2. Moisture and particulates are removed from the gas by filters. The gas stream is passed through a cell in which sulfur dioxide is measured by an infrared (IR) absorption detector. Sulfur dioxide absorbs IR energy at a precise wavelength within the IR spectrum. Energy is absorbed as the gas passes through the cell body in which the IR energy is being transmitted: thus, at the detector, less energy is received. All other IR energy is eliminated from reaching the detector by a precise wavelength filter. Thus, the absorption of IR energy can be attributed only to sulfur dioxide whose concentration is proportional to the change in energy at the detector. One cell is used as both a reference and a measurement chamber. Total sulfur as sulfur dioxide is detected on a continuous basis.

3.1.1 One procedure for Method A uses coal or coke reference materials to calibrate the sulfur analyzer. A second

[1] This test method is under the jurisdiction of ASTM Committee D05 on Coal and Coke and is the direct responsibility of Subcommittee D05.21 on Methods of Analysis.

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[2] 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.

[3] Available from International Organization for Standardization (ISO), 1, ch. de la Voie-Creuse, Case postale 56, CH-1211, Geneva 20, Switzerland, http://www.iso.ch.

***A Summary of Changes section appears at the end of this standard**

procedure for Method A uses a pure substance, BBOT, to calibrate the sulfur analyzer.

3.2 *Combustion Method B (1150°C)*—A weighed test portion of sample is burned in a quartz combustion tube in a stream of oxygen with an equal or excess weight of tungsten trioxide (WO_3). Sulfur is oxidized during the reaction of the sample and WO_3. The tube furnace is operated at a minimum combustion tube operating temperature of 1150°C and tin (Sn) sample boats are utilized. Moisture and particulates are removed from the combustion gas by filters. The gas stream is then passed through a cell in which sulfur dioxide is measured by an infrared (IR) absorption detector. Sulfur dioxide absorbs IR energy at a precise wavelength within the IR spectrum. Energy is absorbed as the gas passes through the cell body in which the IR energy is being transmitted: thus, at the detector, less energy is received. All other IR energy is eliminated from reaching the detector by a precise wavelength filter. Thus, the absorption of IR energy can be attributed only to sulfur dioxide whose concentration is proportional to the change in energy at the detector. One cell is used as both a reference and a measurement chamber. Total sulfur as sulfur dioxide is detected on a continuous basis.

4. Significance and Use

4.1 Sulfur is part of the ultimate analysis of coal and coke.

4.2 Results of the sulfur analysis are used for evaluation of coal preparation and cleaning, evaluation of potential sulfur emissions from coal and coke combustion or conversion processes, and evaluation of coal and coke quality in relation to contract specifications, as well as for scientific purposes.

4.3 The competency of laboratories with respect to use of this standard can be established through reference to Practice D7448.

5. Sample

5.1 Pulverize the sample to pass No. 60 (250-µm) sieve and mix thoroughly in accordance with Practice D2013 or Practice D346.

5.2 Analyze a separate portion of the analysis sample for moisture content in accordance with Test Method D3173, or D7582 or ISO 11722 for calculations to other than the as-determined basis.

5.3 Procedures for calculating as-determined sulfur values obtained from the analysis sample to other bases are described in Practices D3176 and D3180.

6. Apparatus

Combustion Method A (1350°C)

6.1 *Measurement Apparatus*—Equipped to combust the sample as described in 3.1 (See Fig. 1).

6.2 *Tube Furnace*—Capable of heating the hot zone or outer surface of the combustion tube, or both (6.3) to at least 1350°C. It is normally heated electrically using resistance rods, a resistance wire, or molybdenum disilicide elements. Specific dimensions can vary with manufacturer's design.

6.3 *Combustion Tube*—Made of mullite, porcelain, or zircon with provisions for routing the gases produced by combustion through the infrared cell. The tube may have a boat stop made of reticulated ceramics heated to 1350°C that serves to complete the combustion of sulfur containing materials.

6.4 *Sample Combustion Boats,* made of iron-free material and of a convenient size suitable for the dimensions of the combustion tube.

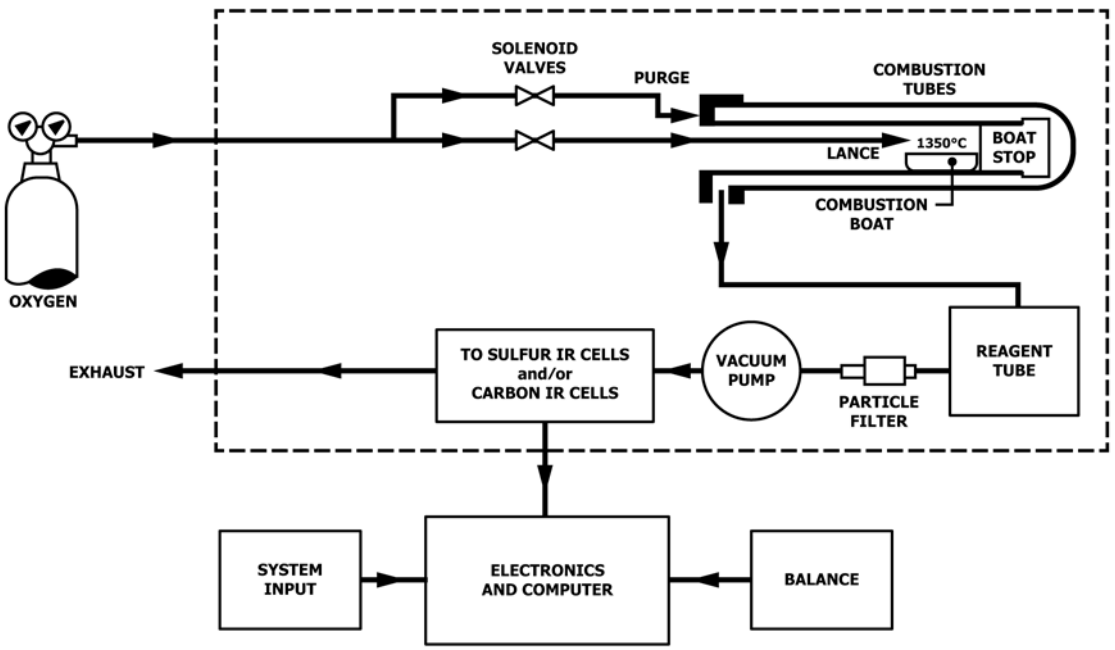

FIG. 1 Apparatus for the Determination of Sulfur by the Infrared Detection, Method A

6.5 *Boat Puller*—Where required, a rod of a heat-resistant material with a bent or disk end to insert and remove boats from the combustion tube.

6.6 *Balance*—A stand-alone balance or a balance integrated with the instrument, with a resolution of at least 0.3% relative of the test portion mass.

Combustion Method B (1150°C)

6.7 *Measurement Apparatus*—Equipped to combust the sample as described in 3.2 (See Fig. 2)

6.8 *Tube Furnace*—Capable of heating the hot zone or outer surface of the combustion tube, or both (6.9) to at least 1150°C. It is normally heated electrically using resistance wire. Specific dimensions can vary with manufacturer's design.

6.9 *Combustion Tube*—Made of quartz with provisions for routing the gases produced by combustion through the infrared cell.

6.10 *Sample Combustion Boat*—Made of an iron-free tin material and of a convenient size suitable for the dimensions of the combustion tube.

7. Reagents

7.1 *Purity of Reagents*—Use reagent grade chemicals in all tests. Unless otherwise indicated, it is intended that all reagents conform to the specifications of the Committee on Available Reagents of the American Chemical Society, where such specifications are available.[4] Other grades can be used, provided it is first ascertained the reagent is of sufficiently high purity to permit its use without lessening the accuracy of the determination.

7.2 *Magnesium Perchlorate*—(**Warning—**Magnesium perchlorate is a strong oxidizing agent. Do not regenerate the absorbent. Do not allow contact with organic materials or reducing agents.)

7.3 *Oxygen, 99.5 % Pure*—Compressed gas contained in a cylinder equipped with a suitable pressure regulator and a needle valve to control gas flow. (**Warning—**Pure oxygen vigorously accelerates combustion. Verify all regulators, lines, and valves are free of grease and oil.)

7.4 *Reference Materials, Reference Material (RM)*—that are coal(s) or coke(s) prepared by a national metrology body. Other materials that are coal(s) or coke(s) with documented traceability to reference material (CRM) coal(s) or coke(s) prepared by a national metrology body can also be used. Only use material(s) with an assigned value and assigned uncertainty for sulfur. The uncertainty expressed as the confidence interval of the assigned value shall be less than the repeatability specified in the appropriate section on Precision and Bias of this test method.

7.4.1 To minimize problems with instrument calibration or calibration verification mix all reference material before removing the test portion from the container. Do not use the reference material for calibration or calibration verification when less than 2 g remain in the container. The remaining material can be used for instrument conditioning.

7.5 *BBOT (2,5-di(5-tert-butylbenzoxazol-2-yl)thiophene, $C_{26}H_{26}N_2O_2S$)*—A pure substance and certified reference material for sulfur (7.47 % sulfur).

7.6 *Tungsten Oxide (WO_3)*—A combustion promoter and a fluxing agent. (**Warning—**Tungsten Oxide is a strong oxidizing agent.)

8. Procedure

8.1 *Instrument Preparation*—Perform apparatus set up system checks in accordance with manufacturer's instructions.

8.1.1 *Balance Calibration*—Calibrate the instrument balance in accordance with manufacturer's instructions.

8.2 *Calibration of the Infrared Detection System*—If the instrument has been previously calibrated in accordance with the section on instrument calibration, proceed to the Analysis Procedure, otherwise carry out a calibration as specified in the following section.

8.2.1 *Calibration with Coal and Coke Certified Reference Materials*—Select reference materials (7.4), in the range of the samples to be analyzed. Use at least three such reference

[4] *Reagent Chemicals, American Chemical Society Specifications* , American Chemical Society, Washington, DC. For suggestions on the testing of reagents not listed by the American Chemical Society, see *Analar Standards for Laboratory Chemicals*, BDH Ltd., Poole, Dorset, U.K., and the *United States Pharmacopeia and National Formulary*, U.S. Pharmacopeial Convention, Inc. (USPC), Rockville, MD.

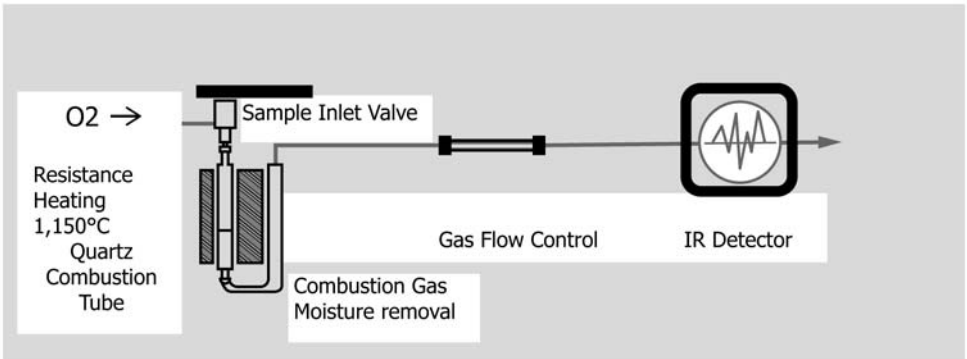

FIG. 2 Apparatus for the Determination of Sulfur by the Infrared Detection, Method B

materials, for each range of sulfur values to be tested. Select one reference material containing at least as much sulfur as the highest level of sulfur expected. Select two additional reference materials, one approximately at the mid-point of the range and one below the lowest level of sulfur expected.

8.2.1.1 Use a mass of material recommended by the apparatus manufacturer to carry out a minimum of three determinations to condition the equipment before calibration. Use a material with a sulfur value near the mid point of the expected calibration range.

8.2.1.2 For each reference coal or coke employed for calibration, use the as-determined sulfur value previously calculated from the certified dry-basis sulfur value and residual moisture determined using either Test Methods D3173 or D7582 or ISO 11722. Use a mass of material and the calibration procedure recommended by the apparatus manufacturer. Weigh in accordance with section 6.6 and evenly spread the test portion of the reference material into the sample combustion boat (6.4). Position the sample in the hot zone of the furnace until the instrument returns to baseline as indicated according to settings recommended by the manufacturer. If the analysis time exceeds the maximum analysis time recommended by the manufacturer take corrective action as recommended by the manufacturer.

8.2.2 *Calibration with BBOT*—To meet the precision requirements of this method, six calibration points are required for a linear fit and eight calibration points are required for a nonlinear fit. A calibration point consists of a determination on a single test portion of calibration material. Select test portions of the calibrant that have at least as much sulfur as the highest level of sulfur expected, test portions of the calibrant that have as much sulfur as the lowest level of sulfur expected and test portions spread evenly in between the highest and lowest levels of sulfur.

8.2.2.1 The mass of the calibrant needed can be calculated using the following equation: (Note 1).

$$M_C = \frac{(M_T \times S_{AD})}{S_C} \quad (1)$$

Where

M_C = Mass of calibrant
M_T = Mass normally used for test samples
S_{AD} = Percent sulfur (as-determined) in the test sample
S_C = Percent sulfur in the pure substance calibrant

NOTE 1—In the interlaboratory study that yielded the data for the precision statement for this method, the mass of BBOT used for calibration ranged from about 15 mg to over 80 mg. Some analyzers may use larger amounts.

8.2.3 *Calibration Verification*—Carry out a minimum of three determinations to condition the equipment before calibration verification (see 8.2.1.1). Verify the instrument calibration prior to analyzing test samples, upon completion of all test samples and as needed to meet quality control requirements. Analyze a test portion of reference material(s) (7.4) using the apparatus conditions employed for instrument calibration (8.2.1). Use a mass that does not exceed the maximum mass used for instrument calibration and with a sulfur value within the range of the instrument calibration. If the value determined for each reference material employed for verification is not within the specified uncertainty for the assigned sulfur value repeat the instrument calibration in accordance with 8.2.1. Repeat all samples analyzed since the last successful calibration verification.

8.3 *Combustion Method A (1350°C)*—Set up the apparatus (see 8.1) and verify the calibration (see 8.2.3).

8.3.1 Raise the furnace temperature as recommended by the manufacturer to at least 1350°C. Weigh a mass of the sample in accordance with section 6.6 and not exceeding the maximum mass of reference material(s) used for calibration. Analyze the test samples using the apparatus conditions employed for calibration (8.2.1).

8.3.2 When the analysis is complete, the instrument indicates the sulfur value.

8.4 *Combustion Method B (1150°C)*—Set up the apparatus (see 8.1) and verify the calibration (see 8.2.3)

8.4.1 Raise the furnace temperature as recommended by the manufacturer to at least 1150°C.

8.4.2 Weigh to the nearest 1 mg and evenly spread into a combustion boat (6.10) a portion of tungsten oxide equal to the target weight, typically 100 mg, of the sample analysis aliquot.

8.4.3 While evenly spreading the sample material into the same combustion boat, weigh in accordance with section 6.6 a sample aliquot equal (± 10 mg) to the weight of tungsten oxide.

8.4.4 For high-rank (Bituminous and Coke) materials add additional tungsten oxide as required to ensure that its weight is 10 mg (± 5 mg) in excess of the sample aliquot weight measured.

8.4.5 For low-rank (Sub-bituminous and lignite and high ash) test samples or any material of unknown BTU content, add additional tungsten oxide as required to ensure that its weight is at least double (± 10 mg) the sample aliquot weight measured.

8.4.6 For test sample materials, do not exceed the maximum mass of reference material(s) used for calibration. Analyze the test samples using the apparatus conditions employed for calibration.

8.4.7 Follow the manufacturer's recommended combustion sample boat handling procedures to position the sample into the hot zone of the furnace and start the analysis.

8.4.8 When the analysis is complete, the instrument indicates the sulfur value.

9. Calculation to Other Bases

9.1 The percent sulfur value obtained is on an as-determined basis.

9.2 Procedures for converting the as-determined value to other bases are described in Practices D3176 and D3180.

10. Report

10.1 Report the following information:

10.1.1 Mass percent sulfur, as determined, dry, or other bases.

10.1.2 Method used; Results were obtained according to Test Method D4239, (Method A, and calibrants, or Method B).

11. Precision and Bias

11.1 *Precision-250 µm (No. 60) Samples*—The precision of this method for the determination of Sulfur in the 60 mesh sample of coal and coke is shown in Table 1 and Table 2 (Method A)[5] and Table 3 (Method B)[6]. The precision is characterized by repeatability (Sr, r) and reproducibility (S_R, R) and is described in Table A1.1, Table A1.2, Table A1.3 and Table A1.4 in Annex A1.

11.1.1 *Repeatability Limit (r)*—the value below which the absolute difference between two test results calculated to a dry basis (Practice D3180) of separate and consecutive test determinations, carried out on the same sample in the same laboratory by the same operator using the same apparatus on samples taken at random from a single quantity of homogeneous material, may be expected to occur with a probability of approximately 95 %.

11.1.2 *Reproducibility Limit (R)*—the value below which the absolute difference between two test results calculated to a dry basis (Practice D3180) carried out in different laboratories using samples taken at random from a single quantity of material that is as homogeneous as possible, may be expected to occur with a probability of approximately 95 %.

11.2 *Bias*—Bias is minimized when reference material(s) are employed to calibrate the instrument.

[5] An interlaboratory study, designed consistent with ASTM Practice E691, was conducted in 1999. Twelve laboratories participated in this study. Supporting data have been filed at ASTM International Headquarters and may be obtained by requesting Research Report RR:D05-1020. In addition, an interlaboratory study, designed consistent with ASTM Practice E691, was conducted in 2012. Eight laboratories participated in this study. Supporting data have been filed at ASTM International Headquarters and may be obtained by requesting Research Report RR:D05-1042.

[6] Supporting data have been filed at ASTM International Headquarters and may be obtained by requesting Research Report RR:D05-1041.

TABLE 1 Method A (Calibration with Coal Reference Materials)—Repeatability and Reproducibility for Sulfur in 250 µm (No.60) Coal

	Range, %	Repeatability Limit (r)	Reproducibility Limit (R)
Coal	0.28–5.61	$0.02 + 0.03\bar{x}^A$	$0.02 + 0.09\bar{x}^A$

[A] Where $\bar{x}$ is the average of two single test results.

TABLE 2 Method A (Calibration with BBOT)—Repeatability and Reproducibility for Sulfur in 250 µm (No.60) Coal and Coke

	Range, %	Repeatability Limit, (r)	Reproducibility Limit (R)
Coal	0.37–5.48	$0.053 + 0.019\ \bar{x}^A$	$0.125 + 0.053\ \bar{x}^A$
Coke	0.6–6.71	$0.051 + 0.01\ \bar{x}^A$	$0.089 + 0.017\ \bar{x}^A$

[A] Where $\bar{x}$ is the average of two single test results.

TABLE 3 Method B-Repeatability and Reproducibility for Sulfur in 250 µm (No.60) Coal and Coke

	Range, %	Repeatability Limit (r)	Reproducibility Limit (R)
Coal and Coke[A]	0.38–5.6	$0.04 + 0.05\bar{x}^B$	$0.07 + 0.07\bar{x}^B$

[A] This precision statement is applicable to coal, metallurgical coke and petroleum coke.
[B] Where $\bar{x}$ is the average of two single test results.

ANNEX

A1. PRECISION STATISTICS

A1.1 The precision of this test method, characterized by repeatability (S_r, r) and reproducibility (S_R, R) has been determined for the following materials as listed in Table A1.1, Table A1.2, Table A1.3 and Table A1.4.

TABLE A1.1 Method A (Calibration with Coal and Coke Reference Materials)—Repeatability (S_r, r) and Reproducibility (S_R, R) Parameters used for Calculation of Precision Statement of 250 µm (No.60) Coal

Material	Rank	Average	S_r	S_R	r	R
91–2	lvb	0.38775	0.011373	0.0248	0.031822	0.06939
91–1	hvAb	1.24325	0.014004	0.030572	0.039183	0.08554
91–5	hvAb	3.083	0.028888	0.060704	0.080829	0.16985
89–4	hvCb	5.6125	0.080452	0.209299	0.225106	0.58562
91–4	hvCb	0.27725	0.018384	0.025722	0.051439	0.07197
90–1	ligA	1.442	0.015066	0.044989	0.042155	0.12588
89–7	subA	0.75475	0.019184	0.028152	0.053677	0.07877
91–6	subA	0.46825	0.023578	0.021137	0.06597	0.05914
89–6	subC	0.5235	0.016595	0.023399	0.046433	0.06547
Lignite	lignite	0.666	0.021000	0.0415	0.058622	0.11605

TABLE A1.2 Method A (Calibration with BBOT)—Repeatability (S_r, r) and Reproducibility (S_R, R) Parameters used for Calculation of Precision Statement of 250 µm (No.60) Coal[A]

Material	Rank	Average	S_r	S_R	r	R
89–2	subB	0.3728	0.0193	0.0534	0.0541	0.1496
89–4	hvCb	5.4838	0.0598	0.1600	0.1675	0.4481
89–6	ligA	0.4670	0.0242	0.0657	0.0677	0.1839
89–7	subA	0.7204	0.0297	0.0723	0.0831	0.2023
89–8	hvBb	1.3698	0.0286	0.0453	0.0800	0.1270
89–9	hvAb	4.1013	0.0402	0.1035	0.1125	0.2899
90–1	subB	1.3696	0.0295	0.1176	0.0827	0.3292
91–2	lvb	0.3930	0.0192	0.0331	0.0536	0.0926
91–5	hvAb	3.0251	0.0385	0.1035	0.1079	0.2899
NIST 2692c	lvb	1.0828	0.0226	0.0410	0.0631	0.1148

[A]Supporting data have been filed at ASTM International Headquarters and may be obtained by requesting Research Report RR:D05-1042. Contact ASTM Customer Service at service@astm.org.

TABLE A1.3 Method A-Repeatability (S_r, r) and Reproducibility (S_R, R) Parameters used for Calculation of Precision Statement of 250 µm (No.60) Coke[A]

Material	Coke Type	Average	S_r	S_R	r	R
Foundry	Met Coke	0.6039	0.0194	0.0360	0.0544	0.1009
Furnace	Met Coke	0.7969	0.0245	0.0437	0.0687	0.1224
DOF	Met Coke	0.6320	0.0148	0.0280	0.0415	0.0785
2017	Calcined Pet Coke	3.5154	0.0310	0.0568	0.0867	0.1590
3050	Green Pet Coke	4.9786	0.0350	0.0739	0.0979	0.2070
4070	Calcined Pet Coke	6.7059	0.0437	0.0630	0.1223	0.1764
5018	Green Pet Coke	6.0884	0.0356	0.0705	0.0998	0.1975
7009	Met Coke	0.7136	0.0142	0.0326	0.0396	0.0911
NIST 2718a	Green Pet Coke	4.7735	0.0409	0.0630	0.1145	0.1764
NIST 2776	Met Coke	0.8478	0.0306	0.0367	0.0857	0.1027

[A]Supporting data have been filed at ASTM International Headquarters and may be obtained by requesting Research Report RR:D05-1042. Contact ASTM Customer Service at service@astm.org.

TABLE A1.4 Method B-Repeatability (S_r, r) and Reproducibility (S_R, R) Parameters used for Calculation of Precision Statement of 250 µm (No.60) Coal and Coke[B]

Material	Average	S	S_r	S_R	r	R
Coal Bank 91-2	0.378833	0.013331	0.008134	0.015078	0.022776	0.042218
NIST 2693 Coal	0.4472	0.010439	0.014004	0.016002	0.039211	0.044804
NIST 2682b Coal	0.482708	0.016355	0.01825	0.022744	0.051101	0.063683
Lignite	1.16275	0.017295	0.060945	0.060945	0.170647	0.170647
NIST 2776 Coke	0.793708	0.00889	0.041466	0.041466	0.116106	0.116106
Metallurgical Coke	0.67025	0.013831	0.018789	0.021356	0.052609	0.059796
NIST 2719 Calcined Petroleum Coke	0.841556	0.015884	0.018552	0.022593	0.051947	0.06326
Coal Bank 91-1	1.213667	0.048008	0.027923	0.053754	0.078184	0.150512
Coal Bank 91-5	3.026083	0.070334	0.059562	0.087222	0.166774	0.244221
Coal Bank 89-4	5.630917	0.091162	0.124971	0.141506	0.349919	0.396216
AR 1713 Lignite	1.272167	0.117201	0.066552	0.130606	0.186346	0.365696
AR 2720 Green Petroleum Coke[A]						
AR 744 Calcined Petroleum Coke	2.383208	0.090416	0.076055	0.111863	0.212955	0.313216
NIST 2718 Green Petroleum Coke	4.563708	0.107464	0.081895	0.128758	0.229306	0.360521

[A] One of the Green Petroleum Coke samples used in this study was excluded from the statistics due to inconsistent results across all participating laboratories.
[B] Supporting data have been filed at ASTM International Headquarters and may be obtained by requesting Research Report RR:D05-1041. Contact ASTM Customer Service at service@astm.org.

SUMMARY OF CHANGES

Committee D05 has identified the location of selected changes to this standard since the last issue (D4239–14) that may impact the use of this standard. (Approved May 15, 2017.)

(1) Precision of the balance in section 6.6 was revised.

(2) Subsections 8.2.1.2, 8.3.1, and 8.4.3 were revised.