



# Standard Practice for Using the Fricke Dosimetry System<sup>1</sup>

This standard is issued under the fixed designation E1026; 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 ( $\epsilon$ ) indicates an editorial change since the last revision or reapproval.

## 1. Scope

1.1 This practice covers the procedures for preparation, testing and using the acidic aqueous ferrous ammonium sulfate solution dosimetry system to measure absorbed dose to water when exposed to ionizing radiation. The system consists of a dosimeter and appropriate analytical instrumentation. The system will be referred to as the Fricke dosimetry system. The Fricke dosimetry system may be used as either a reference standard dosimetry system or a routine dosimetry system.

1.2 This practice is one of a set of standards that provides recommendations for properly implementing dosimetry in radiation processing, and describes a means of achieving compliance with the requirements of Practice E2628 for the Fricke dosimetry system. It is intended to be read in conjunction with Practice E2628.

1.3 The practice describes the spectrophotometric analysis procedures for the Fricke dosimetry system.

1.4 This practice applies only to gamma radiation, X-radiation (bremsstrahlung), and high-energy electrons.

1.5 This practice applies provided the following are satisfied:

1.5.1 The absorbed dose range shall be from 20 to 400 Gy (1).<sup>2</sup>

1.5.2 The absorbed-dose rate does not exceed  $10^6$  Gy·s<sup>-1</sup> (2).

1.5.3 For radioisotope gamma sources, the initial photon energy is greater than 0.6 MeV. For X-radiation (bremsstrahlung), the initial energy of the electrons used to produce the photons is equal to or greater than 2 MeV. For electron beams, the initial electron energy is greater than 8 MeV.

NOTE 1—The lower energy limits given are appropriate for a cylindrical dosimeter ampoule of 12 mm diameter. Corrections for displacement effects and dose gradient across the ampoule may be required for electron

beams (3). The Fricke dosimetry system may be used at lower energies by employing thinner (in the beam direction) dosimeter containers (see ICRU Report 35).

1.5.4 The irradiation temperature of the dosimeter should be within the range of 10 to 60°C.

1.6 *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.*

## 2. Referenced Documents

### 2.1 ASTM Standards:<sup>3</sup>

C912 Practice for Designing a Process for Cleaning Technical Glasses

E170 Terminology Relating to Radiation Measurements and Dosimetry

E178 Practice for Dealing With Outlying Observations

E275 Practice for Describing and Measuring Performance of Ultraviolet and Visible Spectrophotometers

E666 Practice for Calculating Absorbed Dose From Gamma or X Radiation

E668 Practice for Application of Thermoluminescence-Dosimetry (TLD) Systems for Determining Absorbed Dose in Radiation-Hardness Testing of Electronic Devices

E925 Practice for Monitoring the Calibration of Ultraviolet-Visible Spectrophotometers whose Spectral Bandwidth does not Exceed 2 nm

E958 Practice for Estimation of the Spectral Bandwidth of Ultraviolet-Visible Spectrophotometers

E2628 Practice for Dosimetry in Radiation Processing

### 2.2 ISO/ASTM Standards:<sup>3</sup>

ISO/ASTM 51261 Practice for Calibration of Routine Dosimetry Systems for Radiation Processing

ISO/ASTM 51707 Guide for Estimating Uncertainties in Dosimetry for Radiation Processing

<sup>1</sup> This practice is under the jurisdiction of ASTM Committee E61 on Radiation Processing and is the direct responsibility of Subcommittee E61.02 on Dosimetry Systems.

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<sup>2</sup> The boldface numbers that appear in parentheses refer to a list of references at the end of this practice.

<sup>3</sup> For referenced ASTM and ISO/ASTM standards, visit the ASTM website, [www.astm.org](http://www.astm.org), or contact ASTM Customer Service at [service@astm.org](mailto:service@astm.org). For *Annual Book of ASTM Standards* volume information, refer to the standard's Document Summary page on the ASTM website.

### 2.3 ISO/IEC Standard:

ISO/IEC 17025 General requirements for the competence of testing and calibration laboratories<sup>4</sup>

2.4 International Commission on Radiation Units and Measurements (ICRU) Reports:<sup>5</sup>

ICRU Report 14 Radiation Dosimetry: X Rays and Gamma Rays with Maximum Photon Energies Between 0.6 and 50 MeV

ICRU Report 35 Radiation Dosimetry: Electrons with Initial Energies Between 1 and 50 MeV

ICRU Report 64 Dosimetry of High-Energy Photon Beams based on Standards of Absorbed Dose to Water

ICRU Report 80 Dosimetry Systems for Use in Radiation Processing

ICRU Report 85a Fundamental Quantities and Units for Ionizing Radiation

2.5 Joint Committee for Guides in Metrology (JCGM) Reports:<sup>6</sup>

JCGM 100:2008 GUM 1995, with minor corrections, Evaluation of measurement data – Guide to the Expression of Uncertainty in Measurement

2.6 National Research Council Canada (NRCC): PIRS-0815 The IRS Fricke Dosimetry System<sup>7</sup>

## 3. Terminology

### 3.1 Definitions:

3.1.1 *approved laboratory*—laboratory that is a recognized national metrology institute; or has been formally accredited to ISO/IEC 17025; or has a quality system consistent with the requirements of ISO/IEC 17025.

3.1.1.1 *Discussion*—A recognized national metrology institute or other calibration laboratory accredited to ISO/IEC 17025 should be used in order to ensure traceability to a national or international standard. A calibration certificate provided by a laboratory not having formal recognition or accreditation will not necessarily be proof of traceability to a national or international standard.

3.1.2 *molar linear absorption coefficient* ( $\epsilon_m$ )—a constant relating the spectrophotometric absorbance ( $A_\lambda$ ) of an optically absorbing molecular species at a given wavelength ( $\lambda$ ) per unit pathlength ( $d$ ) to the molar concentration ( $c$ ) of that species in solution:

$$\epsilon_m = \frac{A_\lambda}{(d \times c)} \quad (1)$$

Unit:  $\text{m}^2 \cdot \text{mol}^{-1}$

3.1.3 *radiation chemical yield* ( $G(x)$ )—quotient of  $n(x)$  by  $\bar{\epsilon}$ , where  $n(x)$  is the mean amount of a specified entity,  $x$ ,

produced, destroyed, or changed by the mean energy,  $\bar{\epsilon}$ , imparted to the matter.

$$G(x) = \left( \frac{n(x)}{\bar{\epsilon}} \right) \quad (2)$$

Unit:  $\text{mol} \cdot \text{J}^{-1}$

3.1.4 *reference standard dosimetry system*—dosimetry system, generally having the highest metrological quality available at a given location or in a given organization, from which measurements made there are derived.

3.1.5 *type I dosimeter*—dosimeter of high metrological quality, the response of which is affected by individual influence quantities in a well-defined way that can be expressed in terms of independent correction factors.

3.2 Definitions of other terms used in this standard that pertain to radiation measurement and dosimetry may be found in Terminology E170. Definitions in E170 are compatible with ICRU 85a; that document, therefore, may be used as an alternative reference.

## 4. Significance and Use

4.1 The Fricke dosimetry system provides a reliable means for measurement of absorbed dose to water, based on a process of oxidation of ferrous ions to ferric ions in acidic aqueous solution by ionizing radiation (4). In situations not requiring traceability to national standards, this system can be used for absolute determination of absorbed dose without calibration, as the radiation chemical yield of ferric ions is well characterized (see Appendix X3).

4.2 The dosimeter is an air-saturated solution of ferrous sulfate or ferrous ammonium sulfate that indicates absorbed dose by an increase in optical absorbance at a specified wavelength. A temperature-controlled calibrated spectrophotometer is used to measure the absorbance (ICRU 80).

## 5. Effect of Influence Quantities

5.1 The Fricke dosimeter response (change in optical absorbance) to a given radiation dose is dependent on irradiation temperature and measurement temperature. Thus, corrections may have to be applied for changes to the radiation chemical yield ( $G$ ) for irradiation temperature and to the molar linear absorption coefficient ( $\epsilon$ ) for measurement temperatures. both  $\epsilon(\text{Fe}^{3+})$  and  $G(\text{Fe}^{3+})$  increase with increase in temperature. The subscripts indicate the temperature of irradiation and measurement, as applicable.

$$\epsilon_{T_{meas}} = \epsilon_{25} [1 + 0.0069 (T_{meas} - 25)] \quad (3)$$

$$G_{T_{irrad}} = G_{25} [1 + 0.0012 (T_{irrad} - 25)] \quad (4)$$

5.2 The radiation chemical yield depends on the type and energy of the radiation employed and, in particular, changes significantly at low photon energies (5).

## 6. Interferences

6.1 The Fricke dosimeter response is extremely sensitive to impurities in the solution, particularly organic impurities. Even in trace quantities, impurities can cause a detectable change in the observed response. For high accuracy, organic materials

<sup>4</sup> Available from American National Standards Institute (ANSI), 25 W. 43rd St., 4th Floor, New York, NY 10036, <http://www.ansi.org>.

<sup>5</sup> Available from International Commission on Radiation Units and Measurements (ICRU), 7910 Woodmont Ave., Suite 400, Bethesda, MD 20841-3095, <http://www.icru.org>.

<sup>6</sup> Document produced by Working Group 1 of the Joint Committee for Guides in Metrology (JCGM/WG1). Available free of charge at the BIPM website (<http://www.bipm.org>).

<sup>7</sup> Available from the National Research Council, Ionizing Radiation Standards, Institute for National Measurement Standards, Ottawa, Ontario. K1A 0R6.

shall not be used for any component in contact with the solution, unless it has been demonstrated that the materials do not affect the dosimeter response.

6.2 Traces of metal ions in the irradiated and unirradiated dosimetric solutions can also affect dosimeter response. Therefore, do not use metal in any component in contact with the solutions.

6.3 If flame sealed ampoules are used as the dosimeters, exercise care in filling ampoules to avoid depositing solution in the ampoule neck. Subsequent heating during sealing of the ampoule may cause undesirable chemical change in the dosimetric solution remaining inside the ampoule neck. For the same reason, exercise care to avoid heating the body of the ampoule during sealing.

6.4 Thermal oxidation (as indicated by an increase in optical absorbance), in the absence of radiation, is a function of ambient temperature. At normal laboratory temperatures (about 20 to 25°C), this effect may be significant if there is a long period of time between solution preparation and photometric measurement. This interference is discussed further in 9.3.

6.5 The dosimetric solution is somewhat sensitive to ultraviolet light and should be kept in the dark for long-term storage. No special precautions are required during routine handling under normal laboratory lighting conditions, but strong UV sources such as sunlight should be avoided.

## 7. Apparatus

7.1 For the analysis of the dosimetric solution, use a high-precision spectrophotometer capable of measuring absorbance values up to 2 with an uncertainty of no more than  $\pm 1\%$  in the region of 300 nm. Use a quartz cuvette with 5- or 10-mm pathlength for spectrophotometric measurement of the solution. The cuvette capacity must be small enough to allow it to be thoroughly rinsed by the dosimeter solution and still leave an adequate amount of that solution to fill the cuvette to the appropriate level for the absorbance measurement. For dosimeter ampoules of less than 2 mL, this may require the use of semi-microcapacity cuvettes. Other solution handling techniques, such as the use of micro-capacity flow cells, may be employed provided precautions are taken to avoid cross-contamination. Either control the temperature of the dosimetric solution during measurement at  $25 \pm 0.5^\circ\text{C}$ , or determine the solution temperature during the spectrophotometric analysis and correct the measured absorbance to  $25^\circ\text{C}$  using Eq 3.

7.2 Use borosilicate glass or equivalent chemically-resistant glass to store the reagents and the prepared dosimetric solution. Clean all apparatus thoroughly before use (see Practice C912).

7.2.1 Store the cleaned glassware in a clean, dust-free environment. For extreme accuracy, bake the glassware in vacuum at  $550^\circ\text{C}$  for at least 1 h (6).

7.2.2 As an alternative method to baking the glassware, the dosimeter containers (for example, ampoules) may be filled with the dosimetric solution and irradiated to a dose of at least 500 Gy. When a container is needed, pour out the irradiated solution, rinse the container at least three times with unirradiated solution and then refill with the dosimetric solution to be irradiated. The time between filling, irradiation and measure-

ment should be as short as practical, preferably no more than a few hours. Refer to Note 2.

7.3 Use a sealed glass ampoule or other appropriate glass container to hold the dosimetric solution during irradiation.

NOTE 2—To minimize errors due to differences in radiation absorption properties between the container material and the Fricke solution, it is possible to use plastic containers (for example, PMMA or polystyrene) to hold Fricke solution. However, the interferences discussed in Section 6 may result in a reduction in accuracy. To reduce these problems, the plastic containers may be conditioned by irradiating them filled with dosimetric solution to approximately 500 Gy. The containers should then be thoroughly rinsed with unirradiated solution before use.

## 8. Reagents

8.1 *Purity of Reagents*—Reagent grade chemicals shall be used. Unless otherwise indicated, all reagents shall conform to the specifications of the Committee on Analytical Reagents of the American Chemical Society where such specifications are available.<sup>8</sup> Other grades may be used, provided it is first ascertained that the reagent is of sufficient high purity to permit its use without lessening the accuracy of the measurements. Methods of obtaining higher purity of chemicals exist (for example, crystallization or distillation), but are not discussed here.

8.2 *Purity of Water*—Water purity is very important since water is the major constituent of the dosimetric solution, and therefore, may be the prime source of contamination. The use of double-distilled water from coupled all-glass and silica stills is recommended. Alternatively, water from a high-quality commercial purification unit capable of achieving Total Oxidizable Carbon (T.O.C.) content below 5 ppb may be used. Use of deionized water is not recommended.

NOTE 3—Double-distilled water distilled from an alkaline permanganate ( $\text{KMnO}_4$ ) solution (2 g  $\text{KMnO}_4$  plus 5 g sodium hydroxide ( $\text{NaOH}$ ) in 2 L of distilled water) has been found to be adequate for routine preparation of the dosimetric solution. High purity water is commercially available from some suppliers. Water labelled HPLC (high pressure liquid chromatography) grade is usually sufficiently free of organic impurities to be used in this practice.

### 8.3 Reagents:

8.3.1 *Ferrous Ammonium Sulfate*— $(\text{NH}_4)_2\text{Fe}(\text{SO}_4)_2 \cdot 6\text{H}_2\text{O}$ .

8.3.2 *Sodium Chloride* ( $\text{NaCl}$ ).

8.3.3 *Sulfuric Acid* ( $\text{H}_2\text{SO}_4$ ).

## 9. Preparation of Dosimeters

9.1 Prepare dosimetric solution:

9.1.1 Dissolve 0.392 g of ferrous ammonium sulfate,  $(\text{NH}_4)_2\text{Fe}(\text{SO}_4)_2 \cdot 6\text{H}_2\text{O}$ , and 0.058 g of sodium chloride,  $\text{NaCl}$ , in 12.5 mL of 0.4  $\text{mol}\cdot\text{L}^{-1}$  sulfuric acid,  $\text{H}_2\text{SO}_4$ . Dilute to 1 L in a volumetric flask with air-saturated 0.4  $\text{mol}\cdot\text{L}^{-1}$  sulfuric acid at  $25^\circ\text{C}$ . To make 0.4 M solution, use 41.0 g of 96.7 % sulfuric acid plus water to make 1 L of solution.

<sup>8</sup> *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. Pharmaceutical Convention, Inc. (USPC), Rockville, MD.

NOTE 4—Sodium chloride is used to reduce any adverse effects on the response of the dosimeter due to trace organic impurities.

9.1.2 If the final solution is not yet air-saturated, it should be done. Shaking of the solution is normally sufficient to achieve this. Alternatively, bubble high-purity air through the solution, taking care to avoid any possible organic contamination of the air. The oxygen concentration in air-saturated solution is adequate to ensure the dosimeter's linear response up to 400 Gy. Store the dosimetric solution in clean borosilicate glass containers in the dark.

9.2 The dosimetric solution has the following concentrations:  $1 \times 10^{-3}$  mol·L<sup>-1</sup> ferrous ammonium sulfate;  $1 \times 10^{-3}$  mol·L<sup>-1</sup> sodium chloride; and 0.4 mol·L<sup>-1</sup> sulfuric acid.

9.3 The dosimetric solution will slowly oxidize at room temperature resulting in an increase in the optical absorbance of the unirradiated solution. If the solution has not been used for some time, measure the absorbance of the unirradiated solution, as described in 10.4. If the absorbance of a 10-mm pathlength sample is greater than 0.1, do not use that solution. Prepare a fresh batch of solution to replace it.

NOTE 5—Oxidation of the solution at room temperature can be significantly reduced by refrigerating the solution, but refrigeration may also change the oxygen concentration.

9.4 Rinse the dosimeter containers (ampoules or other types) at least three times with the dosimetric solution before filling them for irradiation. Even with careful rinsing, there will always be solution remaining; subsequent rinsing will help mitigate this effect.

9.5 Fill clean containers with the dosimetric solution. If flame sealing the dosimeters, observe the precautions in 6.3.

9.6 An alternative method of preparation using concentrated stock solution is described in Appendix X1. Each dilution made from the stock solution should be treated as a separate batch for the purposes of calibration.

## 10. Calibration of the Dosimetry System

10.1 Prior to use, the dosimetry system (consisting of a specific batch of dosimeters and specific measurement instruments) shall be calibrated in accordance with the user's documented procedure, that specifies details of the calibration and quality assurance requirements. This calibration shall be repeated at regular intervals to ensure that the accuracy of the absorbed dose measurement is maintained within required limits. Calibration methods are described in ISO/ASTM Practice 51261.

NOTE 6—The quality of the Fricke dosimetry system is potentially high if prepared and used correctly and it is capable of dose determination using published  $\epsilon$  or  $G$  values, or the value of their product. However, doses determined in this way cannot be considered traceable to national or international standards without additional evidence, such as comparison with known traceable standards. For completeness, details of this method are given in Appendix X3, but it is not recommended in situations where traceability to national or international standards is a regulatory requirement.

10.2 *Calibration Irradiation of Dosimeters*—Irradiation is a critical component of the calibration of the dosimetry system.

10.2.1 When the Fricke dosimeter is used in a reference standard dosimetry system, calibration irradiations shall be performed at an approved laboratory, as defined in 3.1.1, and have demonstrable traceability to nationally or internationally recognized standards.

10.2.2 When the Fricke dosimeter is used in a routine dosimetry system, the calibration irradiation may be performed in accordance with 10.2.1, or at a production or research irradiation facility together with reference- or transfer-standard dosimeters from a system that has measurement traceability to nationally or internationally recognized standards.

10.2.3 Specify the calibration dose in terms of absorbed dose to water.

10.2.4 For calibration with photons, the Fricke dosimeter shall be irradiated under conditions that approximate electron equilibrium.

10.2.5 When using an electron beam for irradiation, locate the dosimeters in a well-characterized position within the radiation field.

10.2.6 Ensure that the radiation field within the volume occupied by the dosimeters is as uniform as possible. The variation in dose rate within this volume should be known, and be within acceptable limits for the uncertainty of calibration required.

10.2.7 The dosimeter shall be calibrated in a radiation field of the same type and energy as that in which it is to be used, unless evidence is available to demonstrate equivalence of response.

10.2.8 Calibrate each batch of dosimeters prior to use.

10.2.9 Separate five dosimeters from the remainder of the batch and do not irradiate them. Use them in determining  $A_0$  (see 10.5.1).

10.2.10 Control (or monitor) the temperature of the dosimeters during irradiation. Calculate or measure the mean irradiation temperature of each dosimeter to an accuracy of  $\pm 2^\circ\text{C}$ , or better.

10.2.11 Use a set of at least three dosimeters for each absorbed dose value.

10.2.12 Irradiate these sets of dosimeters to at least five known dose values covering the range of utilization in order to determine the calibration curve for the dosimetry system.

10.3 *Measurement Instrument Calibration and Performance Verification*—For the calibration of the instruments, and for the verification of instrument performance between calibrations, see ISO/ASTM Practice 51261 and instrument-specific operating manuals.

10.3.1 Check the wavelength scale of the spectrophotometer and establish its accuracy. The emission spectrum from a low-pressure mercury arc lamp can be used for this purpose. Such a lamp may be obtained from the spectrophotometer manufacturer or other scientific laboratory instrument suppliers. Other appropriate wavelength standards are holmium oxide filters or solutions. For more details, see Practices E275, E925, and E958.