Designation: C1720 – 11ε1

Standard Test Method for Determining Liquidus Temperature of Immobilized Waste Glasses and Simulated Waste Glasses[1]

This standard is issued under the fixed designation C1720; 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.

ε[1] NOTE—Units statement was editorially corrected in April 2015.

1. Scope

1.1 These practices cover procedures for determining the liquidus temperature (T_L) of nuclear waste, mixed nuclear waste, simulated nuclear waste, or hazardous waste glass in the temperature range from 600°C to 1600°C. This method differs from Practice C829 in that it employs additional methods to determine T_L. T_L is useful in waste glass plant operation, glass formulation, and melter design to determine the minimum temperature that must be maintained in a waste glass melt to make sure that crystallization does not occur or is below a particular constraint, for example, 1 volume % crystallinity or $T_{1\%}$. As of now, many institutions studying waste and simulated waste vitrification are not in agreement regarding this constraint (**1**).

1.2 Three methods are included, differing in (*1*) the type of equipment available to the analyst (that is, type of furnace and characterization equipment), (*2*) the quantity of glass available to the analyst, (*3*) the precision and accuracy desired for the measurement, and (*4*) candidate glass properties. The glass properties, for example, glass volatility and estimated T_L, will dictate the required method for making the most precise measurement. The three different approaches to measuring T_L described here include the following: (*A*) *Gradient Temperature Furnace Method (GT)*, (*B*) *Uniform Temperature Furnace Method (UT)*, and (*C*) *Crystal Fraction Extrapolation Method (CF)*. This procedure is intended to provide specific work processes, but may be supplemented by test instructions as deemed appropriate by the project manager or principle investigator. The methods defined here are not applicable to glasses that form multiple immiscible liquid phases. Immiscibility may be detected in the initial examination of glass during sample preparation (see 9.3). However, immiscibility may not become apparent until after testing is underway.

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

2. Referenced Documents

2.1 *ASTM Standards:*[2]

C162 Terminology of Glass and Glass Products

C829 Practices for Measurement of Liquidus Temperature of Glass by the Gradient Furnace Method

D1129 Terminology Relating to Water

D1193 Specification for Reagent Water

E177 Practice for Use of the Terms Precision and Bias in ASTM Test Methods

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

E2282 Guide for Defining the Test Result of a Test Method

2.2 *Other Documents:*

SRM-773 National Institute for Standards and Technology (NIST) Liquidus Temperature Standard

SRM-674b NIST X-Ray Powder Diffraction Intensity Set for Quantitative Analysis by X-Ray Diffraction (XRD)

SRM-1976a NIST Instrument Response Standard for X-Ray Powder Diffraction

Z540.3 American National Standards Institute/National Conference of Standards Laboratories (ANSI/NCSL) Requirements for the Calibration of Measuring and Test Equipment

3. Terminology

3.1 *Definitions:*

[1] This test method is under the jurisdiction of ASTM Committee C26 on Nuclear Fuel Cycle and is the direct responsibility of Subcommittee C26.13 on Spent Fuel and High Level Waste.

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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.

3.1.1 *air quenching*—to pour or place a molten glass specimen on a surface, for example, a steel plate, and cool it to the solid state.

3.1.2 *anneal*—to prevent or remove materials processing stresses in glass by controlled cooling from a suitable temperature, for example, the glass transition temperature (T_g) (modified from Terminology C162).

3.1.3 *annealing*—a controlled cooling process for glass designed to reduce thermal residual stress to an acceptable level and, in some cases, modify structure (modified from Terminology C162).

3.1.4 *ASTM Type I water*—purified water with a maximum total matter content including soluble silica of 0.1 g/m^3, a maximum electrical conductivity of 0.056 μΩ/cm at 25°C and a minimum electrical resistivity of 18 MΩ × cm at 25°C (see Specification D1193 and Terminology D1129).

3.1.5 *cleaning glass*—glass or flux used to remove high viscosity glass, melt insolubles, or other contamination from platinum-ware.

3.1.6 *crystallize*—to form or grow, or both, crystals from a glass melt during heat-treatment or cooling.

3.1.7 *crystallization*—the progression in which crystals are first nucleated and then grown within a host medium. Generally, the host may be a gas, liquid, or another crystalline form. However, in this context, it is assumed that the medium is a glass melt.

3.1.8 *crystallization front*—the boundary between the crystalline and crystal-free regions in a test specimen that was subjected to a temperature gradient heat-treatment.

3.1.9 *furnace profiling*—the process of determining the actual temperature inside of a furnace at a given location; this involves different processes for different types of furnaces.

3.1.10 *glass*—an inorganic product of fusion that has cooled to a rigid condition without crystallizing (see Terminology C162); a noncrystalline solid or an amorphous solid (**2**).[3]

3.1.11 *glass ceramic*—solid material, partly crystalline and partly glassy (see Terminology C162).

3.1.12 *glass sample*—the material to be heat-treated or tested by other means.

3.1.13 *glass specimen*—the material resulting from a specific heat treatment.

3.1.14 *glass transition temperature (T_g)*—on heating, the temperature at which a glass transforms from a solid to a liquid material, characterized by the onset of a rapid change in several properties, such as thermal expansivity.

3.1.15 *gradient furnace*—a furnace in which a known temperature gradient is maintained between the two ends.

3.1.16 *hazardous waste glass*—a glass composed of glass forming additives and hazardous waste.

3.1.17 *homogeneous glass*—a glass that is a single amorphous phase; a glass that is not separated into multiple amorphous phases.

3.1.18 *inhomogeneous glass*—a glass that is not a single amorphous phase; a glass that is either phase separated into multiple amorphous phases or is crystallized.

3.1.19 *liquidus temperature*—the maximum temperature at which equilibrium exists between the molten glass and its primary crystalline phase.

3.1.20 *melt insoluble*—a crystalline, amorphous, or mixed phase material that is not appreciably soluble in molten glass, for example, noble metals, noble metal oxides.

3.1.21 *mixed waste*—waste containing both radioactive and hazardous components regulated by the Atomic Energy Act (AEA) (**3**) and the Resource Conservation and Recovery Act (RCRA) (**4**), respectively; the term "radioactive component" refers to the actual radionuclides dispersed or suspended in the waste substance (**5**).

3.1.22 *mold*—a pattern, hollow form, or matrix for giving a certain shape or form to something in a plastic or molten state. **Webster's**[4]

3.1.23 *nuclear waste glass*—a glass composed of glass-forming additives and radioactive waste.

3.1.24 *observation*—the process of obtaining information regarding the presence or absence of an attribute of a test specimen or of making a reading on a characteristic or dimension of a test specimen (see Terminology E2282).

3.1.25 *phase separated glass*—a glass containing more than one amorphous phase.

3.1.26 *preferred orientation*—when there is a stronger tendency for the crystallites in a powder or a texture to be oriented more one way, or one set of ways, than all others. This is typically due to the crystal structure. **IUCr**[5]

3.1.27 *primary phase*—the crystalline phase at equilibrium with a glass melt at its liquidus temperature.

3.1.28 *radioactive*—of or exhibiting radioactivity; a material giving or capable of giving off radiant energy in the form of particles or rays, for example, α, β, and γ, by the disintegration of atomic nuclei; said of certain elements, such as radium, thorium, and uranium and their products. **American Heritage**[6] **Webster's**[7]

3.1.29 *Round-Robin*—an interlaboratory and intralaboratory testing process to develop the precision and bias of a procedure.

3.1.30 *section*—a part separated or removed by cutting; a slice, for example, representative thin section of the glass specimen. **Webster's**[4]

3.1.31 *set of samples*—samples tested simultaneously in the same oven.

[3] The boldface numbers in parentheses refer to a list of references at the end of this standard.

[4] Webster's New Universal Unabridged Dictionary, 1979.
[5] IUCr Online Dictionary of Crystallography, 2011.
[6] American Heritage Dictionary, 1973.
[7] Webster's New Twentieth Century Dictionary, 1973.

3.1.32 *simulated nuclear waste glass*—a glass composed of glass forming additives with simulants of, or actual chemical species, or both, in radioactive wastes or in mixed nuclear wastes, or both.

3.1.33 *standard*—to have the quality of a model, gage, pattern, or type. **Webster's**[7]

3.1.34 *standardize*—to make, cause, adjust, or adapt to fit a standard (**5**); to cause to conform to a given standard, for example, to make standard or uniform. **Webster's**[7]

3.1.35 *surface tension*—a property, due to molecular forces, by which the surface film of all liquids tends to bring the contained volume into a form having the least possible area.

3.1.36 *test determination*—the value of a characteristic or dimension of a single test specimen derived from one or more observed values (see Terminology E2282).

3.1.37 *test method*—a definitive procedure that produces a test result (see Terminology E2282).

3.1.38 *test observation*—see *observation*.

3.1.39 *test result*—the value of a characteristic obtained by carrying out a specific test method (see Terminology E2282).

3.1.40 *uniform temperature furnace*—a furnace in which the temperature is invariant over some defined volume and within some defined variance.

3.1.41 *vitrification*—the process of fusing waste with glass making chemicals at elevated temperatures to form a waste glass (see Terminology C162).

3.1.42 *volatility*—the act of one or more constituents of a solid or liquid mixture to pass into the vapor state.

3.1.43 *waste glass*—a glass developed or used for immobilizing radioactive, mixed, or hazardous wastes.

3.2 *Abbreviations:*

3.2.1 *AEA*—Atomic Energy Act

3.2.2 *ANSI*—American National Standards Institute

3.2.3 *ASTM*—American Society for Testing and Materials

3.2.4 *CF*—crystal fraction extrapolation

3.2.5 C_F—crystal fraction in a sample or specimen

3.2.6 *EDS*—energy dispersive spectrometry

3.2.7 η—viscosity

3.2.8 *FWHM*—full width of a peak at half maximum

3.2.9 *GF*—gradient temperature furnace

3.2.10 *GT*—gradient temperature

3.2.11 *HF*—hydrofluoric acid

3.2.12 *HLW*—high-level waste

3.2.13 *ID*—identification

3.2.14 *NBS*—National Bureau of Standards

3.2.15 *NCSL*—National Conference of Standards Laboratories

3.2.16 *NIST*—National Institute for Standards and Technology (formerly NBS)

3.2.17 *OM*—optical microscope or optical microscopy

3.2.18 *PDF*—powder diffraction file

3.2.19 *RCRA*—Resource Conservation and Recovery Act

3.2.20 *RIR*—relative intensity ratio

3.2.21 *RLM*—reflected light microscopy

3.2.22 *SEM*—scanning electron microscope or scanning electron microscopy

3.2.23 *SRM*—Standard Reference Material

3.2.24 $T_{1\%}$—temperature where glass contains 1 volume % of a crystalline phase

3.2.25 T_a—primary UT measurement above T_L

3.2.26 T_c—primary UT measurement below T_L

3.2.27 T_g—glass transition temperature

3.2.28 T_L—liquidus temperature

3.2.29 *TLM*—transmitted light microscopy

3.2.30 T_M—melting temperature for glass preparations

3.2.31 *UF*—uniform temperature furnace

3.2.32 *UT*—uniform temperature

3.2.33 *WC*—tungsten carbide

3.2.34 *XRD*—X-ray diffraction

4. Summary of Test Method

4.1 This procedure describes methods for determining the T_L of waste or simulated waste glasses. Temperature is defined as the maximum temperature at which equilibrium exists between the molten glass and its primary crystalline phase. In other words, T_L is the maximum temperature at which a glass melt crystallizes. Fig. 1 illustrates an example T_L for a simple two-component liquid on a binary phase diagram.

4.1.1 *(A) Gradient Temperature Furnace Method (GT)*—This method is similar to Practice C829, "Standard Practices for Measurement of Liquidus Temperature of Glass by the Gradient Furnace Method," though it has been modified to meet the specific needs of waste and simulated waste glass measurements. The most pronounced differences between this method and the Practice C829 "boat method" are the sample preparation and examination procedures.

4.1.1.1 Samples are loaded into a boat, for example, platinum alloy (Fig. 2) with a tight-fitting lid, and exposed to a linear temperature gradient in a gradient furnace (Fig. 3) for a fixed period of time. The temperature, as a function of distance, *d*, along the sample, is determined by its location within the GF, and the T_L is then related to the location of the crystallization front in the heat-treated specimen (Fig. 4).

4.1.1.2 Following the heat-treatment, the specimen should be annealed at or near the glass transition, T_g, of the glass (this should be previously measured or estimated) to reduce specimen cracking during cutting and polishing.

4.1.1.3 The specimen should then be scored or marked to signify the locations on the specimen located at different depths into the gradient furnace, that is, locations heat-treated at specific temperatures.

4.1.1.4 If the specimen is optically transparent, it can be observed with transmitted light (that is, transmitted light microscopy or TLM) or reflected light microscopy (RLM) to look for bulk or surface crystallization, respectively. If the specimen is not optically transparent or is barely optically