Designation: C1308 − 08 (Reapproved 2017)

Standard Test Method for Accelerated Leach Test for Diffusive Releases from Solidified Waste and a Computer Program to Model Diffusive, Fractional Leaching from Cylindrical Waste Forms[1]

This standard is issued under the fixed designation C1308; 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 provides procedures for measuring the leach rates of elements from a solidified matrix material, determining if the releases are controlled by mass diffusion, computing values of diffusion constants based on models, and verifying projected long-term diffusive releases. This test method is applicable to any material that does not degrade or deform during the test.

1.1.1 If mass diffusion is the dominant step in the leaching mechanism, then the results of this test can be used to calculate diffusion coefficients using mathematical diffusion models. A computer program developed for that purpose is available as a companion to this test method (Note 1).

1.1.2 It should be verified that leaching is controlled by diffusion by a means other than analysis of the leach test solution data. Analysis of concentration profiles of species of interest near the surface of the solid waste form after the test is recommended for this purpose.

1.1.3 Potential effects of partitioning on the test results can be identified through modeling, although further testing and analyses are required to determine the cause of partitioning (for example, if it occurs during production of the material or as a result of leaching).

1.2 The method is a modification of other semi-dynamic tests such as the IAEA test (**1**)[2] and the ANS 16.1 Leach Test wherein elevated temperatures are used to accelerate diffusive release to an extent that would only be reached after very long times at lower temperatures. This approach provides a mechanistic basis for calculating diffusive releases at repository-relevant temperatures over long times, provided that the leaching mechanism does not change with temperature.

1.2.1 Tests can be conducted at elevated temperatures to accelerate diffusive release and provide a mechanistic basis for calculating diffusive releases that would occur at lower temperatures over long times. Tests conducted at high temperatures allow the temperature dependence of the diffusion coefficient to be determined. They also demonstrate that the diffusion mechanism is rate-limiting through the measured extent of diffusive release.

1.2.2 Releases at any temperature can be projected up to the highest cumulative fractional release value that has been measured for that material (at any temperature), provided that the mechanism does not change. The mechanism is considered to remain unchanged over a range of temperatures if the diffusion coefficients show Arrhenius behavior over that range.

NOTE 1—A computer program in which the test results are evaluated using three diffusion models is briefly described in Annex A1 and in the Accelerated Leach Test Method and User's Guide for the "ALT" Computer Program (**2**). The data are fit with model equations for diffusion from a semi-infinite solid, diffusion from a finite cylinder, and diffusion with partitioning of the species of interest to determine effective diffusion coefficients and quantify the goodness of fit. The User's Guide contains several typographical errors; these are identified in Annex A1.

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:*[3]

C1220 Test Method for Static Leaching of Monolithic Waste Forms for Disposal of Radioactive Waste

[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.

Current edition approved Jan. 1, 2017. Published January 2017. Originally approved in 1995. Last previous edition approved in 2008 as C1308 – 08. DOI: 10.1520/C1308-08R17.

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

[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.

D1193 Specification for Reagent Water

2.2 *ANSI/ANS Standard:*

ANSI 16.1 Measurement of the Leachability of Solidified Low-Level Radioactive Wastes by a Short-Term Test Procedure[4]

3. Terminology

3.1 *Definitions:*

3.1.1 *cumulative fraction leached*—the sum of the fractions of a species leached during all sampling intervals prior to and including the present interval divided by the amount of that species in the test specimen before the test.

3.1.2 *diffusion coefficient (diffusivity)*—an intrinsic property of a species that relates *(1)* its concentration gradient to its flux in a given medium (Fick's first law), *(2)* its spatial rate of change in the direction of the concentration gradient to the time rate of change in its concentration in a given medium (Fick's second law), or *(3)* its mean square displacement to time in a given medium (the Einstein equation).

3.1.3 *effective diffusion coefficient (D_e)*—the diffusion coefficient as modified by other processes (for example, adsorption) or physical constraints (for example, tortuosity and constrictivity).

3.1.4 *finite cylinder (finite medium)*—a bounded body for which Fick's diffusion equation can be solved.

3.1.5 *incremental fraction leached*—the amount of a species leached during a single sampling interval divided by the amount of that species in the test specimen before the test.

3.1.6 *leachant*—the initial solution with which a solid is contacted and into which the solid dissolves or is leached.

3.1.7 *leachate*—the final solution resulting from a test in which a solid is contacted by a solution and leaches or dissolves.

3.1.8 *leaching*—the preferential loss of components from a solid material into solution leaving a residual phase that is depleted in those components, but structurally unchanged.

3.1.9 *leaching interval*—the length of time during which a given volume of leachant is in contact with a specimen.

3.1.10 *leaching mechanism*—the set of processes that controls the rate of mass transport of a species out of a specimen during leaching.

3.1.11 *matrix material*—the solid material used to immobilize the waste or species of interest.

3.1.12 *reference leach test*—a leach test conducted under defined conditions, the results of which are used as a standard against which the results of other leach tests are compared. In this test method, a reference leach test is one that is conducted at 20°C using demineralized water.

3.1.13 *semi-dynamic leach test*—a leach test method in which the specimen is exposed to fresh leachant on a periodic schedule.

3.1.14 *semi-infinite medium*—a body having a single planar surface and extending infinitely in the directions parallel to the surface and in one direction normal to the surface.

3.1.15 *source term*—in this test method, the concentration of a species of interest in a specimen prior to leaching.

3.1.16 *specimen volume*—for purposes of this test method, the volume of a monolithic specimen calculated from macroscopic measurements of its dimensions by assuming a simple geometric shape, such as a right circular cylinder.

3.1.17 *surface area*—for purposes of this test method, the geometric surface area of a monolithic specimen that is calculated from macroscopic measurements of its dimensions by assuming a simple geometric shape, such as a right circular cylinder.

3.1.18 *waste form*—the waste material and any encapsulating or stabilizing matrix in which it is incorporated.

4. Summary of Test Method

4.1 This test method is a semi-dynamic leach test in which a cylindrical specimen is immersed in a leachant that is completely replaced after specified intervals. The concentration of an element of interest in the recovered test solution is measured after each exchange; this is referred to as the *incremental fraction leached (IFL)*. The accumulated amount of the species of interest in the intervals prior to and including the interval of interest is analyzed to determine if the release from the solid can be described using a mass diffusion model. The amount accumulated through a particular test duration is referred to as the *cumulative fraction leached (CFL)*.

4.2 Tests at a single temperature are adequate to compare the leaching behaviors of different materials.

4.3 The results of tests at repository-relevant temperatures can be extrapolated to long times if data from tests run at elevated temperatures and data from tests run at the reference temperature (20°C) can be modeled using a diffusion model and the diffusion coefficients show Arrhenius behavior.

4.3.1 Elevated temperatures are used to accelerate the release of a species of interest and collect enough data to show that the release is controlled by diffusion and determine the value of the diffusion coefficient.

4.3.2 Tests must be performed at a minimum of three temperatures to verify that the leaching mechanism does not change over that temperature range.

4.3.3 By generating data over a range of temperatures, an Arrhenius plot can be produced to interpolate values of the diffusion coefficient within the temperature range that was tested. Values cannot be extrapolated to temperatures that are higher or lower than the temperature range spanned by the tests.

4.3.4 A computer program that plots the experimental data and a regression curve calculated using a finite cylinder model (**2**) is available from ASTM (see Note 1). The program provides the value of the effective diffusion coefficient, the modeled *IFL* and *CFL* values, and a measure of the goodness of fit of the model.

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

4.4 If the data from the accelerated tests, the reference test, and the fit of the modeled curve agree within defined criteria, the leaching mechanism can be taken to be diffusion-controlled and a diffusion model can be used to calculate releases from full-scale waste forms for long times.

4.4.1 The accelerated leach test provides the maximum cumulative fractional release to which the modeled data can be extrapolated. The maximum cumulative fractional release measured represents the maximum extent of reaction for which the consistency of the mechanism has been verified for that material.

4.4.2 Because the cumulative fraction leached is a function of the specimen surface area-to-volume ratio, the results of tests with the small-scale specimens used in the ALT directly represent leaching from large-scale waste forms having the same aspect ratio.

4.4.3 The effective diffusion coefficient can be used to calculate diffusive releases from waste forms with other shapes.

4.5 If the diffusion model does not fit the data within defined criteria, no extrapolation can be made in time or specimen size. However, other models can be applied to the data to evaluate the leaching process.

4.5.1 A model including diffusion with partitioning of the species of interest between phases having different release behaviors is included in the computer program (**2**).

4.5.2 The possibility of a solubility-limit to the release of the species of interest is addressed in the computer program (**2**).

4.6 If the data cannot be fit with a diffusion model within the defined criterion, then graphical comparisons of the data are recommended for added insight: For example, a plot of the cumulative fraction leached (*CFL*) from ALT conducted at an elevated temperature against the *CFL* from ALT conducted at the reference temperature can be used to verify that the accelerated data are consistent with the reference data and that the accelerated test appropriately accelerates the release, even though the release is not diffusion-limited.

5. Significance and Use

5.1 This test method can be used to measure the release of a component from a cylindrical solidified waste form into water at the reference temperature of 20°C and at elevated temperatures that accelerate the rate and extent of leaching relative to the values measured at 20°C.

5.2 This test method can be used to:

5.2.1 Compare releases of waste components from various types of solidification agents and formulations.

5.2.2 Determine the diffusion coefficients for the release of waste components from waste forms at a specific temperature.

5.2.3 Promote greater extents of reaction than can be achieved under expected service conditions within a laboratory time frame to provide greater confidence in modeled diffusive releases.

5.2.4 Determine the temperature dependence of diffusive release.

5.3 Fitting the experimental results with a mechanistic model allows diffusive releases to be extrapolated to long times and to full-scale waste forms under the following constraints:

5.3.1 Results of this test method address an intrinsic property of a material and should not be presumed to represent releases in specific disposal environments. Tests can be conducted under conditions that represent a specific disposal environment (for example, by using a representative groundwater) to determine an effective diffusion coefficient for those conditions.

5.3.2 Projections of releases over long times requires that the waste form matrix remain stable, which may be demonstrated by the behavior of the specimen in ALTs at elevated temperatures.

5.3.3 Extrapolations in time and scale are limited to values that correspond to the maximum *CFL* value obtained in an accelerated test.

5.3.4 The mechanism must be the same at all temperatures used in the extrapolation. The same model that describes the results of tests conducted at elevated temperatures must also describe the results of tests run at the reference temperature of 20°C.

6. Apparatus

6.1 A forced-air environmental chamber or a circulating water bath capable of controlling leachant temperatures to within 1°C of the target test temperature shall be used.

6.2 *Balance*—The balance shall be accurate to 0.1 % of the test load.

7. Reagents and Materials

7.1 *Leachant*—The leachant can be selected with regard to the material being tested and the information that is desired. Demineralized water, synthetic or actual groundwaters, or chemical solutions can be used. The effects of the leachant solution on the species of interest (that is, the species for which the diffusion coefficient is to be measured) and the solid must be considered. For example, the leachant should not degrade the host solid. In general, the leachant should be devoid of the species of interest to minimize solution feedback and solubility limit effects. If the leachant does contain a non-negligible amount of the species of interest, blank tests should be conducted to provide background concentrations to calculate the amounts released from the solid by using the concentrations measured in the tests. If demineralized water is used, it must meet or exceed the standards for types II or III reagent water specified in Specification D1193.

7.2 *Leaching Containers*—Leaching containers shall be made of a material that does not react with the leachant, leachate, or specimen. It is particularly important to select materials that are not susceptible to plate-out of species of interest from solution. High density polyethylene has been found to be a suitable container material. The top of the container shall fit tightly to minimize evaporation. The mass of the vessel must be checked before sampling to verify that evaporative losses are less than 1 % of the leachant mass (or volume) over every test interval.