



Designation: C 1308 – 95 (Reapproved 2001)

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 Forms¹

This standard is issued under the fixed designation C 1308; 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 a method for accelerating the leach rate of solidified waste and determining if the release is diffusion-controlled. This test method is applicable to any material that does not degrade, deform, or change leaching mechanism during the test.

1.1.1 If diffusion is the dominant leaching mechanism, then results of this test can be used to model long-term releases from waste forms. Diffusion can be confirmed as the leaching mechanism through the use of a computerized mathematical model for diffusion from the finite cylinder (Note 1).

1.1.2 The leaching mechanism should be verified as diffusion-controlled by a means other than analysis of the leach test data. For this purpose, analysis of post-leaching concentration profiles within the solid waste form is recommended.

NOTE 1—The computer program and the models are briefly described in Annex A1 and in the Accelerated Leach Test Method and User's Guide for the "ALT" Computer Program (1).

1.2 *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:

E 632 Standard Practice for Developing Accelerated Tests to Aid Prediction of the Service Life of Building Components and Materials²

D 1193 Standard Specification for Reagent Water³

2.2 ANSI/ANS Standard:

ANSI 16.1 Measurement of the Leachability of Solidified

Low-Level Radioactive Wastes by a Short-Term Test Procedure⁴

3. Terminology

3.1 *cumulative fraction leached*—the sum of the fractions of a species leached during all sampling intervals calculated as $CFL = \sum a_n / A_O$ and assuming no radioactive decay, where a_n is the quantity of a species in the leachate during any interval and A_O is the quantity of that species originally present in the sample.

3.2 *diffusion coefficient (diffusivity)*—based on Fick's Laws for diffusion, the diffusion coefficient is the ratio of the rate of transfer of a diffusing substance through the unit area of a section to the concentration gradient measured normal to the section.

3.3 *effective diffusion coefficient (effective diffusivity)*—the diffusion coefficient that results from diffusion as it is modified by other processes (for example, adsorption) or physical constraints (for example, tortuosity and constrictivity).

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

3.5 *incremental fraction leached*—the fraction leached of a species of interest during a single sampling interval calculated as $IFL = a_n / A_O$ and assuming no radioactive decay.

3.6 *leachant*—the liquid that contacts the specimen during a leach test or contacts a waste form in the disposal environment.

3.7 *leachate*—the leachant after contacting the specimen or the waste form.

3.8 *leaching*—the process (or processes) by which mass transport from a solid to a liquid takes place.

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

3.10 *leaching mechanism*—the process that controls the rate of mass transport out of a specimen during leaching.

3.11 *reference leach test*—a leach test conducted under defined conditions, the results of which are used as a standard

¹ This test method is under the jurisdiction of ASTM Committee C26 on Nuclear Fuel Cycle and is the direct responsibility of Subcommittee C26.07 on Waste Materials.

Current edition approved Sept. 10, 1995. Published April 1996.

² *Annual Book of ASTM Standards*, Vol 14.02.

³ *Annual Book of ASTM Standards*, Vol 11.01.

⁴ Available from American National Standards Institute, 11 W. 42nd St., 13th Floor, New York, NY 10036.

against which the results of other leach tests (for example, accelerated) are compared. In this test method a reference leach test is one that is conducted at 20°C.

3.12 *semi-dynamic leach test*—a leach test method that exposes the specimen to fresh leachant on a periodic schedule.

3.13 *semi-infinite medium*—a body used in diffusion theory whose outer boundary is effectively at an infinite distance from the inner region.

3.14 *source term*—the original concentration, prior to leaching, of a species of interest in a specimen or a waste form.

3.15 *surface area*—for purposes of this test method, surface area is defined as the geometric surface area of a specimen calculated from macroscopic measurements of its dimensions.

3.16 *volume*—for purposes of this test method, volume is defined as the volume of a specimen calculated from macroscopic measurements of its dimensions.

3.17 *waste form*—a stable, solid body composed of the waste and a solidification agent.

4. Summary of Test Method

4.1 This test method is a semi-dynamic leach test, that is, the leachant is sampled and replaced periodically. It is based on earlier semi-dynamic tests such as the IAEA test (2) and the ANS 16.1 Leach Test.⁴ Elevated temperatures, large volumes of leachant, frequent leachant changes, and small specimen size are used to obtain accelerated releases. This is a short-term test, requiring sampling on each of eleven or more successive days. Tests must be performed to demonstrate that the leaching mechanism does not change at the elevated test temperature. This may be done by performing leach tests at a minimum of three temperatures to ascertain that the mechanisms and structural controls of leaching do not change with increasing temperature. If the test is to be used only for making comparisons among specimens, then testing at a single temperature is adequate.

4.2 The results of this accelerated test can be extrapolated to long times if the data from tests run at elevated temperatures and the tests run at the reference temperature (20°C), and can be modeled by diffusion. A computer program that plots the experimental data and a curve calculated from an effective diffusion coefficient for diffusion from a finite cylinder (Fig. 1) is available from ASTM. If the data from the accelerated tests, the reference test, and the modeled curve fit within defined criteria, the leaching mechanism is taken to be diffusion. In this case, the model can be used to project releases from full-scale waste forms and to long times. The accelerated test provides a measure of the maximum fractional release to which the modeled data can be extrapolated. By generating data over a specified temperature range, an Arrhenius plot can be produced allowing projections to be made at temperatures other than those tested. If the diffusion model cannot fit the data, other models (for example, diffusion plus partitioning and solubility limited leaching) contained in the computer program can be used to indicate the leaching mechanism that controls releases. No extrapolations are allowed with these models. If no model fits the data, then an alternative graphical comparison of the data is recommended. A linear plot of modeled CFL plotted against experimental CFL verifies that the accelerated data is

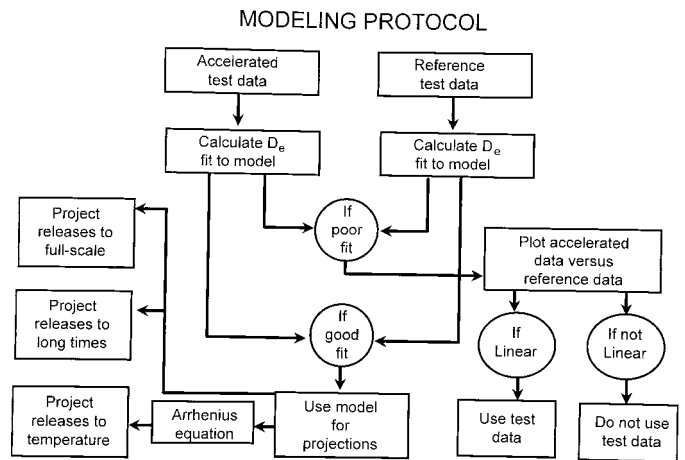


FIG. 1 A Schematic of the Concepts Used in This Test Method to Combine Experimental and Modeling Approaches

comparable to the reference data, showing that the accelerated test is appropriate. With this technique, no extrapolation of data can be made.

5. Significance and Use

5.1 This test method measures mass transport from a cylindrical solidified waste form into water under conditions that accelerate leaching. Test parameters, such as the volume of leachant and the frequency at which the leachant is changed, have been optimized to eliminate experimental effects (for example, saturation effects that can complicate modeling of the net forward diffusion rate).

5.2 This test method can be used to:

- 5.2.1 Provide diffusion coefficients for waste forms;
- 5.2.2 Obtain higher fraction releases than can be achieved with expected service conditions for greater confidence in waste form properties;
- 5.2.3 Obtain measurable leachate concentrations from materials that have very low release rates under service conditions; and
- 5.2.4 Compare releases from various types of solidification agents and formulations.

5.3 Modeling the experimental observations obtained in the test allows the extrapolation of leaching results to long times and to full-scale waste forms under the following constraints:

5.3.1 Results of this test method cannot be taken to apply to releases in specific disposal environments unless tests are conducted to determine the leaching mechanism under those conditions.

5.3.2 Projections of releases require the long-term stability of the waste form, which may or may not be indicated adequately by short-term tests.

5.3.3 Extrapolations are limited to the maximum CFL obtained with the accelerated test.

5.3.4 The leaching mechanisms observed in tests conducted at elevated temperatures must be the same as the mechanisms observed in the tests run at the reference temperature.

6. Apparatus

6.1 A forced-air environmental chamber or a circulating water bath capable of controlling leachant temperatures to $\pm 1^\circ\text{C}$ 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 shall be distilled or deionized water meeting or exceeding standards for types II or III reagent water as specified in Specification D 1193.

7.2 *Containers*—Leaching containers shall be made of a material that does not react with the leachant/leachate and the specimen. It is particularly important to select materials that allow very little plate-out of radionuclides 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. This fit must be checked at test temperatures to ensure that evaporative losses are less than 1 % over 24 hours.

7.3 *Specimen Supports*—Supports for the specimens shall be made of a material that does not react with the leachant/leachate or the specimen and does not allow plate-out onto the support. The method of support should not impede leaching by obstructing the surface area of the specimen by more than 1 %. Moreover, it should not interfere with replacement of the leachate. It is often convenient to suspend the waste form from the cover of the leaching container using monofilament string.

7.4 *Sample Containers*—Containers to hold aliquots of leachate for storage prior to analysis should not allow any plate-out of radionuclides. The containers should be checked to ascertain that evaporation over long times is acceptably small.

7.5 *Stirrers*—Stirrers that are long enough to reach the bottom of the leaching containers are required. Wood or plastic tongue depressors are adequate.

7.6 *Filtration Equipment*—If particulates are present in the leachate, filtration is necessary. Tests must be conducted to ensure that the filter and the filtration apparatus do not adsorb the species of interest. The filter medium should be capable of removing particulates that are 0.45 μm in diameter. Disposable syringe filters are recommended.

8. Specimens

8.1 Right circular cylindrical specimens shall be used with a diameter-to-height ratio between 1:1 and 1:2. A convenient size is 2.5 cm diameter by 2.5 cm height. Smaller sizes should be avoided to preclude problems of mixing and producing non-homogeneous samples.

8.2 Specimens shall be representative of the full-scale solidified waste form. Particular attention should be paid to ensuring that the laboratory specimen is homogeneous. The specimens should be prepared using the same techniques as those used to produce full-scale waste forms. Curing conditions, especially the temperatures experienced by the large waste forms, should be duplicated for laboratory-scale specimens. Care should be taken to ensure that surfaces of the laboratory specimens reflect the structure of surfaces of large waste forms. This refers to surfaces that are cast against container walls as opposed to free or cut surfaces.

8.3 A minimum of three specimens should be tested at any given temperature.

8.4 The dimensions, weight, composition, and curing history shall be recorded for each specimen. Accurate determination of the source term (A_0), *that is, the amount of the species of interest in the specimen at the start of the leach test, shall be made and recorded.*

9. Procedure

9.1 *Removal of Specimen from Mold*—After removing the specimen from its container or mold, any excess material should be removed prior to weighing the specimen. The mold should be rinsed in a volume of water equal to the volume of the specimen. If the quantity of the species of interest contained in this rinse water represents more than 0.5 % of the total quantity in the specimen, the value should be subtracted from the source term (A_0).

9.2 *Leachant Volume*—The leachant volume used for each interval is $100 \times$ the surface area of the specimen as calculated below:

$$\frac{\text{Leachant volume (cm}^3\text{)}}{\text{Specimen surface area (cm}^2\text{)}} = 100 \text{ cm} \pm 2 \% \quad (1)$$

This ratio requires a large volume of water, for example, a 2.5 cm \times 2.5 cm specimen has a surface area of approximately 30 cm^2 , giving a leachant volume of 3000 mL. Specimens that are much larger than this will require volumes of water that need more sophisticated means of wastewater handling (such as peristaltic pumps for draining the containers), since large volumes are too unwieldy for pouring.

NOTE 2—Some waste form materials, such as glass, have such low leach rates that a volume-to-surface area ratio of 100 cm is not necessary. In some cases, such a large volume of leachant can make analysis challenging, even for major constituents of the specimen. Under these circumstances, the volume-to-surface area ratio may be reduced to 10 cm.

9.3 *Temperature*—For materials and formulations that have not been tested previously, leach tests shall be conducted at a minimum of three temperatures to establish that leaching increases systematically with higher temperatures. One temperature must be 20°C . The recommended maximum temperature is 50°C , which is below the threshold of anomalous releases observed so far (3). Temperatures above 50°C can be used if it is demonstrated that releases follow the trend observed at low temperatures, that is, that the leaching mechanism has not changed.

9.4 *Leachant Replacement*—Leachant replacements shall take place at the time intervals shown in Table 1. The time at which the specimen is first placed in the leachant should be noted. The leachant shall be brought to the test temperature before the specimen is placed in it.

9.4.1 If the specimen is suspended from the top of the container, the most convenient method of changing the leachant is to lift off the cover (with the specimen attached) and place it on a new container full of fresh leachant that is at the test temperature. This can be replaced in the temperature-controlled environment while the leachate in the other container is being sampled. During leachant changes, the specimen should be exposed to air for as short a time as possible.

TABLE 1 Replacement Intervals for the Accelerated Test Leachant

Interval	Incremental Time (t)	Cumulative Time (Σt)
1	2 h	2 h
2	5 h	7 h
3	17 h	1 day
4	1 day	2 days
5	1 day	3 days
6	1 day	4 days
7	1 day	5 days
8	1 day	6 days
9	1 day	7 days
10	1 day	8 days
11	1 day	9 days
12	1 day	10 days
13	1 day	11 days

9.5 *Leachate Sampling*—Immediately after the specimen has been moved to fresh water, the old leachate should be stirred thoroughly and sampled to minimize any artifacts caused by cooling (for example, precipitation). These samples will be used to determine quantities of the species of interest that have leached from the specimen during each interval and should be preserved in a way appropriate for the analytical techniques. This may require that several aliquots be taken during each sampling. If particulates are present in the leachate, it is necessary to account for the quantity of the species of interest associated with them. Two approaches can be used. One requires filtration of the leachate and subsequent analysis of both the filtrate and the particulate material on the filter. The other is to acidify the leachate to dissolve the particulates and thereby include the associated species of interest in the leachate.

9.6 *Analysis and Standards*—Analysis of species in the leachate will be conducted by standard methods. Appropriate standards will be used for analysis. If necessary, standards will be prepared to match the matrix elements in the samples. For radioactive specimens, the preferred method of analysis includes use of a standard prepared from an aliquot of the original solution (or waste) used to make the specimens. From this aliquot, dilutions are made from which the standard is taken. With this method, leachate samples can be counted relative to standards without the need for absolute standards, detector efficiencies, or decay corrections.

10. Calculations

10.1 *Incremental Fraction Leached*—Several parameters shall be calculated from the data obtained. The incremental fraction leached (IFL) is calculated by the following equation:

$$IFL = \frac{iA_n}{rA_0} \tag{2}$$

where:

iA_n = the quantity of species i observed in the leachate at any given time interval. This value is corrected for radioactive decay to the time of the beginning of the test.

A_0 = the source term, the total original quantity contained in the leaching specimen at the beginning of the test.

10.1.1 The rate of release for any interval can be calculated by dividing IFL by the elapsed time of the interval. The rate can then be divided by the surface area of the specimen to obtain the fraction released per square cm per s.

10.2 *Cumulative Fraction Leached*—Another parameter is cumulative fraction leached, which is calculated as:

$$CFL = \frac{\sum r_i A_i}{rA_0} \tag{3}$$

which is the sum of the fractions of a species of interest leached during the test. This value for each interval, plotted against cumulative time, provides a useful means of graphically comparing data to other test results and to modeling results. An example of this type of plot is shown in Fig. 2.

10.3 *Diffusion Coefficient*—This test method has an associated computer program that calculates a best fit effective diffusion coefficient based on the solution to diffusion from the finite cylinder (4, 5). The computer program calculates D_e from the test data so that the curve $\sum r_i A_i / A_0$ calculated from D_e can be plotted against time and readily compared to the data itself. This should be done for data from the accelerated test and from the reference test. If the curves of the data and the model all fall within a designated value describing “goodness of fit,” then it can be concluded that diffusion is the dominant leaching mechanism. In this case, the model can be used to project releases to long times. This projection requires the assumption that the waste form remains intact and the leaching mechanism does not change with time.

10.3.1 The computer program that accompanies this test method contains two models in addition to the two diffusion models. One model partitions the source term into a leachable component and an unleachable component. It then uses the diffusion models to determine releases from the leachable portion of the source term. The other model is used to determine if solubility constraints are limiting leaching. This

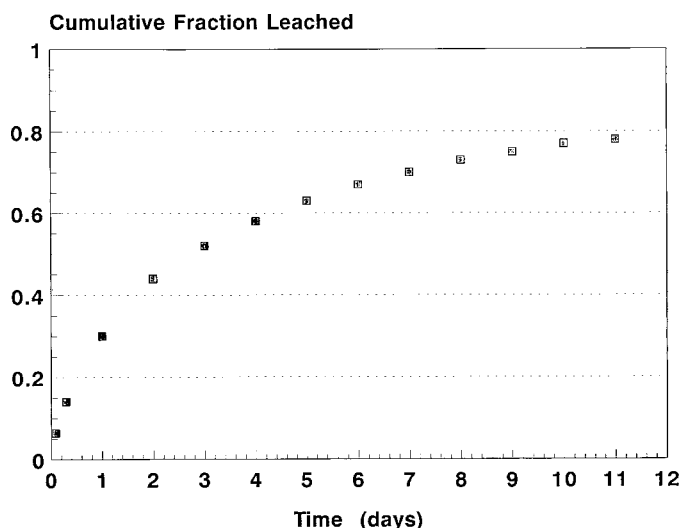


FIG. 2 The Cumulative Fraction Leached Plotted Versus Time. The Top of the Plot (CFL = 1) Represents 100 % Release