Designation: D8091 – 16

An American National Standard

Standard Guide for Impregnation of Graphite with Molten Salt[1]

This standard is issued under the fixed designation D8091; 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 guide covers procedures for the impregnation of graphite with molten salt under a consistent pressure and temperature. Such procedures are necessary if the user wishes to prepare graphite specimens for testing that have been exposed to a molten salt environment that may not necessarily represent material exposed to an operating reactor environment.

1.2 The values stated in SI units are to be regarded as standard. No other units of measurement are included in this guide.

1.3 *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]

C559 Test Method for Bulk Density by Physical Measurements of Manufactured Carbon and Graphite Articles

D7775 Guide for Measurements on Small Graphite Specimens

3. Terminology

3.1 *Definitions:*

3.1.1 *impregnation pressure (P_I), n*—the pressure of cover gas used in the impregnation.

3.1.2 *impregnation temperature (T_I), n*—the system temperature before the graphite sample has been immersed in the molten salt.

3.2 *Definitions of Terms Specific to This Standard:*

3.2.1 *Parameter D*—provides a measure of the extent of penetration of the graphite porosity by the molten salt. If there was no damage to the microstructure of the graphite during impregnation, then parameter *D* based upon open pore volume would be unity at saturation. At high impregnation pressures, closed porosity may be broken into by the molten salt and parameter *D* based upon open pore volume could have values greater than unity. It is therefore useful to express parameter *D* in terms of both open pore volume and total (open and closed) pore volume.

3.2.1.1 *Discussion*—Parameter *D* is based upon open pore volume (typically applicable at low impregnation pressures) and is calculated as follows:

$$D_o = \frac{W_2 - W_1}{\rho V_o} \quad (1)$$

where:

W_1 = the weight of the graphite sample before impregnation,
W_2 = the weight of the graphite sample after impregnation,
ρ = the density of molten salt, and
V_o = the open pore volume in the sample.

3.2.1.2 *Discussion*—Parameter *D* based upon total pore volume (typically at high impregnation pressures) is calculated as follows:

$$D_t = \frac{W_2 - W_1}{\rho V_t} \quad (2)$$

where:

V_t = the total pore volume in the sample.

4. Summary of Guide

4.1 This guide provides guidance on the impregnation of graphite with molten salt. The guide gives the various factors which need to be considered to perform the impregnation procedure. These include pretreatment of graphite samples, immersion of graphite in the molten salt, safe handling of the molten salt, and selection and control of the impregnation pressures and temperatures.

5. Significance and Use

5.1 The molten salt reactor is a nuclear reactor which uses graphite as reflector and structural material and fluoride molten salt as coolant. The graphite components will be submerged in the molten salt during the lifetime of the reactor. The porous structure of graphite may lead to molten salt permeation, which

[1] This guide is under the jurisdiction of ASTM Committee D02 on Petroleum Products, Liquid Fuels, and Lubricants and is the direct responsibility of Subcommittee D02.F0 on Manufactured Carbon and Graphite Products.

Current edition approved Dec. 1, 2016. Published January 2017. DOI: 10.1520/D8091-16.

[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.

can affect the thermal and mechanical properties of graphite. Consequently, it is important to assess the effect of impregnation of molten salt on the properties of the as-manufactured graphite material.

5.2 The purpose of this guide is to report considerations that should be included in the preparation of graphite samples representative of that after exposure to a molten salt environment. The degree to which the molten salt will infiltrate the graphite will depend upon a number of factors, including the type of graphite and the type and extent of porosity, the properties of the molten salt, the impregnation pressure and temperature, and the duration of the exposure of the graphite to the molten salt.

5.3 The user of this guide will need to select impregnation parameters sufficiently representative of those in a molten salt reactor based on parameters provided by the designer. Alternatively, the user may select a standard set of impregnation conditions to allow comparisons across a range of graphites.

5.4 This guide is not intended to be prescriptive. A typical apparatus and associated procedure is described. Some indication of the sensitivity of the procedure to graphite type and impregnation conditions is given in He, et al.[3]

5.5 There are four major practical issues that must be addressed during the impregnation process:

5.5.1 The density of molten salt is greater than that of graphite. A specially designed tool is required to submerge graphite samples in the molten salt during the impregnation process.

5.5.2 Some molten salts (for example, FLiBe) are poisonous and it is therefore necessary to provide containment by performing procedures within a glove box.

5.5.3 The graphite must be kept away from air to avoid oxidation at high temperature. This can be achieved by performing the impregnation process within a glove box with a controlled atmosphere.

5.5.4 Pressure control of the molten salt can be difficult to achieve. A specially designed autoclave is needed to hold the sample and molten salt.

5.6 In order to assess the quantity of molten salt in the graphite, parameter D is used as a variable in measuring the mechanical and thermal material properties. For a low impregnation pressure, the parameter D_o is the ratio of salt volume to open pore volume. For a high impregnation pressure, the parameter D_t is the ratio of salt volume to total pore volume. The saturated value of D_o can be greater than 1 when the molten salt impregnation takes place at high pressure. It is postulated that the internal microstructure of graphite has been damaged by the impregnation pressure and some closed pores have been opened. In this case, the parameter D_t is more appropriate to represent the impregnation process.

6. Apparatus

6.1 *Autoclave:*

6.1.1 Since fluoride molten salts are toxic and a small amount of water can significantly affect the wetting behavior of graphite, the conditions for the procedure should be strictly controlled during molten salt impregnation. The schematic diagram of an impregnation setup is shown in Fig. 1.

6.1.2 An autoclave is used as a sealed container and should meet a leakage rate <0.25 %/h. To avoid corrosion by molten salt, the autoclave should be made of a high temperature and corrosion-resistant alloy.

6.1.3 A graphite crucible is placed inside the autoclave to hold the molten salt. There should be enough clearance between the wall of autoclave and graphite crucible to prevent damage of the graphite crucible due to differences in thermal expansion.

6.1.4 The graphite samples must be secured in a sample holder. For example, the sample holder might comprise two graphite plates with the samples placed between the plates (see Fig. 2).

6.1.5 In this example, the sample holder is fixed to a sample rod. The sample rod can move up and down inside the autoclave to ensure that the entire graphite sample holder is submerged in the molten salt during the procedure.

6.1.6 High purity inert gas (>99.99 %) is pumped into the autoclave and is used to adjust the impregnation pressure of molten salt. A pressure gauge should be installed to measure the gas pressure inside the autoclave.

6.2 *Electric Furnace*—An electric furnace with temperature control is used to heat the autoclave. The furnace should be able to provide the desired temperature of T_1 °C ± 20 °C.

6.3 *Analytical Balance*—An analytical balance is used to measure the weight of the graphite sample before and after the impregnation procedure. The error of the scale should be less than ±0.1 mg.

6.4 *Glove Box:*

6.4.1 All the testing equipment, including the autoclave, the electric furnace, and the analytical balance should be placed inside a glove box. A hoist system may be installed inside the glove box to aid lifting the autoclave and the graphite crucible.

6.4.2 The water and oxygen contents inside the glove box should be less than 1 ppm to prevent external contamination or

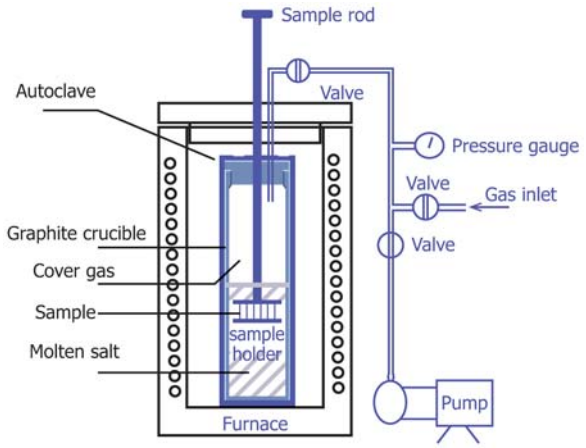

FIG. 1 Schematic Diagram of the Procedural Setup

[3] He, Zhoutong, et al., "Molten FLiNaK Salt Infiltration into Degassed Nuclear Graphite Under Inert Gas Pressure," *Carbon*, Vol 84, 2015, pp. 511–518.

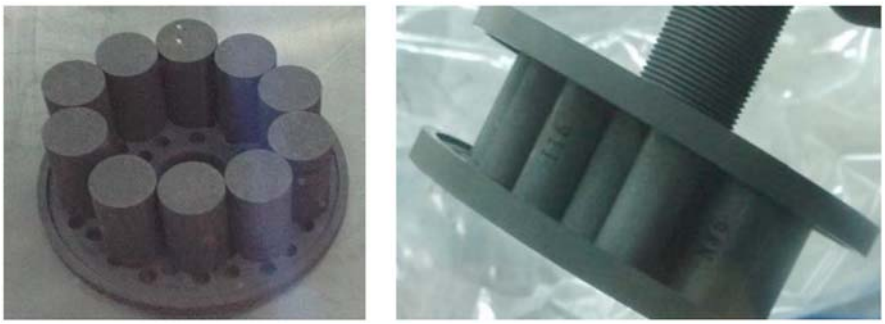

FIG. 2 Sample Holder: Graphite Plates and Sample Rod

oxidation. The glove box is maintained at negative pressure to prevent release of toxic salt vapor/dust from the glove box.

6.4.3 The glove box can be divided into two zones. One zone may be used for temporary storage of graphite samples and the weighing of graphite samples; the other contains the furnace and the impregnation equipment.

7. Procedure

7.1 Graphite specimens must be individually marked (for example, surface laser etching) or tracked.

7.2 For regular specimen geometries, the volume should be evaluated from the dimensions measured in accordance with Test Method C559. For non-regular specimen geometries, the volume must be evaluated by another method such as immersion method (Guide D7775).

7.3 The graphite crucible and graphite sample must be dried in an oven at a temperature above 110 °C for 2 h to remove any sorbed water.

7.4 Measure the open pore volume, V_o, of the graphite sample by using helium pycnometry.

7.5 The weight of graphite specimen is measured at room temperature by using the analytical balance inside the glovebox and recorded as W_1.

7.6 Calculate the total pore volume, V_t, from the difference between the sample bulk density and the graphite crystal density taken as 2.24 g/cm^3.

7.7 The autoclave should be preheated at a temperature above 110 °C for at least 2 h.

7.8 Molten salt is poured into the graphite crucible before the impregnation such that the level is no more than half height of the graphite crucible. The graphite crucible is placed into the autoclave.

7.9 The specimen holder containing the graphite specimens is placed inside the graphite crucible and held by the sample rod above the molten salt level.

7.10 The autoclave flange is sealed. The temperature of the autoclave is increased to the desired impregnation temperature. The sample holder containing the graphite specimens is immersed into the molten salt by pushing the sample rod after the impregnation temperature has been reached.

7.11 High purity inert gas is used to adjust the impregnation pressure inside the autoclave. Once the impregnation pressure has been reached, the impregnation start time is recorded and the duration measured.

7.12 The desired impregnation pressure and temperature should be kept constant over the whole impregnation period.

7.13 Retrieve the specimen after impregnation:

7.13.1 Once the target impregnation time has been reached, withdraw the sample holder to a position above the molten salt level.

7.13.2 Turn off the furnace and allow the autoclave to cool down to room temperature.

7.13.3 Release the pressure valve and open the autoclave flanges to retrieve the specimens.

7.13.4 The weight of each graphite specimen is measured at room temperature by using the analytical balance inside the glove box and recorded as W_2.

NOTE 1—After the test, some solidified salt may have been deposited on the surface of graphite specimen. The user may carefully remove the salt on the surface before the measurement of specimen weight. Confirm that the individual dimensions of the sample are within 0.5 % of the original dimensions.

7.13.5 The specimen is stored in the temporary storage zone of the glove box for use in subsequent tests.

8. Test Data Record

8.1 The actual impregnation time, the specimen dimension, the specimen weight before and after the impregnation, the impregnation pressure, and temperature should be recorded.

8.2 The D parameters should be calculated as in Eq 1 or Eq 2 and should be recorded.

9. Report

9.1 The report should include all the test data records together with relevant information on graphite grade designation, lot number, billet number, orientation, and location. Information on the molten salt shall be provided.

10. Keywords

10.1 graphite; impregnation; molten salt