Test Methods for Cementitious Seal Materials for Borehole Disposal-17352

E. G. Ferreira,* J. T. Marumo,* R. Vicente,* C. A. Langton, A. J. Duncan, and M. R. Williams

Savannah River National Laboratory, Savannah River Site, Aiken, SC 29808
*Nuclear and Energy Research Institute, IPEN-CNEN/SP, Sao Paulo, Brazil
michael.williams@srnl.doe.gov

ABSTRACT

The Nuclear and Energy Research Institute (IPEN-CNEN/SP) in Sao Paulo-Brazil, is considering borehole disposal of disused sealed radioactive sources (DSRS), which were used in smoke detectors, lightning arrestors, radiotherapy, industrial gauges, irradiators, and other applications. The current IPEN concept of borehole is a modified BOSS system developed by the International Atomic Energy Agency (IAEA). It differs from the IAEA's design in that the disposal zone is below 300 m, and the DSRS are placed directly in the disposal packages constituted of stainless steel containers with 1.5 L capacity. A borehole drilled through sediment and granite, stabilized with stainless steel casing and cement grout is proposed for the conceptual model. Standard cement-material test methods and performance specific test methods, including accelerated aging methods, are needed for modeling the performance of the candidate cementitious grouts. A cross-disciplinary team from the USA Savannah River National laboratory is providing technical assistance to IPEN in this performance assessment.

INTRODUCTION

A cross-disciplinary team from several SRNL departments is providing technical assistance to the Brazilian Nuclear and Energy Research Institute (IPEN-CNEN/SP) in devising a strategy for safe, permanent disposal of disused sealed radioactive sources (DSRS) which were used in smoke detectors, lightning arrestors, radiotherapy, industrial gauges, irradiators, and other applications within Brazil. DSRS are considered radioactive waste and IPEN is proposing their disposal in a deep borehole.

The current IPEN concept of borehole is a modified BOSS system developed by the Nuclear Energy Corporation of South Africa under supervision of the Atomic Energy Agency (IAEA). It differs from the IAEA's design in that the borehole is deeper, the disposal zone is below 300 m, versus 30 m of the BOSS, and will be used to dispose of a much larger number of DSRS. The sources are disposed of in waste packages (WP) that are cylindrical stainless steel containers with approximately 1.5 liters of storage capacity. The IPEN concept also differs from the IAEA design in that the DSRSs are placed directly in the WP rather than in a capsule imbedded in grout inside the WP. This allows for disposal of more sources per WP.

A borehole drilled through sediment and granite and stabilized with stainless steel casing is proposed for the conceptual model. Cement grout will be placed: (1) between the outer surface of the casing and above the disposal zone to physically stabilize the casing and to seal saturated formations; and (2) between the inner surface of the casing and the WP again to provide physical stabilization and seal and to provide a solid spacer between WP. Ideally this spacer will also provide a level surface on which the overlying WPs can be positioned.

The Brazilian modified BOSS case is significantly different from the IAEA design and generic safety case in that capsules are not used within the waste package to contain the disused sealed radioactive sources (DSRS) and the borehole is significantly deeper at 300 meters below the surface. Thus, requiring accelerated testing at least to the rigor of the Generic Safety Assessment of the IAEA BOSS GSA.

Because borehole performance is required for thousands of years, accelerated testing coupled with an understanding of aging/degradation mechanisms is needed to evaluate the evolution of the grout material under borehole conditions. Accelerated tests for borehole grouts can utilize increasing rate controlling variables such as temperature, and concentrations of corroding chemicals provided that the aging/degradation processes at accelerated stress levels are the same as that occurring under the expected service conditions. Micro- and nano-scale characterization of surfaces and interfacial regions of grout coupons/samples subjected to accelerated conditions is proposed as a means of further enhancing the understanding of mechanisms and rates. Scanning and transmission electron microscopy and associated chemical analysis coupled and micro-tomography of surfaces regions and interfacial regions are proposed for material comparisons and mechanistic understanding of grout evolution in a borehole environment. This information coupled with thermodynamic models and assessment of natural analogues is needed to assess grout materials in borehole environments and time frames for which engineering and field data are not available.

Functional performance must address both fresh properties needed for placement the borehole application and cured properties. For boreholes up to a few hundred meters deep, standard drilling technology and grout rheology and tests are expected to be adequate for installing and stabilizing the borehole and casing. These requirements are specific to the borehole stratigraphy, diameter, etc.

Where appropriate, literature data can be used for an initial assessment of cured grout performance, refine grout designs, and to prioritize potential degradation scenarios. Understanding the effects of (1) temperature over the range from ambient downhole, e.g., about 36 °C at about 300 m to the maximum expected due to energy imparted from radioactive decay (up to 100°C for the IPEN DSRS inventory and WP distributions in the borehole); (2) pressure; (3) ground water chemistry; (4) radiation dose; and (5) time (aging) on the grout-casing and grout-WP seal properties.

The first step in identifying test methods was to define the functions and requirements of the cement grouts. Cured grout requirements include: physical properties, i.e., compressive strength, dimensional stability, porosity, density, thermal conductivity, thermal expansion, gas and water permeability. In addition, borehole specific tests are needed to evaluate and compare candidate materials under simulated borehole conditions. Additional performance tests are needed to evaluate borehole grouts with respect to: (1) physically sealing the rock formation-casing and the casing-WPs contacts/interfaces, (2) minimizing corrosion of the stainless steel casing and WPs, and providing chemical retardation of long-lived radionuclides that may be released if the WPs are breeched.

GENERIC SCENARIO OF THE ENGINEERED BARRIER SYSTEM

The Waste Form or Waste Package must not contain liquids, particulates, chemically reactive or combustible materials and the materials/components of the Engineered Barrier System (EBS) are designed to provide-assuming anticipated processes and events substantially complete containment of the low level and intermediate level DSRS radioactive material for 360 years [1] at a minimum. The EBS should be designed to work in combination with the borehole natural barriers, such that the performance assessment of the EBS demonstrates conformance to the annually reasonable expected individual dose protection standard of the BOSS design [2].

The technical information and test data regarding the actual behavior of waste forms and materials that are actually used in the EBS and exposed to repository conditions for such long periods of time will not be sufficient to develop fully validated models in the classical sense. Rather the necessarily short term accelerated data acquisition, and use of the data in formulating reliable long-term predictive models is to be used to support the design, performance assessment, and even the selection of waste package/EBS materials (e.g., low confidence in a degradation model may justify the selection of alternative EBS barrier such as schedule 80 stainless steel 316-L pipe versus schedule 40 in the BOSS design).

The ASTM Standard C1174 and others provide standard practice and methods for making useful predictions of long-term behavior of materials from such sources as test data, scientific theory, and analogs for geological disposal of radioactive waste. The EBS environment of interest is that defined by the natural conditions (for example, minerals, moisture, biota, and mechanical stresses) as modified by effects of time, repository construction and operations, and the consequences of radionuclide decay, for example radiation damage, heating, and radiolysis effects. Environmental conditions associated with both anticipated and unanticipated scenarios will be considered from the initial concepts and investigations of the repository site, candidate EBS, and the Waste Package materials proposed based on the geologic environment and conceptual design. The IPEN proposed dimensions of the EBS for the disposal of DSRS is shown in the Figure 1.

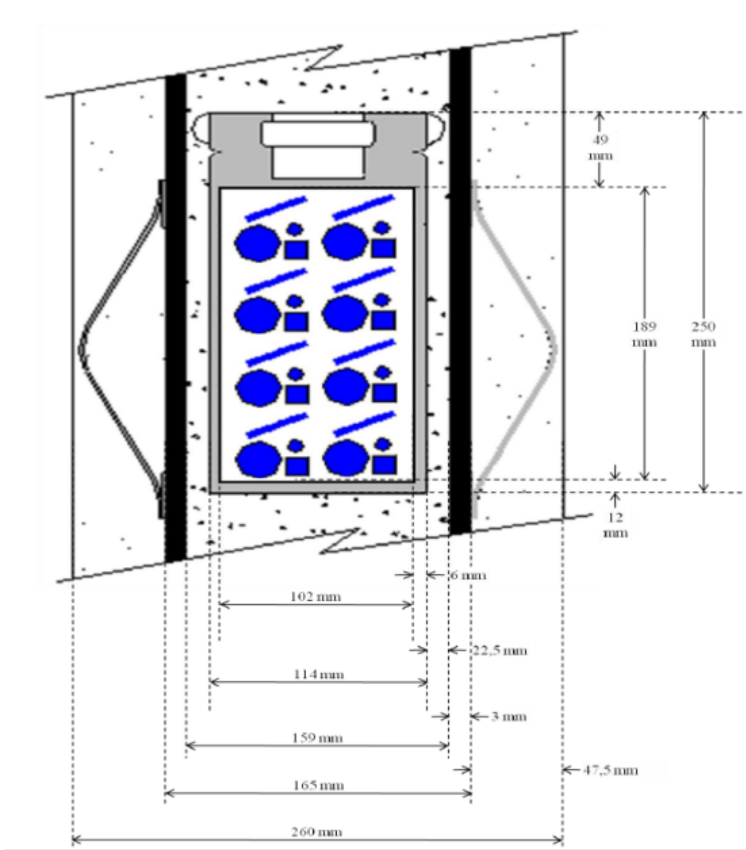

Figure 1. Dimensions of the components of the IPEN proposal

TESTING APPROACH FOR GEOLOGICAL DISPOSAL OF RADIOACTIVE WASTE

Testing is required to establish whether materials proposed will perform the function for which they were intended, i.e., effectively contain radionuclides for the containment period. Tests will be conducted to develop and parameterize more detailed models that can be used to predict materials behavior over time periods longer than tested directly. Tests conducted over a comparatively short period, for example, less than 20 years, will be used to support development of predictive behavior models for the response of the materials to the repository environment for the regulatory compliance period. The testing program will address the development, validation, and confirmation of these models. Materials testing programs are designed with the goal of supporting the development and application of materials behavior models, as well as the minimizing of the uncertainties in the test data, the models, and the use of the models in calculations of long-term behavior in the IPEN-CNEN/SP EBS.

ASTM C1174 standard practice establishes categories of testing according to the information they provide and how it is used for model development and use [3]. These tests may include, but are not limited to the following:

- Attribute tests to measure intrinsic material properties,
- Characterization tests to measure the effects of material and environmental variables on behavior,
- Accelerated tests to accelerate alteration and determine important mechanisms and processes that can affect the performance of waste packages and EBS materials,
- Service condition tests to confirm the appropriateness of the model and variables for anticipated disposal conditions,
- Confirmation tests to verify the predictive capacity of the model, and
- Tests or analyses performed with analog materials to identify important mechanisms, verify the appropriateness of an accelerated test method and to confirm log-term model predictions.

A single test method can simultaneously serve more than one function. For instance, a single test procedure could serve as both a characterization test and as an accelerated test depending on the test parameters that are used. These tests may be applied to analog materials to provide insight into long-term mechanisms of alteration.

Attribute Tests: Intrinsic materials properties, such as chemical composition and phases, density, specific surface area, thermal properties, grain size, hardness, tensile strength, radiation effects, etc. can be usually be obtained from literature sources of material properties. Data obtained from attribute tests are used for initial material selection and for monitoring changes as a function of time and environmental conditions. Material characterization data are used to parameterize long term predictive performance models. It is expected that most of the required information concerning the DSRSs material attributes will be available in the literature.

Characterization Tests: Characterization tests provide a mechanistic understanding for important processes of material alteration expected in the repository environment and measuring model parameter values. These tests are used to establish both the suitability and the basic form of the behavior model. Test conditions may depart significantly from the expected repository conditions, and so it may be necessary to investigate the sensitivity of the alteration mechanisms to variations in the values of a particular test parameter. Examples of these tests include anodic polarization tests, radionuclide solubility measurements, phase changes detected x-ray diffraction analyses, etc. which are used to characterize material evolution as a function of time and environmental conditions.

Accelerated Tests: Accelerated tests are used to address numerous needs including:

- Alter the state of a material in a short time to simulate long time exposures, and thereby produce artificially "aged" materials.
- Measure the rates of slow reactions provided that the rate equation is known,
- Promote the formation of a sufficient quantity of alteration phases for identification and characterization,
- Promote the approach to solution saturation, and
- Age the solution that contacts the material to represent conditions that may occur after long reaction progress.

An accelerated test typically results in increasing the rate of an alteration mechanism or the extent of reaction progress, when compared with expected service conditions, without changing the basic alteration mechanisms of the alteration mode under investigation. Therefore, some knowledge of the aging or degradation mechanisms that are operative under in-service conditions is needed for the design of accelerated test data.

Processes may be accelerated by increasing one or more external test parameters including: temperature, pressure, humidity, radiation dose, porosity, material surface area, particle size, or roughness, solution volume and flow rate, solute concentrations, electrical current, mechanical stress, microbial activity, etc., relative to the in-service values. If the alteration mechanism that is operative in the accelerated test differs from that which is operative under the in-service conditions or changes over a range of accelerating test conditions, the accelerating test conditions must be re-evaluated.

Confirmation Testing: Constructing, instrumenting, monitoring, and evaluating individual elements, multiple elements or small- or full-scale mock-up designs prior to actual to constructing the actual disposal system can provide the opportunity to refine the design prior to implementation. In one of a kind or new disposal system designs confirmation testing is performed to reduce unknown issues and reduce overall project risks. Instrumentation of the actual disposal site is typically implemented to monitor and confirm performance during an intimal period of institutional control after the site / borehole is closed.

Analogue Material: Analyses of analogs, both natural [5] and man-made [6], are useful input to long-term predictions concerning phase evolution of materials used in the disposal environment. Examples of areas in which analogue systems can provide improved understanding and predictive capabilities include:

- Phase evolution of metals, cement based materials, geologic media and contents of the waste package in the disposal environment over time.
    - The role of redox processes in mobilizing and/or stabilizing radionuclides the speciation and solubility of radionuclides in groundwater
    - The downstream retardation processes affecting remobilized radionuclides, including diffusion into host geology porous rocks

- Use of natural decay series disequilibria to estimate the longevity of various mobilization and deposition processes in the EBS-host geology environment.

IPEN WASTE PACKAGE AND ENGINEERED BARRIER MATERIALS

The conceptual design of the IPEN borehole disposal concept is illustrated in Figure 2. The materials identified for the components of the design are listed below:

- Waste form or Waste Package i.e. sealed sources and foils (hundreds per WP) packed into a 4 inch 316L schedule 40 stainless steel segment of pipe with an inert dry atmosphere capped on the top and bottom of IAEA BOSS design

- Borehole casing i.e. 316L stainless steel pipe

- Casing grout as specified in the IAEA BOSS design [4]

- Borehole back fill grout as specified in the IAEA BOSS design [4]

- Geological formation w/wo brine[1].

[1] Measurement of dissolved oxygen in ground water is essential to understanding and predicting the corrosion rate of metal components in the borehole. Details of measurement methods are provided elsewhere [7, 8].

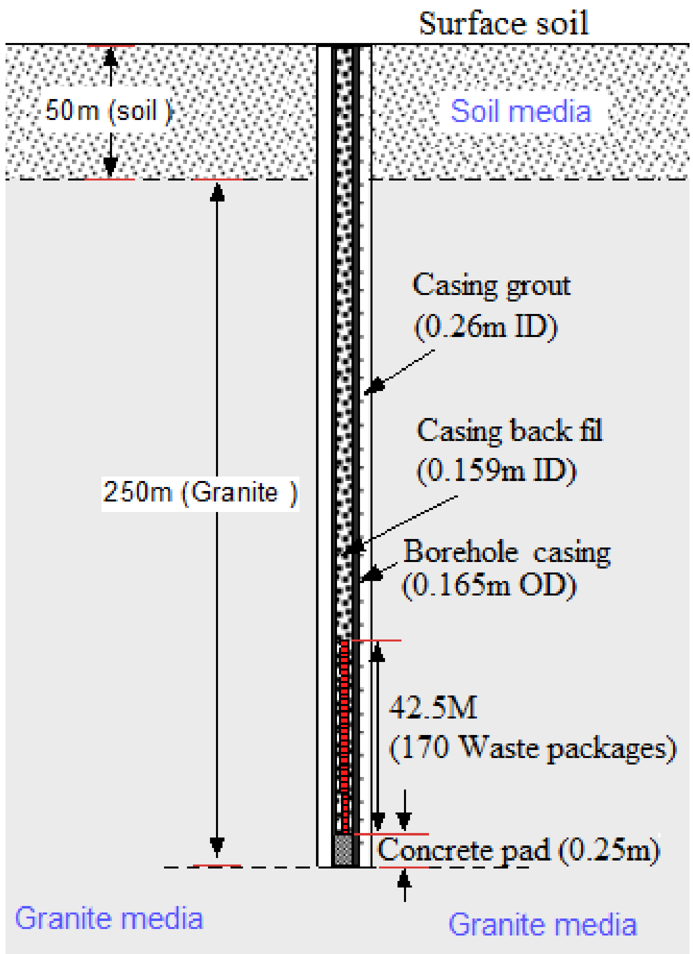

Figure 2. IPEN Geological Borehole Predictive Modeling Boundaries Containing 170 Waste Packages

Establishing the functions of the materials, components, and systems in the IPEN borehole disposal conceptual design is necessary to develop a test program to evaluate the individual features and the integrated performance.

Table 1. Proposed functions of the components in the Borehole Disposal Concept

Component	Function	Examples of Parameters	Information Needed
Environmental / geologic media	Limit access of human, animal, plant intrusion into the waste zone Limit spread of radionuclides into aquifers and near	Borehole siting criteria specific to regulatory and programmatic needs including performance time for geologic isolation.	Waste zone mineralogy, temperature, pressure; porosity, hydraulic conductivity, ground water chemistry

	surface environment Limit impact of disposal on environmental media		including dissolved oxygen and flux
Casing	Physically stabilize borehole during drilling and disposal	Pipe thickness and material	Material and pipe properties for casing speciation and durability during operation and institutional control
Casing grout (between casing and environment) Drilling mud	Stabilize borehole during drilling and casing placement Drilling mud needed to lubricate and keep the borehole open during construction is identified but not considered here	Grout fresh properties for pumping and placement	Consistency as a function of temperature, pressure and working time, Set time, strength, bonding to metal and rock, interaction with drilling mud and residue, compatibility with groundwater/brine, physical, chemical, and mineralogical durability over time in disposal environment.
DSRS Canister (Modified BOSS conceptual design)	Provide leak tight package for a time after which radioactive decay has rendered the sources no longer a concern.	Corrosion rate of material and welds in borehole-DSRS environment.	Thermal transients in WP and rock formation, Chemistry of ground water chemistry and concentrated groundwater, Effects of radiation, Pressure evolution inside WP
	Consider machined refractory spacer to minimize effects of welding heat on sources	Effect of welding process (heating) on DSRS in container Weld integrity of aged canisters in borehole	Leak tight tests after simulated aging, i.e., (temperature pressure, corrosion, exposure to radiation) Xenon tracer gas in canisters coupled with gas

			chromatograph leak detection
DSRS Canister atmosphere	Minimize void volume minimize chemical reactions inside the canister. Evaluate filling canister with inert gas and /or inert solid	Void volume Void space material (gas and or inert solid)	Effect of WP contents on long term performance (gas generation, chemical compatibility, expansion due to corrosion products
Grout between casing and canister	Physically stabilize WPs in the casing. Chemical buffering (pH) of corrosive conditions	Same parameters as casing grout but placement techniques may differ	Same as casing grout but
Leak detection instrumentation	Detect release of radionuclides from WM	Detection of tracer gas Other as specified	Concentration of tracer or actual waste species at surface or subsurface locations
Additional plugs and seals	Provide leak tightness to gas, water, and/or soluble species	Mechanical plug performance parameters	Plug specific

ACCELERATED TESTING FOR BOREHOLE DISPOSAL CONCEPT

Accelerated testing can be used to investigate aging mechanism and rates for chemical, physical, mechanical, thermal, and biological characteristics. Accelerated durability testing may take a long time (months and years) before relevant data are provided. An accelerated test must result in increasing the rate of an alteration mode or the extent of reaction progress, when compared with expected service conditions, without changing the basic alteration mechanisms of the alteration mode under investigation. Therefore, some knowledge of the mechanism that is operative under in-service conditions is needed for the design of accelerated test data.

Processes may be accelerated by increasing various test parameters, including temperature, pressure, material surface area, particle size, or roughness, solution volume and flow rate, solute concentrations, humidity, etc., relative to their in-service values. If the alteration mechanism that is operative in the accelerated test differs from that which is operative under the in-service conditions or changes over a range of accelerating test conditions, the accelerating test conditions must be re-evaluated.

Because of the large number of components and independent variables in the borehole disposal scenario, e.g., temperature, pressure, radiation dose, mechanical stress, groundwater chemistry, and multiple component materials including the DSRSs, synergistic and/or competing effects must be considered.

The range over which these variables can be increased for accelerated testing programs depends on the variable. For example, the range for temperature is limited for hydrated cementitious materials which are formulated for specific temperature and pressure conditions. In the IPEN borehole scenario the design range is between 35 and 95 °C, for an empty 300 m deep borehole and a borehole in which DSRSs are disposed, respectively [9]. The range over which ground water and be concentrated can most likely be increased over one or more orders of magnitude. The range over which radiation dose rate needs to be increased in order to produce a conservative approximation of the effect of the maximum dose on the materials.

Given that IPEN has identified the effect of gamma radiation to be an important parameter in the performance of the WP with respect to thermal transients, radiolytic gas generation of the hydrated materials, and radiation effects on the DSRS themselves, the consequences of gamma dose which in itself is accelerated through dose rate must be incorporated into the overall test plan.

Unique accelerated tests are required to evaluate the effects of gamma irradiation of a multicomponent disposal configuration which includes groundwater, stainless steel, DSRSs, hydrated Portland cement based grouts in contact with stainless steel and granite. The gamma source available to IPEN has limited capacity and sample size is another limiting issue.

Sample size limitation in the Co-60 gamma irradiator and the multicomponent nature of the system has focused attention on micro- and nano-scale characterization of surfaces and interfacial regions as the accelerated testing approach for identifying mechanism and estimating alteration rates. A three component sample consisting of cement grout-stainless steel-granite in an aqueous environment with ground water and dry environment without ground water are proposed and will be evaluated. The relationship between these materials is in the IPEN borehole disposal concept is illustrated in Figure 3. Scanning and transmission electron microscopy and associated chemical analyses coupled with comparisons of cementitious material evolution in borehole environments

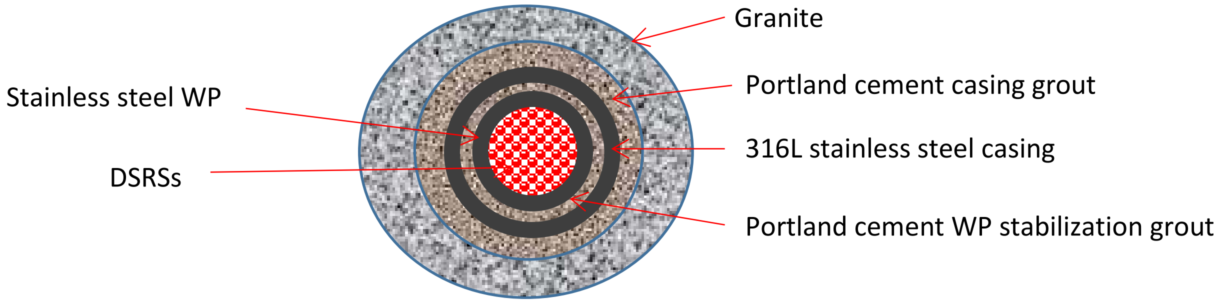

Figure 3. Cross section of multi component sample for immersion in borehole water and irradiation in Co-60 source.

DISCUSSION

Test methods have been presented to support the Brazilian Borehole disposal concept for DSRS, based on IAEA BOSS modified Waste Packages utilizing existing well technology from the oil and gas industry. Applying the results of material properties and focused micro-scale tests to predictive modeling of the borehole can be used to meet regulatory requirements in Brazil and/or the IAEA. SRNL has collaborated with the IPEN-CNEN/SP organization bringing extensive experience involving the study of cementitious radioactive waste forms at the Savannah River Site and testing for other radioactive waste locations in the DOE complex.

Confirmation tests are recommended to further support and validate acceptable performance of the as designed and as-constructed borehole disposal site. During the pre-closure period of the repository, testing (particularly in-situ testing) is recommended to validate key aspects of materials behavior for the Waste Package and EBS. Tests begun as service condition tests can be extended, so as to serve the purpose of confirming the predicted materials alteration behavior.

Testing the use of a xenon tracer gas added to the inert gas fill of the WP to confirm leak tightness of the WPs should be evaluated as a potential long-term method of evaluating performance in the geologic setting.

CONCLUSIONS

Sufficient test methods have been presented to provide data in support of Brazil Borehole behavioral model development and prediction of performance over the regulatory required period of performance. The non-radioactive and radioactive test methods applicable to the Brazil Borehole Disposal of Low-Level and Intermediate Level DSRS material can also be applied to the DOE Disposal of cesium and strontium radioactive capsules in the Waste Encapsulation and Storage Facility at Hanford as well as Spent Nuclear Fuel utilizing Borehole technology.

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