Contract No:

This document was prepared in conjunction with work accomplished under Contract No. DE-AC09-08SR22470 with the U.S. Department of Energy (DOE) Office of Environmental Management (EM).

SRNL-STI-2016-00157

Proceedings of the ASME 2016 Pressure Vessels & Piping Conference
PVP2016
July 17-21, 2016, Vancouver, BC, Canada

PVP2016-63884

CHLORIDE-INDUCED STRESS CORROSION CRACK GROWTH UNDER DRY SALT CONDITIONS – APPLICATION TO EVALUATE GROWTH RATES IN MULTIPURPOSE CANISTERS

Robert L. Sindelar
Materials Science and Technology
Savannah River National Laboratory
Aiken, SC 29808, USA
robert.sindelar@srnl.doe.gov

Joe T. Carter
Nuclear Programs
Savannah River National Laboratory
Aiken, SC 29808, USA
joe.carter@srnl.doe.gov

Andrew J. Duncan
Materials Science and Technology
Savannah River National Laboratory
Aiken, SC 29808, USA
andrew.duncan@srnl.doe.gov

Brenda L. Garcia-Diaz
Materials Science and Technology
Savannah River National Laboratory
Aiken, SC 29808, USA
brenda.garcia@srnl.doe.gov

Poh-Sang Lam
Materials Science and Technology
Savannah River National Laboratory
Aiken, SC 29808, USA
ps.lam@srnl.doe.gov

Bruce J. Wiersma
Materials Science and Technology
Savannah River National Laboratory
Aiken, SC 29808, USA
bruce.wiersma@srnl.doe.gov

ABSTRACT

The conditions of continued dry storage of the spent nuclear fuel in multipurpose canisters render the canisters, a component for confinement in dry storage cask systems, susceptible to chloride-induced stress corrosion cracking (SCC). The requisite conditions involve deposits of chloride-bearing marine salts and/or dust that deliquesce on the external surface of the cooling canister to create brine at weld residual stress regions. The subcritical crack growth rate at this "dry salt" condition, investigated by several researchers, has shown a relatively slow growth rate compared to chloride-cracking under aqueous conditions. A new SCC growth rate test specimen configuration has been developed to enable an initially dried salt assemblage to deliquesce under temperature and humidity conditions to load the fatigue pre-cracked, wedge-opening-loaded (WOL) specimen with the brine and enable measurements of crack growth rate (da/dt) under falling stress intensity factor, K_J, conditions. The application of the results to a canister weldment with a residual stress profile to predict crack extension in time is described. The results are evaluated in terms of development of acceptance standards for this type of flaw, should SCC be identified and characterized through inservice inspection (ISI).

INTRODUCTION

Many dry cask storage systems for spent nuclear fuel are of a dry shielded canister (DSC) design that includes a welded construction (and weld-sealed) austenitic stainless steel multipurpose canister that is placed within a concrete overpack and stored on an outside pad. Tests and experiments have shown that stainless steels typical of those used for the canisters (e.g. 304, 304L, 316, or 316LN) are susceptible to stress corrosion cracking with chloride-containing salts [1-5]. The degradation scenario is that chloride-rich salts and other debris deposit on the canister through atmospheric convection over time and, if the local relative humidity at the canister surface is "high," deliquesce to form a moist salt or brine. The wetted chloride salt deposit is aggressive to stainless steel for pitting and cracking corrosion, particularly at weld heat-affected-zone regions with sensitized microstructures and residual stresses.

No observation or evidence of pitting or cracking attack of canisters due to corrosion has been observed to date. Nevertheless, the U.S. Nuclear Regulatory Commission is requiring that the dry storage licensee institute an Aging Management Program (AMP) to demonstrate that the structural integrity and confinement function of the canisters is maintained for extended (re-license) storage conditions in consideration of the potential for SCC [6]. Consensus rules for inservice inspection of the canisters are being developed, and these rules can serve as the foundation for the canister AMP. A Task Group was constituted in 2015 under Section XI, Division 1 of the ASME Boiler and Pressure Vessel Code to draft a new Code Case N-860, "Examination Requirements and Acceptance Standards for Spent Nuclear Fuel Storage and Transportation Containment Systems."

Flaw acceptance criteria are needed for the disposition of the inspection results should relevant indications be found during the examination. Disposition of flaws that would be identified as stress corrosion cracks, and characterized or sized using visual, surface, and volumetric examination methods requires, as inputs to the technical bases, both evaluation of flaw stability under design basis loading cases, and also allowance for flaw growth between inspection intervals. Even if shown as mechanically stable, through-wall flaws would not be acceptable due to their challenge to the confinement function of the canister.

The stability of flaw postulates, both parallel and perpendicular to circumferential (girth) and longitudinal (axial) canister weldments has been reported in several studies. Canister-specific dimensions, material properties, and loading conditions, were applied in flaw stability analyses using ASME SCXI [7] for the limit-load method [8], and using API/ASME Level 2 assessment [9] for the failure assessment diagram (FAD) method [10] with welding residual stress profiles estimated from the ASME/API methodologies. Work presented at this conference [11] compares the flaw tolerance in stainless steel piping (Nominal Pipe Size 32-inch with 0.75-inch wall thickness) using Nonmandatory Appendix C (Article C-5000 limit load criterion and also Article C-6000 elastic-plastic fracture mechanics criterion), and Nonmandatory Appendix H (FAD) methods under ASME SCXI. The common conclusion is that the static, low loading conditions of the austenitic stainless steel canister is highly tolerant of flaws. However, it is observed that with the residual stress included as part of the loading, and with the consideration of elastic-plastic fracture mechanics through the FAD formulation, the instability crack sizes are significantly reduced from those based on the net section yielding limit load failure criterion [10]. The instability crack sizes (length and depth) for more realistic, semi-elliptic surface crack configurations were also presented at this conference [12] using R6 Option 1 FAD procedure [13] and the through-wall welding residual stress distributions used in Reference [10].

Results vary widely among the available literature on crack growth rate (CGR) testing under conditions relevant to the deliquescing salt conditions on the canister. This appears to be due, in part, to the difficultly in establishing a crack growth front in a test specimen subject to a constant, controllable salt brine condition. This paper presents the design of a new "dry salt" SCC specimen test configuration to overcome that limitation. The test specimen configuration provides a crack growth rate test specimen charged on the edge of the crack plane with an initially-dry salt assemblage that would deliquesce under temperature and humidity conditions and thereby environmentally load a fatigue pre-cracked, wedge-opening-loaded specimen with the naturally-formed brine. The specimen design and test configuration enables measurements of crack growth rate (da/dt) under falling stress intensity, K_J, conditions (the falling K is caused by compliance change due to crack growth under constant crack opening displacement, or COD, loading). This data would be applied to estimate subcritical crack growth in the residual stress fields of canisters weldments.

CRACK GROWTH KINETICS

Deposits on the surface of a DSC, especially those near a seacoast with a source of atmospheric marine salts, or an industrial setting with a source of fly-ash particulates, could be anticipated to be a mixture of dust and chloride-bearing salts and compounds. The initiation of stress corrosion cracks with these deposits is dependent on the salt/dust assemblages, temperature and humidity, and the material and its surface stress condition [1-5]. Further considerations of the requisite conditions and the timeframe of the potential initiation of cracks are not developed in this paper.

If a flaw in a canister has been identified as a stress corrosion crack with consensus NDE (nondestructive examination) methods and qualified inspection personnel, the characterization of the flaw and an understanding of its likelihood and rate for further propagation are needed for its disposition.

There are several specimen designs and approaches to test (measure) crack velocity or da/dt under a stress intensity factor, K, loading. Figure 1 shows the various geometries and loading modes for the crack growth rate test specimens.

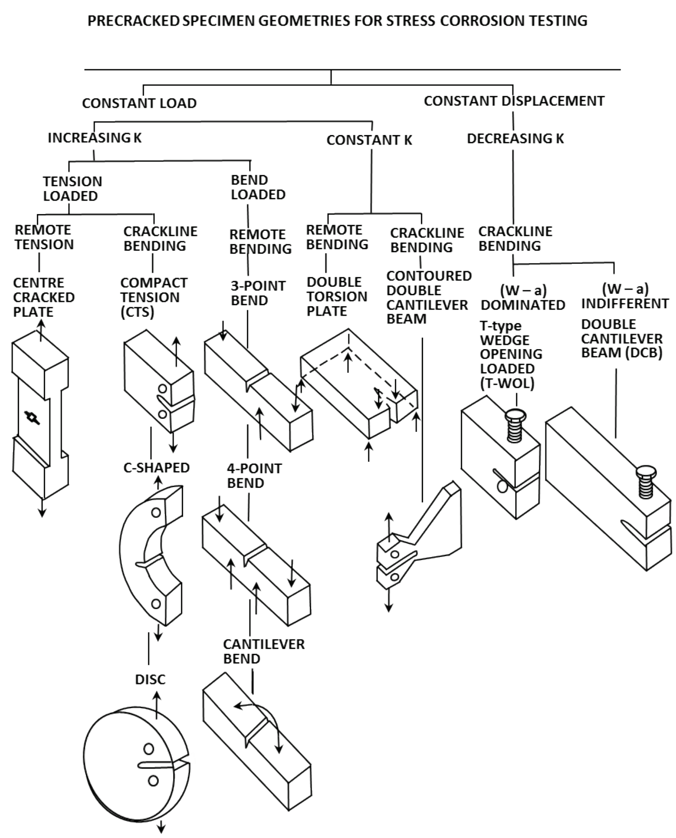


FIGURE 1 PRECRACKED SPECIMEN GEOMETRIES FOR STRESS CORROSION TESTING (figure from the United States Naval Research Laboratory, Washington, DC)

The challenge with generating data relevant and directly applicable to the DSC is the environmental loading and testing for what we will call "dry salt" conditions. That is, with initial fully dried deposits on the canister surface, and under falling canister temperatures with extended times, the deposits with deliquescing salts will create the aggressive environment.

Crack growth rate testing aimed at conditions for a DSC in natural exposure settings and with salt assemblages loaded onto the specimens has been performed over the past several years. Researchers at Japan's Central Research Institute of Electric Power Industry have generated the largest data sets to date [5, 14-17]. The specimen designs included 4-point bend [3], 3-point bend [15, 16], and compact tension (CT) [14, 17]. Figures 2 to 4 show a sampling of the specimen designs and mechanical loading with the corresponding crack growth rate results.

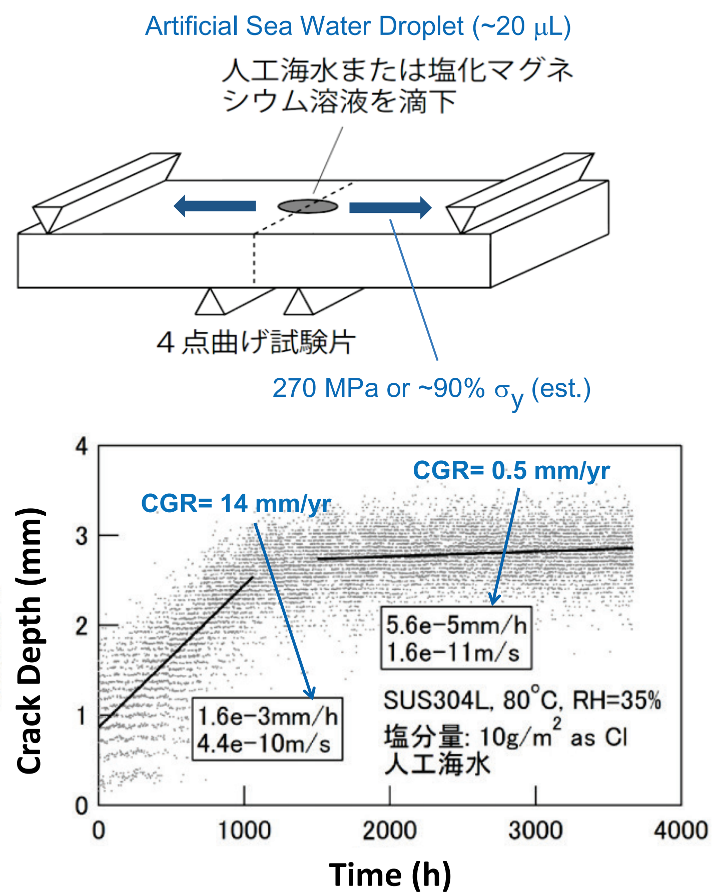


FIGURE 2 4-POINT BEND TEST SPECIMEN CONFIGURATION AND RESULTS (Reproduced from [3], the specimen may not have contained a pre-crack)

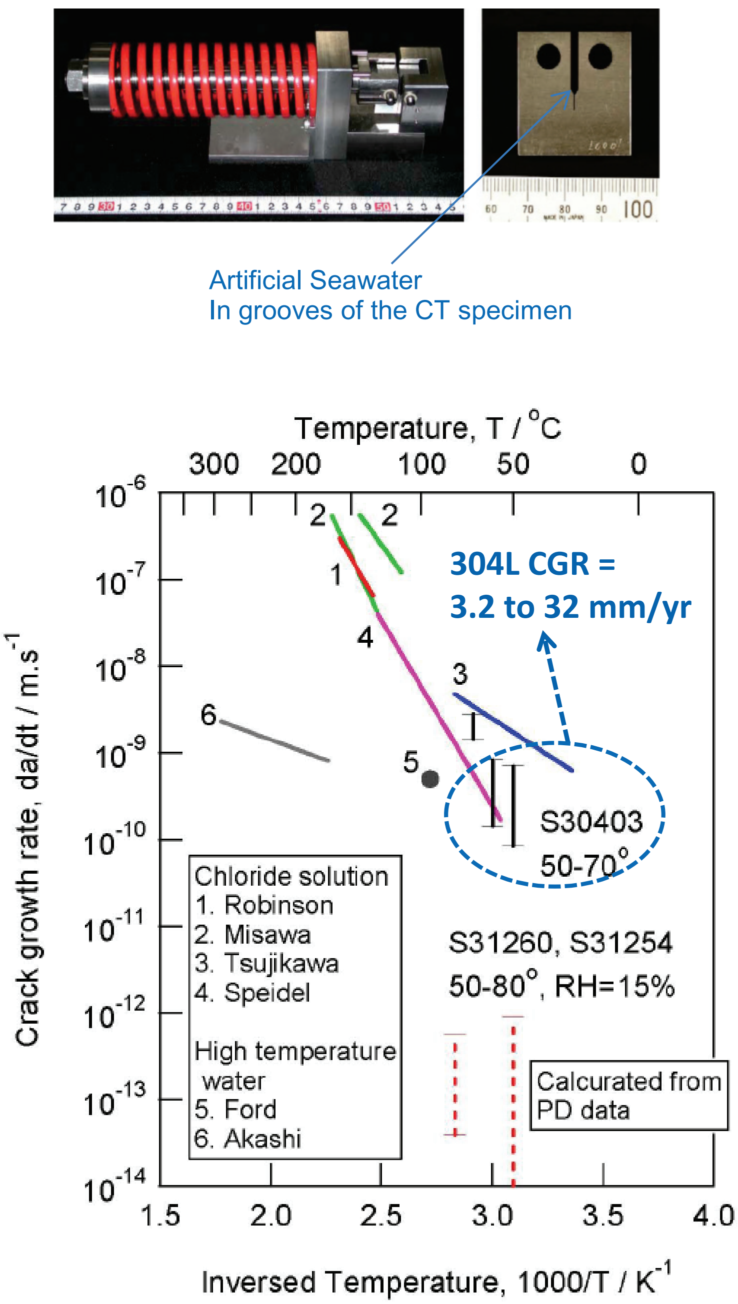
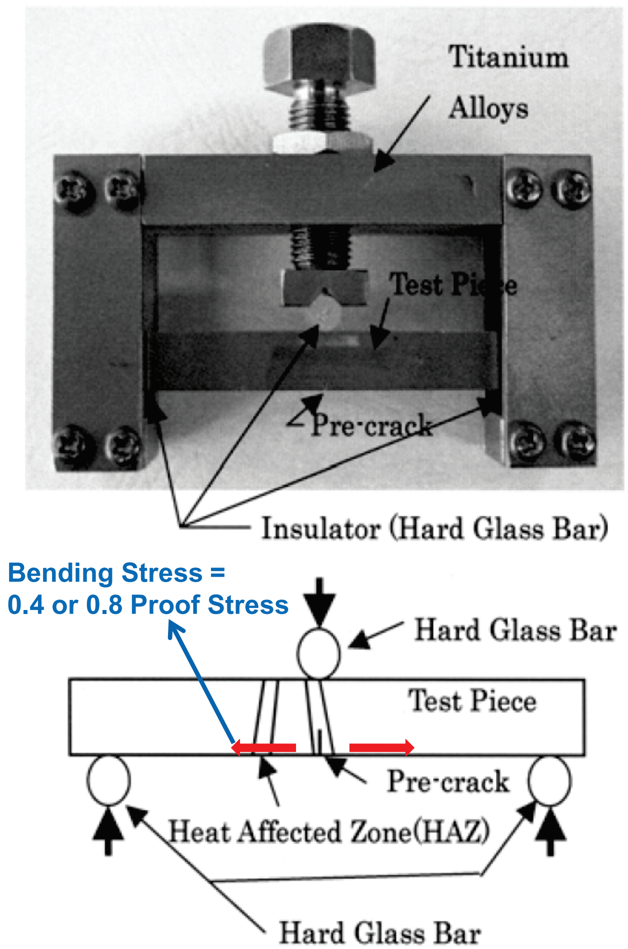


FIGURE 3 CT TEST SPECIMEN CONSTANT LOAD CONFIGURATION AND RESULTS (Reproduced from [14])



Materials: 304 and 304L

Natural Exposure (Miyakojima Island):
CGR=$1.2x10^{-12}$ to $1.8x10^{-11}$ m/s
= 0.04 mm/yr to 0.5 mm/yr

Accelerated Test (60°C, 95RH, NaCl steam mist):
CGR= $1.0x10^{-10}$ to $3.5x10^{-9}$ m/s
= 3 mm/yr to 110 mm/yr

FIGURE 4 3-POINT BEND TEST SPECIMEN CONFIGURATION AND RESULTS (Reproduced from [15, 16])

Specimens can be tested without pre-cracks, however pre-cracks are typically used to provide a defined initial crack front in a specimen. Semi-elliptical cracks can be electro-discharge machined (EDM) in the 4-point bend specimen design, although it appears that this was not used in the CRIEPI testing in Figure 2 based on the cross-section containing the SCC [3]; fatigue pre-cracking in air was not always used in the testing with 3-point bend [15,16] and CT [14] design specimens in the CRIEPI testing. The aggressive environmental conditions used to load the crack growth test specimens was chloride solutions or artificial seawater. Natural exposure testing was also used in the 3-point bend specimen testing [15,16].

For the CT testing, the chloride solution was placed as droplets (~20 μL) at the pre-crack region [14] of the specimen and allowed to dry. The specimen was then exposed to temperature and humidity conditions.

Phenomenologically, stress corrosion cracking typically has a dependency on the applied stress intensity factor, K, as shown in Figure 5. A threshold stress intensity factor, K_{ISCC}, is observed below which SCC does not propagate and/or propagates very slowly and would not grow to threaten the structural integrity of a component over its lifetime. A region of crack growth occurs that is relatively insensitive to the K loading, followed by a region of strong dependency of subcritical crack growth on K prior to mechanical instability. The mechanical instability is the classic intrinsic fracture toughness of the material, conventionally labeled as K_{IC}.

Ideally a test configuration would allow salt loading to be prototypic to the canister in that it is initially a dried salt assemblage with dust or inert material. The salt would be allowed to deliquesce under the test conditions of temperature and humidity to form brine that could be drawn by capillary forces to the crack tip. Further, the configuration would allow the characterization of the K dependency in a single test setup. A new test configuration, described in the next section, has been developed at the Savannah River National Laboratory; the testing is being initiated at the time of this paper.

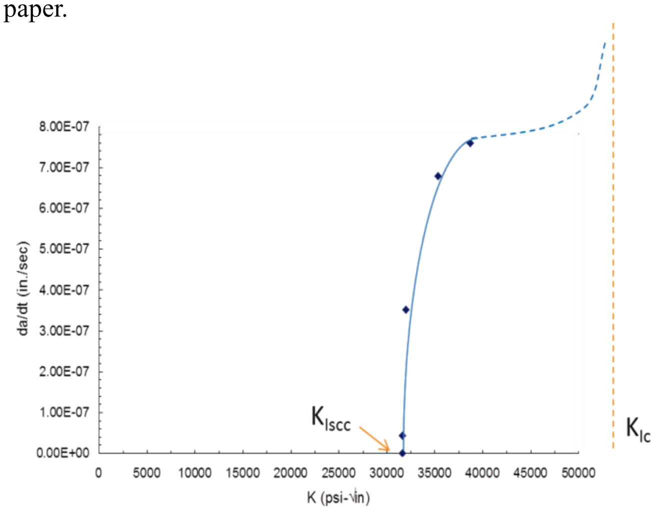


FIGURE 5 EXAMPLE SCC GROWTH DEPENDENCY ON APPLIED K (the SRNL data shown is for A285 carbon steel under conditions of nitrate stress corrosion cracking [18])

DRY SALT CRACK GROWTH KINETICS - WEDGE OPENING LOADED SPECIMEN DESIGN

Figure 6 shows the sketch of the new dry salt SCC test configuration concept that uses a wedge opening loaded specimen per ASTM E1681, "Standard Test Method for Determining Threshold Stress Intensity Factor for

Environment-Assisted Cracking of Metallic Materials," that is loaded with an instrumented bolt, and that contains a special cradle to load a salt mixture and hold it against the external faces of the specimen. The SCC testing is under "falling K" kinetics that would provide data to enable construction of a da/dt vs K curve such as shown in Figure 5. Since the strict dimensions to enable plane strain conditions in the specimen design with austenitic stainless steel are not met, the derived K is termed K_J to reflect that plastic conditions exist at the crack tip in the testing.

Figure 7 shows the actual test assembly that has been constructed for this concept. The glass vessel shown would contains brine following ASTM E104, "Standard Practice for Maintaining Constant Relative Humidity by Means of Aqueous Solutions," at the bottom of the vessel to control the relative humidity (RH) at a desired target level (e.g. $MgCl_2$ to control RH at 30%). The vessel is placed in an oven for temperature control. The feed-through on both the glass vessel and the oven enable a DAS to acquire load drop with time for a fixed displacement (COD). The ASTM-based algorithms are applied to estimate the crack length and applied K real time. Post-test visual check of the crack lengths on the sides of the specimen, and also the specimen crack face with destructive examination will validate and calibrate the estimations.

The salt mixture cradle is machined from an aquarium bubbler, and its porosity allows a humid air contact to the salt mixture. At the time of submittal of this paper, the testing was being initiated. Two conditions are being tested. The test salt assemblages are 1) CeO_2 + 2% mixture of $NaCl/KCl/CaCl_2$ and 2) a mixture of dust and artificial sea salt. The temperature and humidity conditions are 45 ºC and 30-50% RH, respectively. The testing is being conducted to shakedown the test configuration before a set of test conditions are selected by an inter-laboratory research team.

APPLICATION OF DRY SALT CRACK GROWTH DATA FOR FLAW DISPOSITION IN ISI

As stated, flaw growth must be considered in disposition of flaws identified and characterized in inspection of a DSC. There are two primary inputs for planar flaw growth evaluation, namely:

(1) residual stress distributions for canister fabrication welds[1]; and

(2) crack growth rate curve(s) or da/dt vs. K for these canister materials and environmental conditions

Estimation of weld residual stresses for a DSC has been performed using the methodology contained in reference [9]. The full results have been reported in reference [10]. As an example, Figure 8 shows the calculated through-wall residual stress distribution perpendicular to a circumferential weld. Measurements of the weld residual stresses on a full-size prototype DSC are in progress at the Sandia National Laboratory [19].

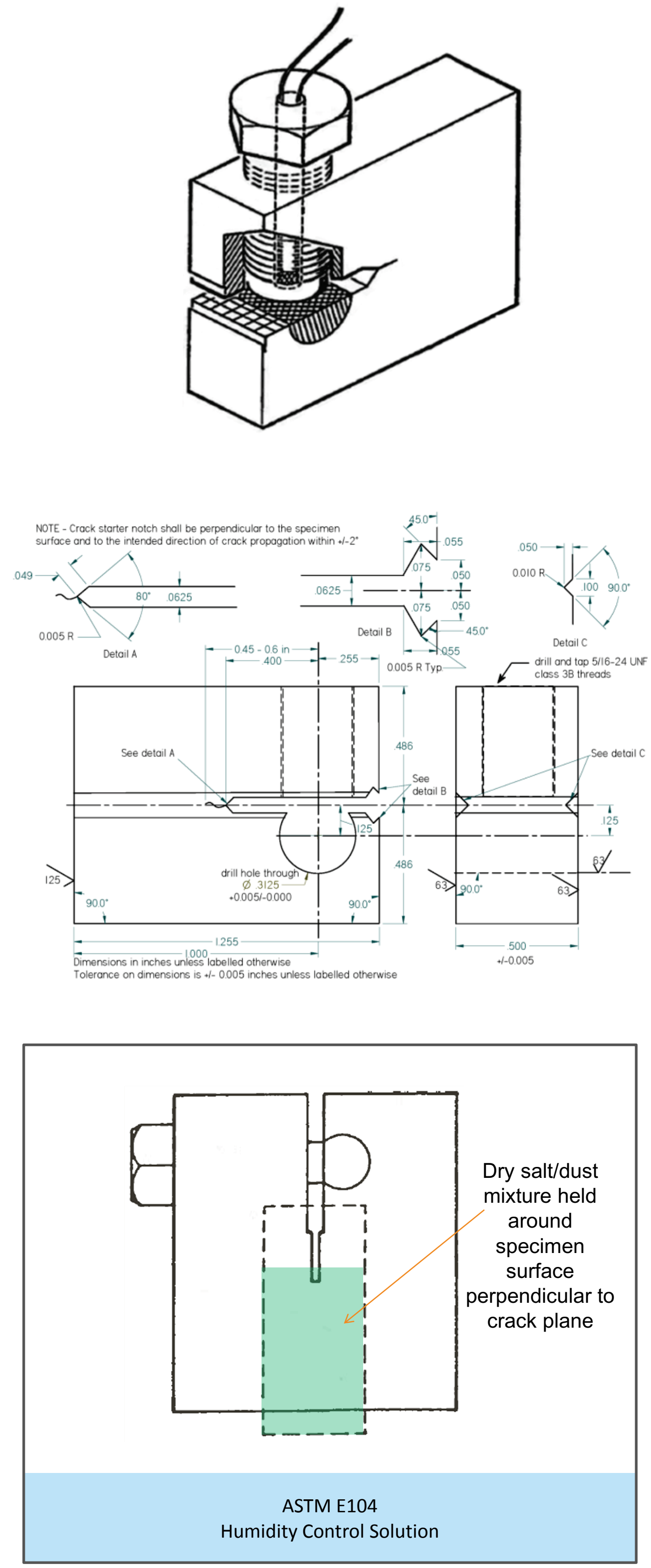


FIGURE 6 ASTM E1681 WEDGE-OPENING LOADED SPECIMEN DESIGN, 20% SIDE-GROOVED, IN A SALT MIXTURE CRADLE. AN INSTRUMENTED BOLT MEASURES THE LOAD DROP WITH CRACK EXTENSION

[1] Applied loads need to be checked for role in subcritical flaw propagation.

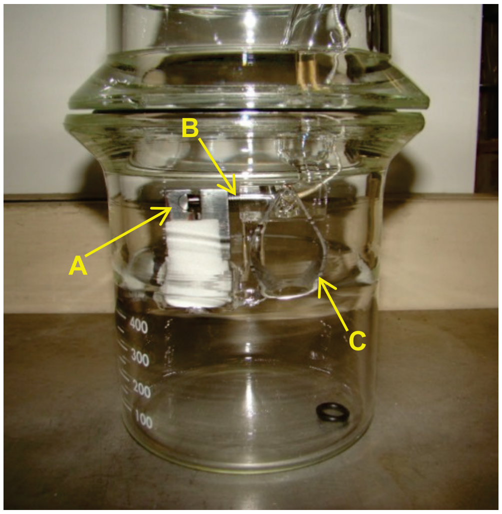


FIGURE 7 SCC TEST CONFIGURATION SHOWING THE CRADLED WOL SPECIMEN (A) WITH AN INSTRUMENTED BOLT (B) AND A COMPANION TEARDROP COUPON (C) IN A CONSTANT HUMIDITY VESSEL

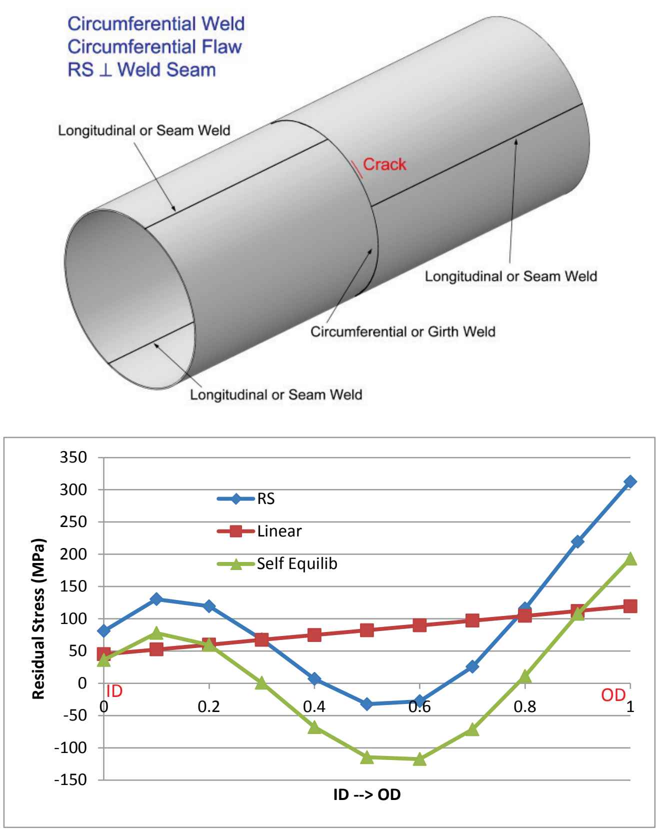


FIGURE 8 CIRCUMFERENTIAL CRACK PARALLEL TO A CIRCUMFERENTIAL WELD - RESIDUAL STRESS PERPENDICULAR TO THE DOUBLE-V NOTCH CIRCUMFERENTIAL WELD (Reproduced from [10])

Appendix C of the ASME SCXI provides guidance for evaluation of crack growth for light-water reactor (LWR) conditions. The guidance from Appendix C can be used in evaluation of flaw growth under dry salt SCC conditions. Figure 9 is the da/dt vs K crack growth model recommended in Appendix C. It is expected that the new testing underway, and with the other literature data will be assembled to construct a recommended SCC growth rate curve (da/dt vs. K) for the N-860 code case.

The available data to date on SCC under "dry salt" conditions, especially the CRIEPI data [16] under natural conditions, suggest that a constant rate of 0.5 mm/year represents the crack growth rate for a range of K loadings. This rate is recommended to be used to estimate SCC growth in the interim until additional testing and consensus evaluation of the data is complete, and the results are published in code case N-860.

ACKNOWLEDGEMENT

This work at the Savannah River National Laboratory was sponsored by the Nuclear Fuel Storage and Transportation (NFST) Planning Project, and by the Used Fuel Disposition (UFD) Campaign under the U.S. Department of Energy, Office of Nuclear Energy; and by the Savannah River Nuclear Solutions, LLC under Contract No. DE-AC09-08SR22470 with the U.S. Department of Energy.

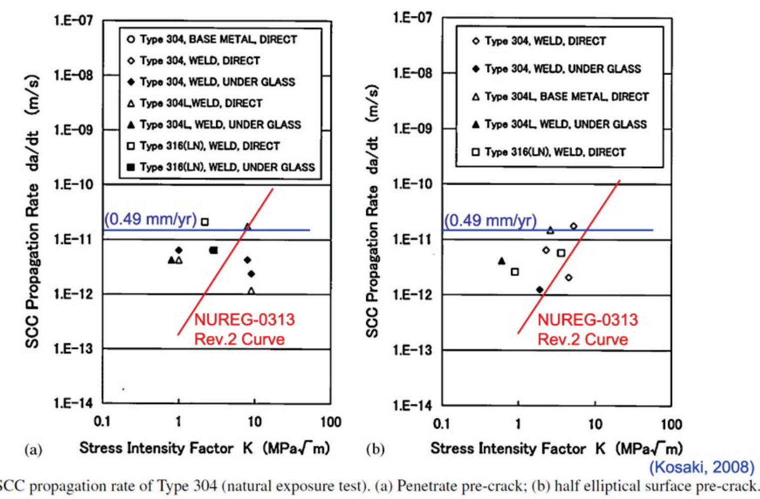


FIGURE 9 da/dt vs K_I for Intergranular Stress Corrosion Cracking (Reproduced from [14] and curve from NUREG-0313 [20] and ASME SCXI, Non-mandatory Appendix C [7]

REFERENCES

[1] Tani, J.I., 2009, "Initiation and Propagation of Stress Corrosion Cracking of Stainless Steel Canister for Concrete Cask Storage of Spent Nuclear Fuel," Corrosion, Vol. 65, No. 3, pp. 187-194.

[2] Caseres, L. and Mintz, T.S., 2010, *Atmospheric Stress Corrosion Cracking Susceptibility of Welded and Unwelded 304, 304L, and 316L Austenitic Stainless Steels Commonly Used for Dry Cask Storage Containers Exposed to Marine Environments*, NUREG/CR-7030, Office of Nuclear Regulatory Research, U.S. Nuclear Regulatory Commission, Washington, D.C., USA.

[3] Shirai, K., Tani, J. ,and Saegusa, T., 2011, *Study on Interim Storage of Spent Nuclear Fuel by Concrete Cask for Practical Use - Feasibility Study on Prevention of Chloride Induced Stress Corrosion Cracking for Type 304L Stainless Steel Canister*, CRIEPI N10035, Central Research Institute of Electric Power Industry , Tokyo, Japan (In Japanese).

[4] He, X., Mintz, T. S., Pabalan, R., Miller, L., and Oberson, G., 2014, *Assessment of Stress Corrosion Cracking Susceptibility for Austenitic Stainless Steels Exposed to Atmospheric Chloride and Non-Chloride Salts*, NUREG/CR-7170, Office of Nuclear Regulatory Research, U.S. Nuclear Regulatory Commission, Washington, D.C., USA.

[5] Enos, D.G., Bryan, C.R., and Norman, K.M., 2013, *Data Report on Corrosion Testing of Stainless Steel SNF Storage Canisters*, SAND2013-8314P, FCRD-UFD-2013-000324, Prepared for the U.S. Department of Energy Used Fuel Disposition Campaign, Sandia National Laboratories, NM, USA.

[6] Draft Report for Comment, 2015, *Standard Review Plan for Renewal of Specific Licenses and Certificates of Compliance for Dry Storage of Spent Nuclear Fuel*,. NUREG-1927, Rev. 1, Office of Nuclear Material Safety and Safeguards, U.S. Nuclear Regulatory Commission, Washington, D.C., USA, ML15180A011.

[7] ASME Boiler and Pressure Vessel Code, Section XI, *Rules for Inservice Inspection of Nuclear Power Plant Components*, 2015, American Society of Mechanical Engineers, New York, NY., USA.

[8] Chu, S., 2014, *Flaw Growth and Flaw Tolerance Assessment for Dry Cask Storage Canisters*, No. 3002002785, Electric Power Research Institute, Palo Alto, CA., USA.

[9] API 579-1/ASME FFS-1, 2007, *Fitness-For-Service* (API 579 Second Edition), American Petroleum Institute, Washington, DC., USA.

[10] Lam, P.S. and Sindelar, R.L., 2016, "Flaw Stability Considering Residual Stress for Aging Management of Spent Nuclear Fuel Multiple-Purpose Canisters," PVT-15-1195, to be published in ASME Journal of Pressure Vessel Technology, DOI: 10.1115/1.4032279.

[11] Broussard, J.E., 2016, "Limiting Flaw Size Evaluation for Welded Austenitic Stainless Steels: A Comparison of Section XI Appendix C and Appendix H Approaches," PVP2016-64037, Proceedings of ASME Pressure Vessels and Piping Conference, Vancouver, Canada.

[12] Shi, J., Wei, L., and Lam, P.S., 2016, "Flaw Stability Analysis of Semi-elliptical Surface Cracks in Canisters under the Influence of Welding Residual Stress," PVP2016-63887, Proceedings of ASME Pressure Vessels and Piping Conference, Vancouver, Canada.

[13] EDF Energy Nuclear Generation Ltd, 2014, *Assessment of the Integrity of Structures Containing Defects*, R6 Revision 4, Including Updates to Amendment 11, December 2014.

[14] Shirai, K., Tani, J., Arai, T., Watatu, M., Takeda, H., Saegusa, T., Windton, P.L., 2008, "Research on Spent Fuel Storage and Transportation in CRIEPI (Part 2 Concrete Cask Storage)," Paper ID P16P1215, 16th Pacific Basin Nuclear Conference (16PBNC), Aomori, Japan; Reprinted by Idaho National Laboratory, INL/CON-08-14592, Idaho Falls, ID, U.S.A.

[15] Kosaki, A., 2006, "SCC Propagation Rate of Type 304, 304L Steels under Oceanic Air Environment," Paper No. ICONE14-89271, Vol. 1, Plant Operations, Maintenance and Life Cycle; Component Reliability and Materials Issues; Codes, Standards, Licensing and Regulatory Issues; Fuel Cycle and High Level Waste Management, International Conference on Nuclear Engineering, July 17-20, Miami, Florida, USA, pp. 443-450.

[16] Kosaki, A., 2008, "Evaluation method of corrosion lifetime of conventional stainless steel canister under oceanic air environment," Nuclear Engineering and Design, 238, pp. 1233–1240.

[17] Tani, J, Mayuzumi, M., Arai, T., and Hara, N., 2007, "Stress Corrosion Cracking Growth Rates of Candidate Canister Materials for Spent Nuclear Fuel Storage in Chloride-Containing Atmosphere," Materials Transactions, Vol. 48, No. 6, pp. 1431-1437.

[18] Zapp, P.E. and Lam, P.S., 2000, *Preliminary Evaluation of Tank 15 Vapor Space Cracking*, WSRC-TR-2000-00187, Westinghouse Savannah River Company, Aiken, SC., USA.

[19] Enos, D.G. and Bryan, C.R., 2015, *Status report: Characterization of Weld Residual Stresses on a Full-Diameter SNF Interim Storage Canister Mockup*, SAND2015-7068R, FCRD-UFD-2015-00123, Prepared for the U.S. Department of Energy, Used Fuel Disposition Program, Sandia National Laboratories, NM, USA.

[20] Hazelton, W. S. and Koo, W. H., 1988, *Technical Report on Material Selection and Processing Guidelines for BWR Coolant Pressure Boundary Piping*, NUREG-0313, Office of Nuclear Regulatory Research, U.S. Nuclear Regulatory Commission, Washington, D.C., USA.