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AN APPROACH TO MODEL ABSTRACTION OF STRESS CORROSION CRACKING DAMAGE IN MANAGEMENT OF SPENT NUCLEAR FUEL AND HIGH-LEVEL WASTE

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ABSTRACT

This paper presents an approach to assess stress corrosion cracking (SCC) damage of a canister for use in confinement management (extended dry storage or geological disposal) of radionuclides from spent nuclear fuel and high-level (radioactive) waste. Localized corrosion, mainly in pitting form and fabrication flaws, were analyzed as a possible precursor to SCC using field/laboratory data. This paper assesses single crack propagation over long time periods and estimates the potential maximum opening area resulting from multiple cracks. This crack propagation model was developed by the Sandia National Laboratories (SNL) for disposal under seismic conditions, and it appears to be conservative with respect to radionuclide releases through the opening area. The SNL model could be applied to the weld and various metals for both management applications. The conservative SNL approach could be used to estimate consequences of radionuclides dispersals, if a canister failed as the confinement barrier.

INTRODUCTION

Canisters[1] provide confinement of radionuclides in the management of spent nuclear fuel (SNF) and high-level waste (HLW) such as vitrified nuclear waste, for hundreds of years in extended dry storage or for thousands of years in geologic disposal. Stress corrosion cracking (SCC) may lead to partial loss of canister confinement if not addressed by design, manufacturing and aging management programs. This paper presents an approach to assess SCC impact on the radionuclide confinement boundary and models for SCC damage (i.e., opening area) of a canister. If the canister failed, limited radionuclides releases through the opening area may need to be assessed. The abstracted model discussed herein is developed with simple representations utilizing a conservative and bounding approach that may be incorporated in a system performance model.

Specifically, the approach considered here include: (i) precursory step for SCC, such as pitting or flaws; (ii) single crack propagation; (iii) a recently developed model for estimating potential canister opening area from a possible maximum number of multiple through-wall cracks for a predominant range of crack aspect ratios (i.e., potential maximum opening area). The model was developed under seismic conditions in a geologic disposal facility by the Sandia National Laboratory ([1]; hereafter referred to as the "SNL model"); and (iv) application of the SNL model in welds and various metals for radioactive waste management applications.

Metals of stainless steels, nickel-based alloys, and carbon steels, and the chloride environment are largely considered. However, the application is not limited to the selected metals or the environment. Residual stresses from welding and other fabrications (e.g., rolling) processes are addressed. External stresses from seismic events are also considered. Chemistry and microstructure may be altered during welding (e.g., heat affected zone [HAZ]). It is noted that inspection, monitoring, mitigation, or remediation, as needed, is part of aging management in extended dry storage. However, in disposal, such opportunities are available but limited only during the initial operations.

Precursory steps considered in this abstracted model are pitting by initial corrosion and flaws formed during welding and other fabrications. For pitting, limited density and size of pits are discussed with respect to existing pitting theories. An example calculation in stainless steel is made on the possibility of pit-induced SCC, and results compared with available test data. Propagation of single SCC crack is discussed with observed test results and interpretations, whether the canister could fail over long time periods. There are large uncertainties associated with single crack propagation in the real canister installations over such long time periods. Therefore, the recently developed SNL model [1] for potential maximum opening area in disposal is assessed for applications in the weld and various metals. The SNL model conservatively assumes that the environment for SCC is present and, thus it is stress-based.

[1] The term canister is used in this paper to refer to a leak-tight metal structure used to contain spent fuel assemblies during extended dry storage or geologic disposal.

NOMENCLATURE

C	Constant
E	Young's modulus
HAZ	Heat affected zone
HLW	High-level waste
J	Energy per unit fracture surface area
K	Stress intensification factor
LWR	Light-water reactor
PDM	Point defect model
SCC	Stress corrosion cracking
SNF	Spent nuclear fuel
SNL	Sandia National Laboratories
t	Time
w(t)	Crack width
YS	Yield stress
σ	Applied stress
δ	Potential maximum opening area of multiple cracks per deformed or weld/HAZ area

PRECURSORY STEPS FOR SCC

Two precursory steps for SCC are considered in this abstracted model: pits formed during early corrosion processes and flaws formed during fabrication and welding.

Pitting Corrosion

Pitting corrosion occurs on the free metal surface exposed to the aqueous environment or inside crevice. Once a pit forms, stress is concentrated at the bottom of the pit. At the concentrated location, the stress intensification factor may reach a threshold value for SCC initiation, depending on pit size and magnitude of applied stress.

In a non-passive metal, such as carbon steel, a pitting factor is defined as the ratio of the pit depth to the general corrosion depth in the given environment [2 to 4]. In carbon steel, the pitting factor will approach 1 with time, implying pitting eventually disappears. In passive metals such as stainless steel or nickel-based alloys, more mechanistic studies have been done. These are summarized below.

References 5 and 6 proposed statistical models for pitting corrosion process. Reference 6 reviewed the historical background of extreme value statistics and development in the theory and application to pitting corrosion. However, statistical models do not traditionally include time as a variable and do not account for time-dependent nature of the pitting corrosion. Reference 5 proposed a model for pit generation, based on a homogeneous (i.e., time invariant pit generation rate) Poisson process. The experimental work of Reference 5 suggests that the type of metal and environmental conditions determine the (electrochemical) potential for corrosion. At low potential, the pit initiation rate is high and the repassivation (i.e., cessation of continuous propagation of an initiated pit) rate is low. While the pit initiation rate depends on the potential, the repassivation rate is independent of the potential. Associated with the Poisson distribution of the pit population, Reference 5 derived an exponential distribution for the initiation time between two successive occurrences of new pits. Further experimental data on the distribution of the initiation time for pit generation revealed that more complex stochastic models may be necessary because the time between the generations of new pits does not fit an exponential distribution.

Reference 7 established a point defect model (PDM) for the pit initiation. More recently, Reference 8 has furthered the theory for the application in waste management. Point defects such as interstitial or vacancy move and form voids at metal-passive (oxide) film interface in chloride solutions. Pits nucleate at a pitting potential. Many of them quickly die by repassivation under open-circuit (i.e., free corroding without applied potential) conditions. Currently, experimental data on pit repassivation, especially delayed repassivation, are sparse.

Pit density (i.e., number of pits per unit surface area) is restricted by the repassivation process. In addition, pit initiation itself is restricted by low cathodic capacity near a pit. When a pit forms, the bottom of the propagating pit acts as an anode and the near free surface area to the pit behaves as a cathode. In the cathodic region, pitting cannot occur. This restricts the pit density, as modeled in References 9 to 11. Reference 11 furthered the restriction considering that a thin-layer of water film in atmospheric condition cannot support localized corrosion condition for extended periods. Pit growth rate is generally understood to decrease with time. Nevertheless, it is believed that pits are a precursor for SCC initiation [12].

Some measurements of pit density are available, along with theoretical studies [13]. Under polarization conditions, the range of measured pit density is 0.1 – 100/cm^2 [14, 15]. The theoretical estimate is on the order of 1000/cm^2 [16], and does not consider the repassivation phenomena.

Flaws from Fabrication and Welding/HAZ

There is a relatively high density of potential incipient surface cracks associated with microscopic discontinuities.

Reference 17 (cited in Reference 1) postulates that the incipient crack size (associated with microscopic crack formation at defect sites such as mechanical flaws) is 0.05 mm (1.97 mils). This value was derived from Quality Assurance work, corroborating data, and expert opinion.

Manufacturing flaws develop largely from welding [1]. The flaw detection and repair size criterion is 1.6 mm (63 mils), as compared to the somewhat more sensitive 1 mm ultrasonic inspection defect-detection threshold used for the weld mockup study. Imperfections uncovered by metallographic examinations but not detected by ultrasonic inspections were gas bubbles, the majority of which were less than 0.08 mm (3 mils). The size distribution of initial weld flaws is exponential, with an upper-bound truncation due to the weld thickness.

Pit-Induced SCC: example calculation

Corrosion data are available for austenitic stainless steels exposed to the coastal environment at test facilities in Kure Beach, North Carolina for 15 years [18]. During this exposure pitting corrosion occurred on the coupons or U-bend specimens with salt deposits. Reference 18 also summarized SCC observations for (i) sensitized 304 stainless steel in Kure Beach and (ii) thermally annealed or sensitized 304 and 304L stainless steels in the coastal environment of Okinawa, Japan, both under stress. Using the reported data from the exposure tests at room temperature, pit sizes are estimated as10 – 100 µm [0.39-39.3 mils], with a uniform distribution. Residual stress can be due to welding or other fabrication (such as rolling), which was not relieved by thermal annealing or other mitigation processes (such as applying compressive stress). Available measured data in Japan for austenitic stainless steel suggest that weld residual stress can be 0 – 600 MPa [0 – 87 ksi] [19]. The stress distribution is assumed to be normal, based on the trend of the measured data. Using the distributions of the pit size and stress, the cumulative probability of stress intensification factor, K (MPa $m^{1/2}$), is calculated, using GoldSim Version 10.11 for a probabilistic simulation [20]. The stress intensification factor was calculated using the formula in Table 1.

Table 1. Cumulative probability of stress intensification factor, K(MPa $m^{1/2}$) = $\pi^{1/2}$ x stress x (crack size)$^{1/2}$

Probability	0.001	0.05	0.25	0.75	0.95
K (MPa $m^{1/2}$)	0.43	1.57	2.59	4.57	6.94

(Conversion factor: 1 MPa $m^{1/2}$ = 0.91 ksi $in^{1/2}$)

The stress intensity factor values (K) were measured using the fracture mechanics SCC tests with salt deposits [18, 19, 21]. The measured K values for SCC ranged between 0.5 – 7.0 MPa $m^{1/2}$ (0.5 – 6.4 ksi $in^{1/2}$). The calculated data are consistent with the values observed in the coastal area tests [18] discussed above.

It is also noted that additional applied stress may be present. Any adjacent component may exert force on a canister. More notably, seismic-induced stress could also occur. The annual probability of annual seismic events may be of a fixed value. However, the occurrence of seismic events is more likely with increasing time, such as geologic disposal [22].

SINGLE SCC CRACK PROPAGATION (PERSPECTIVES)

After SCC initiates, the induced cracks are generally understood to propagate continuously. However, this understanding is based on conservatisms such as sustained applied stress or sharp crack geometry. In the passive system used in the management of SNF and HLW, materials characteristics and the stress state at the crack tip will change during crack propagation. The residual or applied stress may decrease, or the crack tip shape may be modified to decrease the stress intensification factor, thereby slowing the crack propagation rate. The following summarizes such modifications, as discussed in various industry literatures (e.g., discussion in Reference 1; [19]).

Residual stress along the thickness of a canister varies near the weld. Stress will be redistributed around the crack tip during crack propagation. If crack branching or a tortuous crack path decreases the local stress, the crack propagation rate would decrease, whether it is inter- or trans-granular cracking. Plasticity in terms of $J_{material}$ may increase more than $J_{applied}$ with crack propagation, which may affect the slip or twin process for film rupture at the crack tip for SCC propagation. Therefore, it may be difficult to sustain constant K value at the crack tip.

Cracks cannot propagate without required stress. Residual stress decreases rapidly as one moves away from the weld area. There will be no significant stress from internal gas pressure inside a canister. Under seismic impact conditions, stress decreases from the outside surface along the thickness. It is expected that the seismic stress is applied to the outer surface during the collision of a canister with neighboring canisters or rocks [1].

The strength of neutron irradiation is insignificant in the management of SNF and HLW if compared to reactor operations. Environmental variations such as seasonal temperatures or salt concentration in air may decrease the crack propagation rate. For example, the SCC in the coastal area could occur with the deliquescence of deposited salts on the canister surface [19]. The deliquescence in turn depends on the temperature and relative humidity (RH) on the canister surface. This temperature and RH are determined primarily by the canister heat loading, and ambient atmospheric temperature and RH. At higher temperatures without salt deliquescence, SCC will not propagate. In addition SCC rates themselves depend on the temperature.

In light-water reactor (LWR) experiences, SNL believes that material degradation due to through-wall growth of neighboring cracks has not been observed [1]. According to Reference 1, depending on the stress distribution SCC may initiate and propagate through-wall. If several cracks were to initiate in the same area, coalesce, propagate through-wall while remaining straight (i.e., perpendicular to the surface), and maintain smooth crack faces, material could fall out. The simultaneous occurrence of all of these events is improbable. For through-wall cracks, tighter and relatively separate cracks are more expected. However, it is noted that some events such as coalescence of multiple cracks are important for the structural integrity assessment of actual nuclear power plants.

Mitigation and remediation processes (such as applying compressive stress), will decrease the probability of crack propagation. They will require prior inspections for precursory indications such as pits or early cracks. For a newly installed canister, thermal annealing if appropriate or applying compressive stress may also mitigate crack initiation.

Based on this discussion, the probability of a single crack penetrating through the canister wall can be low. An example from the literature is shown in Figures 1 and 2 below.

Figure 1. Crack tip near the outside diameter surface of a type 304 stainless steel piping system contaminated with chlorides. Trans-granular crack is arrested. Crack depth in the picture is about 1.2 mm (50 mils) [23] (reprint permission by ASM International)

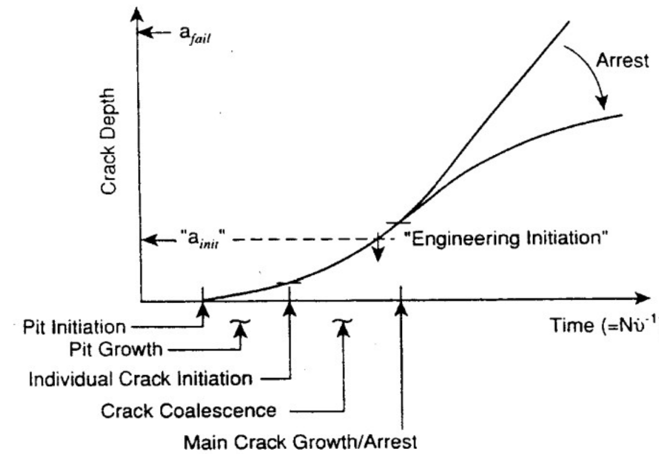

Figure 2. Proposed sequence of crack initiation, coalescence, and growth for steel under-going subcritical cracking in aqueous environments [24]. Crack depth for initiation is "a_{int}" and crack depth for failure is "a_{fail}"

There are other literature data that show gradual increase in crack growth rate with increase of stress intensification factor or time [19, 25, 26]. This means crack growth rate can decrease as the local stress intensification factor decreases for various reasons. Unlike the reactor studies, however, the probability of the through wall cracking has not been quantitatively studied in the management of SNF and HLW. The assessment of single crack propagation or environment/materials conditions for SCC has large uncertainties over long time periods.

Initiation of SCC requires a relatively narrow range of environmental and materials conditions. The environmental conditions include electrochemical potential, pH, temperature, aqueous chemistry (e.g., carbonate, or chloride), and applied stress (typically only in the weld/HAZ or deformed area). In humid atmospheric corrosions, appropriate relative humidity is also a factor. The thermal treatment of materials may lead to microstructure alteration in many cases. More comprehensive discussion is referred to in literature [12] and in the author's ongoing work [27]. This range of conditions initially limits the SCC susceptibility on the canister surface.

POTENTIAL MAXIMUM OPENING AREA OF MULTIPLE CRACKS

The above sections discussed that the opening area of multiple cracks will be limited by: (i) density and size of pits and flaws; (ii) likelihood of cracks extending through the wall; (iii) environmental and materials conditions for SCC; and (iv) if inspection, mitigation and remediation are implemented. However, the assessment of single crack propagation or environment/materials conditions for SCC has large uncertainties over long time periods, and data under various anticipated conditions are lacking. The SCC issue has been considered for some specific disposal environments. For extended dry storage canisters, SCC is being studied and, to date, no SCC has been observed based on very limited observation. If the canister failed, a potential maximum opening area has been used as a conservative and bounding approach in disposal for the assessment of radionuclide releases. This opening area is for a possible maximum number of multiple through-wall cracks for a predominant range of crack aspect ratios. Ideally, the analytic model representing the potential maximum opening area should be simple, for incorporating in a system performance model.

SNL Model [1]: basis

Models for potential maximum opening area of multiple cracks under seismic impact scenarios have been developed for disposal canisters in an open drift repository design [1]. The SNL seismic model allows all possible surface cracks to penetrate through the wall thickness, and is likely to be conservative and bounding. The following summarizes the SNL model and its applicability for confinement assessment.

The SNL model is stress based, and conservatively assumes that the environment for SCC exists. The centers of two cracks are separated by a parameter (approximately the canister thickness, related to crack geometry) due to stress attenuation. The distance between two neighboring through-wall cracks would need to be greater than the wall thickness for the stress (and resultant stress intensity) to be sufficient to drive a flaw through-wall. This conclusion is based on stress field interactions between closely spaced parallel cracks. The crack aspect ratio (ratio of length to depth) has distribution for various possible crack geometries in a probabilistic system approach, and each value was sampled in the confinement assessment.

Examples of such a crack network are illustrated in Fig.3. In the SNL model, this type of crack network is generated only in the limited area of the canister surface deformed from seismic impact [22]. Once the crack penetrates through the wall, further crack growth will not occur due to stress relief.

To support this hypothesis of the crack separation by stress attenuation, a stress analysis was conducted with a crack network [28]. Figure 4 shows the analysis results. The upper figure shows a number of cracks separated horizontally and vertically. The lower figure shows longitudinal stress distribution along the center. The stress is attenuated near the crack. This implies that a new crack cannot form between two cracks spaced by the parameter.

SNL Model [1]: mathematical description

Although the model in Reference 1 primarily dealt with the seismic case, supporting data for crack behavior were from LWR experiences without seismic impacts. The SCC induced by weld residual stress was also assessed. The contribution from weld residual stress was insignificant in this case because the weld residual stress was mitigated by applying compressive stress as part of the fabrication process. Also, unlike the LWR case, the environment in storage and disposal does not have pressure-induced primary stress or severe neutron effects.

At a crack length, $a(t)$, the crack width, $w(t)$, is

$$w(t) = C\,\sigma\, a(t)/E \qquad (1)$$

Where
- σ: applied stress (MPa)
- E: Young's modulus (MPa)
- C: geometric constant
- t: time

This equation applies to conditions of plane stress and infinite size (conservative assumption).

Each crack area is a product of crack length and crack width. The number of cracks is proportional to the sample area divided by $a(t)^2$. The potential maximum opening area of multiple cracks is the product of each crack area and the number of cracks. The density per unit deformed or weld/HAZ area (cm^2/cm^2) is δ

$$\delta = C\,\sigma/E \qquad (2)$$

SNL Model: application

Results of the numerical analysis of crack spacing are not sensitive to Young's modulus and Poisson's ratio, for tested stress on the order of 207 MPa (30.0 ksi) [28]. This implies that the SNL model can be applied to various metals.

In an example LWR case, the number and size of surface cracks were reported from welds at the Nine Mile Point Unit 1 main recirculation lines [29]. The total number from Nine Mile Point surface cracks in welds is comparable with the above SNL model. However, only a small fraction of surface cracks penetrated through the wall thickness, with a low probability by observation and prediction [30]. Although the number of through-wall cracks may increase with time, the maximum number is limited in the SNL model due to the stress attenuation between neighboring cracks. The crack aspect ratio is considered to increase with large surface cracks up to 10 to 150 [29]. The dominant area of surface cracks appears to fall in a range of crack size.

The SNL model appears to be conservative and bounding inferred by stress analysis. The SNL model also appears to be partly consistent with the LWR example exercises of surface cracks above with respect to the most probable dominant crack size in the opening area and potential crack aspect ratio present for dominant crack sizes. Therefore, the SNL model is likely to be applicable to extended dry storage and disposal at some potential sites. This approach may be sufficient for the initial assessment of radionuclide releases if a canister failed (e.g., [31]). This is also partly supported by estimating the potential maximum opening area of cracks for various metals.

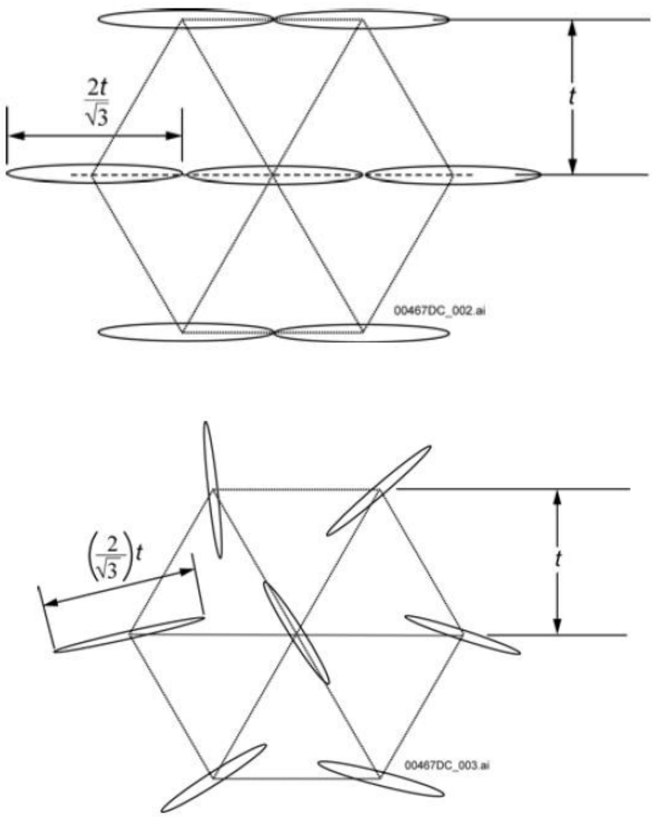

Figure 3. Crack network from SCC with the depicted densities of cracks [1]. The symbol "t" denotes distance between two cracks, and is also the wall thickness.

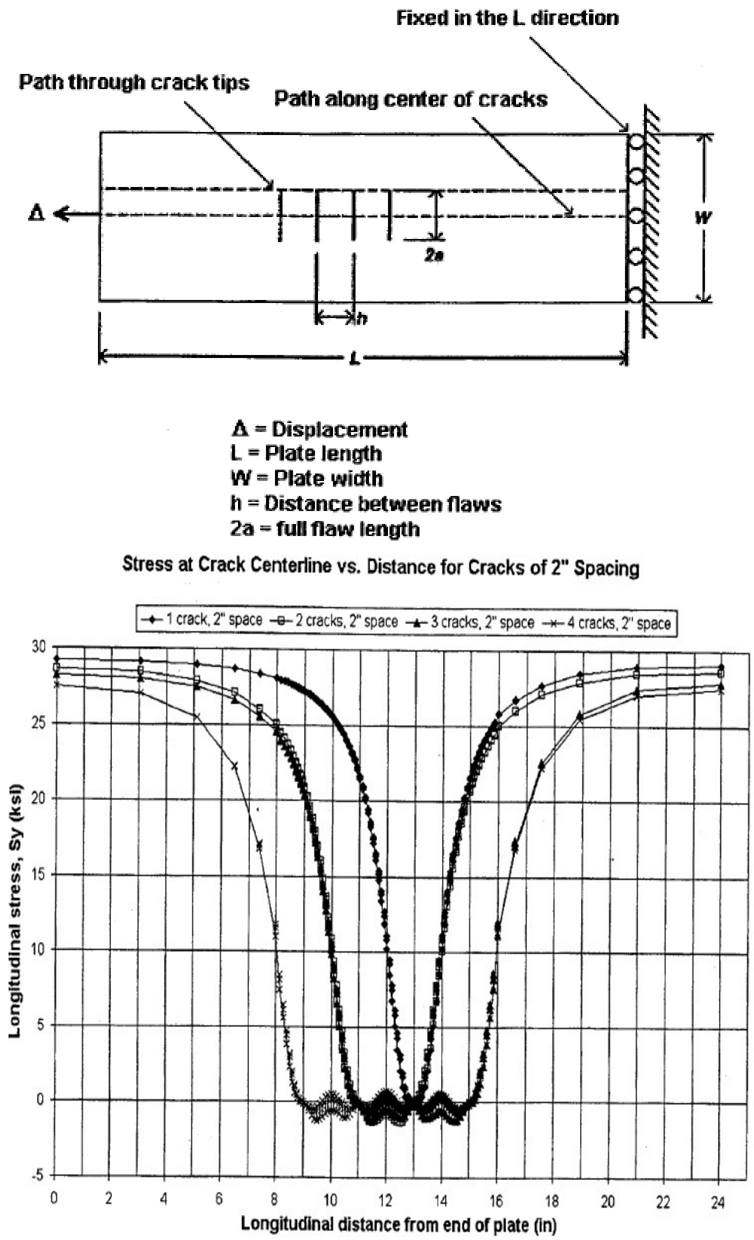

Figure 4. Stress analysis with crack network in top surface view [28]. Dimensions of the plate and setup for the analysis (upper) and longitudinal stress distribution along center with 5.1 cm (2 inch) spacing between cracks (lower), 1 ksi = 6.9 MPa

An exercise was conducted [32] for various metals with example parameter values for the various metals. Yield stress is used as an example applied stress. In the probabilistic confinement assessment, the yield stress is one of the sampled values.

Table 2. Materials properties for various metals [32]

Conversion factor: 1 MPa = 145 psi; E (Young's Modulus)

Materials	Yield Stress (MPa) (example applied stress)	E (MPa) x 10^3	YS/E (mean) x 10^3
Stainless Steel	170 - 310	193-207	1.2
Carbon Steel	207	207	1.0
Copper	70-310	108-117	1.7
Zircaloy	241	99	2.4

Using these values the potential maximum opening area of multiple cracks was calculated for each metal. For stainless steel, the mean value of potential maximum opening area of multiple cracks per unit weld/HAZ or deformed area is approximately 1.2×10^{-3} (fraction, cm^2/cm^2) for 170-310 MPa (24.6-44.9 ksi) of applied stress, $(193\text{-}207) \times 10^3$ MPa ($[28.0\text{-}30.1] \times 10^3$ ksi) of Young's modulus. The weld/HAZ area fraction is about $10^{-2} – 10^{-1}$ [33]. The fraction of the open area is small.

If the calculation from this method shows fast radionulide releases without substantial dilution by dispersion, then a more rigorous approach as used in reactors (e.g., [29, 30, 34, 35]) should be pursued.

SUMMARY

- An approach is presented on abstracting analytic models for SCC damage of canisters to assess the confinement and releases of radionuclides in management of SNF and HLW. Under certain environmental conditions, localized corrosion mainly in pitting form may be limited in density and size due to repassivation and cathodic capacity. An example calculation with field data shows that SCC of stainless steel can be initiated at pits in chloride environments.

- The propagation rate of single SCC may decrease during propagation, if the local stress intensification factor decreases. The values of environmental and material parameters for SCC are in narrow ranges. However, they are uncertain as performance periods increase in time.

- For disposal under seismic conditions, the possible potential maximum opening area of multiple cracks is estimated based on the SNL model. From the weld behavior at operating reactors and the SNL stress analysis, the SNL model appears to be conservative. The SNL model can be used for the weld and various metals for extended dry storage and geologic disposal. For stainless steel, the fraction of possible maximum crack opening area is limited based on the SNL model. The conservative SNL approach could be used for conditions where radionuclides could be substantially dispersed if a canister failed.

DISCLAIMER

The NRC staff views expressed herein are preliminary and do not constitute a final judgment or determination of the matters addressed or of the acceptability of any licensing action that may be under consideration at the NRC.

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