An Approach to Model Stress Corrosion Cracking in Nuclear Fuels Containing a Graphite Coating

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Summary

This work builds on existing fuel models developed at RMCC using the COMSOL Multiphysics platform. The fuel parameters relevant to Stress Corrosion Cracking (SCC) for in-reactor conditions are being modelled to develop an improved tool to predict SCC. The model will incorporate a critical sheath stress to initiate cracking of the protective oxide layer and the impact of CANLUB on iodine concentrations with burnup. Modelling of a stress-dependant critical crack depth to initiate sheath rupture is also being explored. The model will be benchmarked against a large database of SCC power reactor and experimental test reactor data available from the open literature. This paper will outline the proposed methodology for development and implementation of the model.

1. Introduction

Stress corrosion cracking (SCC) is a process through which the Zircalloy sheath surrounding nuclear fuel is corroded away by fission product, such as iodine. Under ideal circumstances a thin oxide layer of ZrO_2, which does not react readily with iodine, protects the sheath from corrosion. Stress in the sheath can cause cracking of this oxide layer, which exposes the underlying Zircalloy to iodine corrosion. Therefore there are two requisite factors for SCC to occur:[1]

1) There must be sufficient stress in the sheath to cause crack initiation and propagation in the oxide layer
2) There must be a sufficient quantity of free fission product iodine in the pellet-sheath gap to corrode through the sheath

SCC occurs during power ramps when the power level of a fuel bundle is increased. This can occur during refuelling when a bundle is shifted from a low power region of the core to a high

power region, or when overall reactor power is raised. During a power ramp, thermal expansion of the pellet increases sheath stress, which can lead to oxide layer cracking. Additionally, the higher power level increases the rate of production of fission product iodine as well as increasing the transport of that iodine to the pellet-sheath interface, where it can cause SCC.

In the early 1970's SCC fuel failures occurred in the Pickering and Douglas Point Reactors due to SCC[2].In addition to changes in refuelling practices, the inside of the fuel sheath was coated with a thin layer of graphite. Its name, CANLUB, reflected the assumption that the graphite would act as a lubricant, reducing stresses due to pellet-cladding interaction (PCI) and thus act as a physical protectant against SCC.

While CANLUB did indeed improve the fuel's resistance to SCC, the mechanism through which this is accomplished is not as originally expected. CANLUB acts principally as a chemical protectant by bonding with and immobilizing iodine, preventing it from corroding the sheath.

The active iodine gettering ingredient in CANLUB is not the graphite itself; impurity elements such as sodiumand potassium[2] and perhapsan ethyl cellulose binder residueleft over from baking during fuel manufacture are hypothesized as the active ingredients.[3]

Whileincidences of SCC in CANDU reactors are rare today, there is interest in understanding the exact mechanism through which CANLUB prevents SCC for a number of reasons. First, it will allow fuel designers to ensure future changes to supply or manufacturing processes do not mitigate this effect.Second, an improved SCC model will further support reactor operation when the existing margins for SCC are tight. Third, the technology may be refined for advanced fuel designs such as DUPIC, MOX and slightly enriched fuel bundles.

2. Existing Fuel Models

Power ramp fuel defects occurred during the early history of CANDU operations. The industry therefore developed 'FUELOGRAMS' to predict the power ramp performance of fuel in CANDU reactors [4]. Both Ontario Hydro and AECL developed probabilistic models for the calculation of fuel defect probabilities, which define an operating envelop of power ramp size (ΔP) and ramped power (P_{max}) for CANDU fuel based on fuel burnup, and predict a probability of fuel failure if fuel is operated outside that envelop. Two distinctly different models emerged, an empirically based one called FUELOGRAMS and the other derived from statistical considerations called FULOMO;FUELOGRAMS were used at AECL while Ontario Hydro used the FULOMO correlation. In response to the need for a predictive tool for CANLUB fuel, daSilva developed the first CAnlubFuelomo Extended (CAFÉ) model in 1986 [4]. This initial formulation of CAFÉ did not find widespread use. However, the power ramp fuel defect experience at Pickering A in 1988 provided important new data on the performance of CANLUB fuel elements. These data were used to revise the CAFÉ formulation and a new tool for the

calculation of power ramp fuel defect probabilities for CANLUB fuel elements was issued in 1990 [1]. An SCC failure database was further compiled [6] [7].

Significant work has been ongoing at RMC to develop anapproachwhich models the specific mechanisms which cause SCC.Two codes in particular will form the basis of this new model:

1. A COMSOL based model developed by Kleckzek et al.[2] (SCC model)models the production of fission product iodine and its diffusion to the fuel-sheath gap. By determining the inventory of iodine present at a potential crack site it is able to determine if there is sufficient corrosive product to corrode through the sheath.
2. The Fuel And Sheath modelling Tool (FAST) is a COMSOL based fuel performance code.It models the thermal and mechanical response of the fuel and sheath system [5]. Of particular interest to this investigation, FAST models stress and strain in the sheath.

3. ProposedModel

A new code is being developed which models the mechanisms causing SCC. This includes modelling stress in the sheath for oxide layer cracking,the gettering effects of CANLUB impurities, specifically alkaline metals and ethyl cellulose binder residue, anda stress dependant critical crack depth to cause fracture.

3.1 Oxide Layer Cracking

A large set of power reactor data available in the open literature will be input to the SCC model to determine iodine inventories for each case. Based on failure due to iodine transport of zirconium away from a crack site and a assumingcylindrical cone crack with a critical volume of $2.8x10^{-9}$ cm^3[2], the iodine inventory can be compared to a critical iodine concentration required to cause sheath failure.

The cases where a critical iodine concentration is present will then be run in FAST to determine sheath stress. If failure did not occur, it can be assumed that the stress level in the sheath has not exceeded a critical stress sufficient to cause oxide cracking. In cases where failure did occur the stress rate must be greater than the critical stress level.By testing the entire set of reactor data and comparing which elements with a critical iodine concentration failed and which ones did not fail, a critical stress rate will be determined.

A recent improvement in the FAST code is an option to model a cracked UO_2pellet [6]. A sensitivity analysis will be conducted by running the power histories both with and without pellet cracking, in order to determine its impact on the critical stress.

A mechanistic approach to modelling crack growth will be investigated in parallel.

3.2 CANLUB Chemistry

The model will account for the gettering effect of CANLUB by considering both alkaline metalimpurities and ethyl cellulose binder residue. The concentration of impurities on the CANLUB layer (molecules/fuel element) will be calculated based on the thickness and impurity content:

$$N_{imp} = \sum(\frac{\omega_n}{A_n})\, \rho_{Canlub}\, t_{Canlub}\, SA_{Fuel}\, N_A [2] \quad (1)$$

Where $\omega_n$is the content by weight %of a particular impurityin CANLUB (g/g), $A_n$is the atomic weight of the impurity (g/mol), ρ_{Canlub} is the density of CANLUB (g/cm^3),t_{Canlub} is the thickness of the CANLUB layer (cm), $SA_{Fuel}$is the surface area of the fuel stack and N_A is Avogadro's number (atoms/mol).

The number of alkaline metal impurities in CANLUB, has been characterised by previous experiments[2]. The concentration of ethyl cellulose binder residue will be determined through chemical analysis at RMCC.

As a simplifying assumption it will be assumed that all impurities will react completely with the iodine. The number of molecules of impurities in the CANLUB will be subtracted from the number of iodine atoms present in the gap:

$$N_{Iodine} = N_{Iodine\ ,initial} - N_{imp} \quad (2)$$

It is known that CANLUB's effectiveness diminishes with increasing burnup[7]. Evidence suggests that by 350 MW·h/kgU the CANLUB layer no longer provides protection from SCC, likely due to radiation damage. To account for this decreasing effectiveness the number of impurities in the model will decrease with burnup. There is no data on the rate at which CANLUB effectiveness is lost, therefore a simple linear decrease will be assumed based on burnup (B):

$$N_{imp} = N_{imp} * \frac{350-B}{350} \quad (3)$$

Fracture Mechanics Model

The propagation of a stress corrosion crack in the sheath will increase hoop strain above the crack site, causing failure before corrosion propagates the entire thickness of the sheath. Based on calculations of the sheath stress, a fracture mechanics model could be implemented to determine a stress dependant critical crack depth to cause sheath rupture. The feasibility of including this effect in the model is currently being investigated.

4. Conclusion

The proposed model as presented in this paper will be an improved tool for modelling SCC. It will include explicit calculation of sheath stress and a more complete account of the relevant chemical ingredients in CANLUB.

5. References

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