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Reconsideration of Hydrogen Source Term in CANDUs to include Oxidation of Steel

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1. Introduction

A loss of coolant accident with a simultaneous loss of emergency coolant injection (LOCA+LOECC) is one of the critical design basis accidents that are analyzed for CANDU reactor licensing. The analyses are also used for development of hydrogen mitigating measures such as igniters and passive auto-catalytic recombiners (PARs). A failure of emergency core cooling (ECC) could result in extensive fuel damage but channel integrity has been assumed to be assured by the moderator acting as a heat sink with moderator cooling when this accident is considered within the design basis accident. In this scenario, although boilers are available as heat sinks, the fuel heatup occurs finally after LOCA due to eventual failure of secondary heat removal from primary coolant loss. This accident has typically been classified as a limited core damage (LCD) accident, distinguished thus from a severe core damage (SCD) accident where channel integrity is lost and overall core disassembly is preceded by a global loss of heat sinks including those at the moderator and the boilers.

In a CANDU LCD accident of a (LOCA+LOECC), the moderator cooling and steam generator cooling will likely be available but neither will be able to stop fuel bundles from heating up to high temperatures. Steam generator availability as a heat sink may help avoid a containment by-pass from tube creep ruptures and maintain low pressures by cooling any steam that flow through it. However there also are possibilities that this depressurization would promote in-leakage into the primary heat transport system (PHTS) from interfacing systems and hence promote longer periods of hot steam supplies for feeder oxidation.

Recall that moderator as an effective heat sink is insufficient that fuel heatup is not avoided if the primary coolant is lost as in a LOCA [1][2]. The 'heat-sink' function is in play only after a ballooned pressure tube comes in contact with calandria tube or if the pressure tube deforms at high temperatures into partial contact with calandria tube with fuel slumped inside at bundle deformation temperatures in excess of 1000°C. Depressurization of the PHTS after a pipe rupture (LOCA) will inhibit pressure tube ballooning unlike from high pressure scenario cases. In this case, the moderator can only become a heat sink when the depressurized pressure tube is hot enough to lose the fuel heat by radiation to submerged calandria tube. In all cases, the fuel remain hot enough to heatup any steam through it to high temperatures. Thus moderator presence or absence has little meaningful bearing on feeder oxidation concerns raised in this possibility study. Given that channel integrity is not lost in this scenario, feeder oxidation may continue as long as a source of water/steam is available while in a severe core damage accident, feeder oxidation may occur only as long as an integral flow path through the feeder is maintained. Therefore there could be design basis accident scenarios to which feeder oxidation may contribute more hydrogen than to severe accident scenarios.

For example, any steam flow through the channels could heatup to temperatures as high as 1500°C (depending on break size and time of onset of fuel heatup) and then transfer the heat to end fittings and feeders. Moderator water inventory as a heat sink, with or without cooling, could not affect peak fuel temperature predictions as long as water is present in moderator. Since appreciable rates of carbon steel oxidation occur at temperatures as low as 600°C, even a fuel heatup to temperatures where zircaloy oxidation has not started, can cause the feeders to oxidize and produce deuterium in quantities that cannot be ignored. This has been demonstrated in the comparison of steel and zircaloy oxidation kinetics in Fig.1. The technical basis of the argument is in the parametric analyses presented in the paper. This is consistent with the methodology used in the safety report where a stylized steam flow is used to arrive at a 'hydrogen' source term. However, as this paper is to urge reconsideration of hydrogen source term in CANDUs from oxidation of steel, the actual analyses of (LOCA+LOECC) will be undertaken afterwards with concrete integrated codes like CAISER [3] or ROSHNI [4] which model every fuel channel with consideration of its feeder flow resistant characteristics in relation to each other.

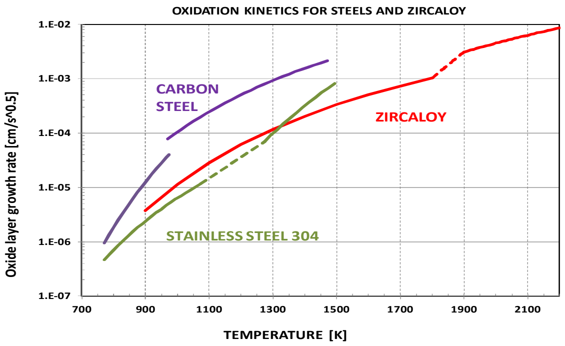

Fig. 1 Oxidation kinetics in steam for metals

In the PHWR licensing analyses, fuel thermal analyses are conducted by single channel models that assume a constant critical flow rate of steam into the channel that maximizes oxidation potential of the fuel string. The analyses are typically carried out for a simulation period of an hour during which the ‘hydrogen’ source term is calculated for a number of representative channels. These source term predictions are based on only fuel channel modeling using computer codes such as CHAN-II [5] or CATHENA [6]. These codes, however, does not try to model oxidation by steam and air of other reactor components, especially carbon steel feeders and main heat transport system piping as well as stainless steel end fittings. The hydrogen mitigating measures such as igniters and PARs in Wolsong are designed to avoid hydrogen explosions for both LCD and SCD which finally lead off-site source terms to be bounded within regulatory limits. In doing this, the hydrogen source terms at CANDU could have remained under-estimated if oxidation of low carbon, low chrome feeders that are susceptible to high rates of oxidation in steam and air is not considered.

Also, while CANDU measures for detection and mitigation are based on the more commonly understood lighter hydrogen isotope, LOCA accidents in CANDU reactors will produce only deuterium, a gas which has distinctly different properties [7] from hydrogen and hence requires dedicated measures, not entirely identical to those for hydrogen. The emphasis in our proposal is therefore on best estimates of production of deuterium gas by oxidation of fuel channel components, end fitting components and feeders with appropriate consideration of available heat sinks and production of appropriate gases.

2. Steel Oxidation Kinetics

The following is an overall summary of feeder data for a CANDU-6 reactor as at Wolsong:

- Material : Low carbon, Low Cr steel (max 0.4%; actual as low as 0.04%); SA106- Grade B
- Total mass:~35,000 kg inlet, 68,000 kg outlet
- Total length : ~4,300m inlet, 5,000m outlet
- Surface area: ~775 m^2 (inlet feeder internal surface), 1,075 m^2 (outlet feeder internal surface)

In this analysis, only internal oxidation by steam is considered and oxidation kinetics is typically modeled as:

$$K_p = Ae^{(Q/RT)}$$

Where,

- k_p= rate of weight gain by oxidation [$g^2/cm^4.s$]
- R = Universal gas constant [J/mol.K]
- A = Arrhenius number [$g^2/cm^4.s$]
- Q = activation energy [J/mol]
- T = temperature [K]

The detailed and discrete correlations [8][9] are given as:

$k_P = 5.87 \times 10^3 e^{\left(-\frac{230000}{RT}\right)}$, for 773<T<973 K

and

$k_P = 3.047 e^{\left(-\frac{157539}{RT}\right)}$, for T > 973 K.

During oxidation, a multi-layer oxide exists (Wustite FeO, Hematite Fe_2O_3, and Magnetite Fe_3O_4) on steel surfaces and for carbon steel a ratio of 95:4:1 exists between these oxides. It is shown in Fig.1 that the steel oxidation by steam starts at lower temperatures; is faster than oxidation of zircaloy at the same temperature; and is almost as energetic with exothermic heat of reaction of ~272 kJ/mole and thus cannot be ignored.

3. Demonstrative Simulation Results

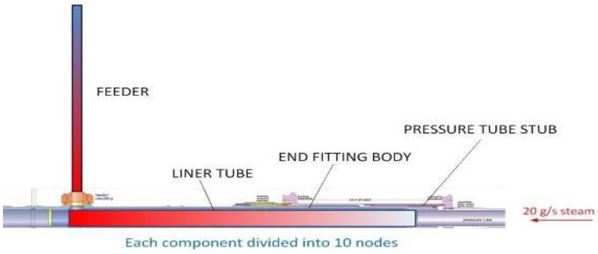

Fig. 2 Detailed Modeling Demonstration

The thermal transient of the end fittings and feeders (see Fig.2) is a function of the steam conditions at the exit of channels. For illustration purposes, only stylized simulations for a constant discharge of steam into the end fitting from fuel over a 3 hour period are presented. This captures the effect of feeder and end fitting thermal and oxidation behavior. It is shown in Fig. 3 and 4 that feeder metal temperatures are raised sufficiently high in about an hour for the high steam temperature of 1200°C at 20g/s, to start exothermic oxidation and production of hydrogen. For this stylized scenario, one outlet feeder produces about 0.5 kg of deuterium in less than 3 hours.

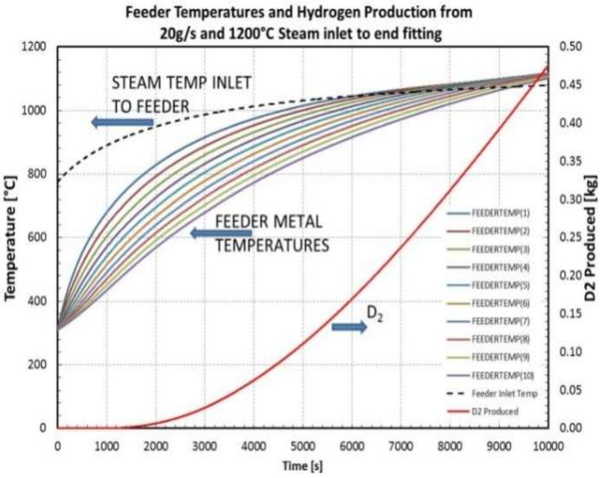

Fig. 3 Feeder end fitting thermal response (for 20g/s 1200°C steam)

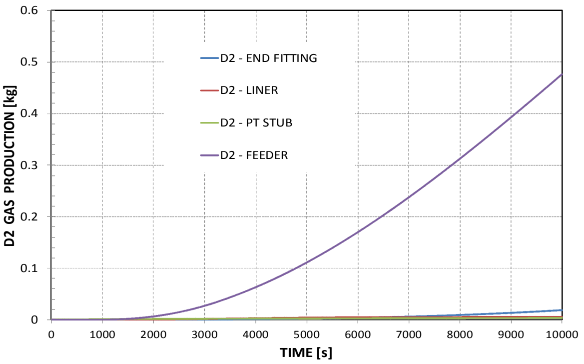

Fig. 4 Deuterium production from feeders, end fittings and the pressure tube stub (for 20g/s 1200°C steam)

Scoping analyses have shown that in less than 12 hours deuterium from oxidation of 380 feeders could exceed deuterium production from fuel and pressure tube oxidation of zircaloy. This does not include oxidation of external surfaces of feeders by air ingress into feeder cabinets.

4. Discussion and Future Work

A stand-alone methodology using detailed material and fluid (D_2O) properties and a discrete end fitting and feeder heatup model has been addressed and employed to illustrate the magnitude of feeder oxidation under the stylized conditions used for (LOCA+LOECC) analyses. While a LOECC event implies loss of water inventory without makeup, a total loss of liquid water from the heat transport circuit prior to significant fuel heatup would not occur immediately. A large inventory of water can be held up in end fittings and large U-shaped piping between the boilers and the pumps. And leakage of water into the depressurized heat transport system from the interfacing systems cannot be precluded. Therefore, all high temperature steam interaction phenomena, including exothermic oxidation of carbon steel feeders etc., needs to be studied. Also, the actual source term for combustible deuterium may be different from that of previously assumed hydrogen species. Hydrogen mitigation measures, such as igniters and recombiners have been sized according to the predicted source term that does not include hydrogen source term from steel oxidation. As a result, the effectiveness of 'hydrogen' detection and mitigation systems needs to be re-examined and a potential for containment failure due to gas explosions needs to be reconsidered as well.

In the meantime, carbon steel oxidation at the outside surfaces of feeders, normally enclosed inside feeder cabinets is not considered in this study. But the feeder cabinet under severe accident circumstances is not expected to survive high temperatures and it is not unlikely that the feeders will oxidize on the outside. Therefore, carbon steel oxidation by steam at the inside surfaces and air at the external surfaces (exposed to containment atmosphere after the insulation in the feeder cabinet burns off) have to be considered together. Furthermore, the feeders are known to be rusted during normal operation (see Fig.5) from moisture and their oxidation after the core damage accident associated with a (LOCA+LOECC) must consider oxidation with consideration of water vapor presence. Increase of oxidation rates for almost all metals in presence of steam [10] is easily expected.

Fig. 5 Rusted feeder during normal operation

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