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Diffusion of hydrogen through the walls of carbon steel pipes experiencing flow-accelerated corrosion under hydrothermal conditions.

D.S. Mancey, R.J. Keyes, M.S.L. Godin, J. M. Smith and D.G. Webb
Atomic Energy of Canada Limited
Chalk River Laboratories, Chalk River, Ontario, K0J 1J0

In the 1950's M. C. Bloom and co-workers at the Naval Research Laboratories (Washington, DC, USA) developed the *hydrogen effusion technique* for measuring the rate of carbon steel corrosion in aqueous systems at elevated temperature and pressure (1). The method was based on the measurement of the hydrogen generated by the corrosion reaction, and involved the collection and measurement of the hydrogen that diffuses through the walls of a sealed steel vessel containing the aqueous solution. Most of the work performed by Bloom and his co-workers involved the study corrosion in aqueous solutions that were stagnant.

Since 2003, work has been conducted at Chalk River Laboratories (CRL) to apply the concepts of Bloom to the study of flow-accelerated corrosion (FAC) of steel under hydrothermal conditions. Of particular interest is the FAC process that occurs in outlet feeder pipes in the primary heat transport system (PHTS) of CANDU reactors (2). The inlet feeder pipes supply the heavy-water (D_2O) coolant to the reactor core at a temperature of about 265°C. After passing through the core and attaining a temperature of about 310°C, the coolant exits the core into the outlet feeders. During reactor operation, the outlet feeder pipes experience FAC in the high-velocity coolant. The FAC rate in outlet feeders are affected by flow conditions and range from about 0.04 mm/a in straight pipe to about 0.17 mm/a near some bends.

The chemistry of the coolant in the PHTS is controlled so as to maintain a dissolved D_2 concentration of 3-10 mL(STP)/kg. In principle, hydrogen atom absorption can arise from corrosion or from the dissociation of dissolved D_2 on the pipe surface. Laboratory tests were performed under simulated PHTS conditions to determine the relative significance of these sources for carbon steel experiencing FAC. The tests demonstrated that corrosion was the dominant source of the absorbed hydrogen, and that the rate of FAC could be calculated from the flux of hydrogen passing through the pipe wall. For carbon steel, the hydrogen produced by corrosion appears to be absorbed with near 100% efficiency. The introduction of low concentrations of dissolved oxygen was observed to completely suppress hydrogen absorption. Similar tests are in progress with selected stainless steels.

Devanathan-Stachurski cells have been used extensively since the 1950's to study the permeation of atomic hydrogen through metal membranes. Commonly, these electrochemical cells have been used to perform experiments with aqueous electrolytes at temperatures < 100°C. The results from such experiments are reviewed and compared to the results of the experiments performed under hydrothermal conditions at CRL. Particular attention is given to the efficiency of the hydrogen atom absorption process at the membrane-electrolyte interface.

The experiments performed at CRL led to a number of significant developments. Firstly, the development of a device for measuring the hydrogen flux passing through the wall of a feeder pipe of an operating CANDU reactor (3). Secondly, the recognition that a significant flux of atomic hydrogen continually passes through the wall of outlet feeder pipes during CANDU reactor operation. This state of affairs has important implications for the ageing of feeder pipes and the susceptibility of feeder pipes to cracking (4).

Lastly, consideration is given to the implications of the CRL experiments with regard to the mechanism of carbon steel corrosion under hydrothermal conditions, and the properties of the magnetite (Fe_3O_4) film present on the steel surface under such conditions.

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