MODELLING OF SPATIAL RADIOLYSIS IN PRIMARY WATER

Background

The radiolysis of water in LWRs (light water-cooled reactors) by gamma, neutron and alpha radiation leads to the production of molecular (H_2O_2, H_2) and radical (H, OH, e^-(aq), H^+) species. These species subsequently react and achieve steady state concentrations. The concentrations of the oxidizing species (O_2, H_2O_2) and dissolved hydrogen significantly influence the corrosion potential of an alloy exposed to the primary coolant and, hence, affect the resistance of the alloy to localized corrosion and stress corrosion cracking particularly.

Objectives

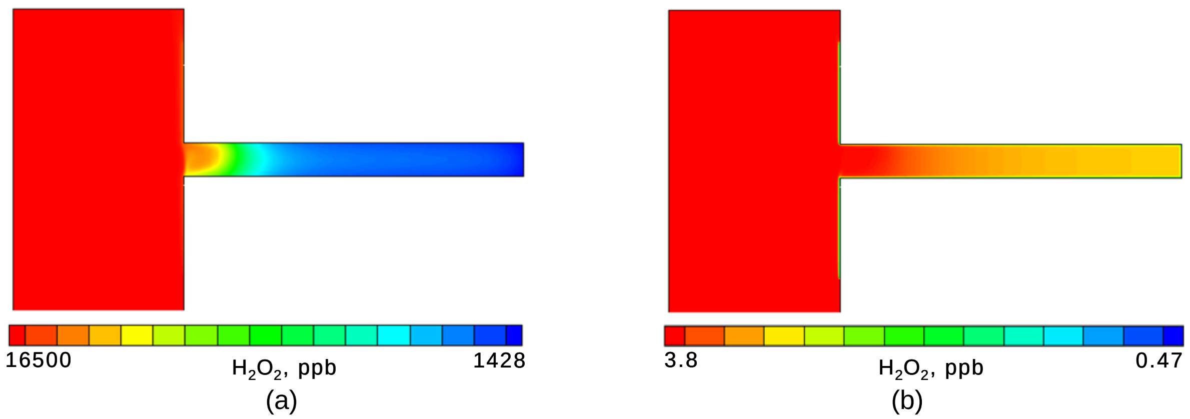
A common approach to estimate the corrosion potential of in-core components is numerical calculation. The steady state concentrations of dissolved hydrogen, oxygen and hydrogen peroxide are estimated by conventional radiolysis codes. These concentrations are used afterwards to calculate the corrosion potential. However, the analytical expressions employed in the calculations for approximation of the mass transport do not take into account radiolysis in the diffusion layer. This can cause significant errors in the estimation of the corrosion potential even for a simple electrode geometry; hence also for occluded regions like baffle bolt crevices. Therefore, to improve the numerical calculation of radiolysis one should take into account simultaneously mass transport effects and radiolysis. This approach has been called spatial radiolysis calculations.

A typical PWR radiolysis model contains some 50 chemical reactions involving some 20 species. The implementation of all these reactions into a finite element code in order to calculate mass transport and radiolysis simultaneously has not been successful. However, a reduced-reaction-set of 17 radiolysis reactions to be used in PWR (Pressurised Water Reactors) water was suggested by J. Henshaw (SERCO) and is a reasonable approximation around pH 7.

Principal results

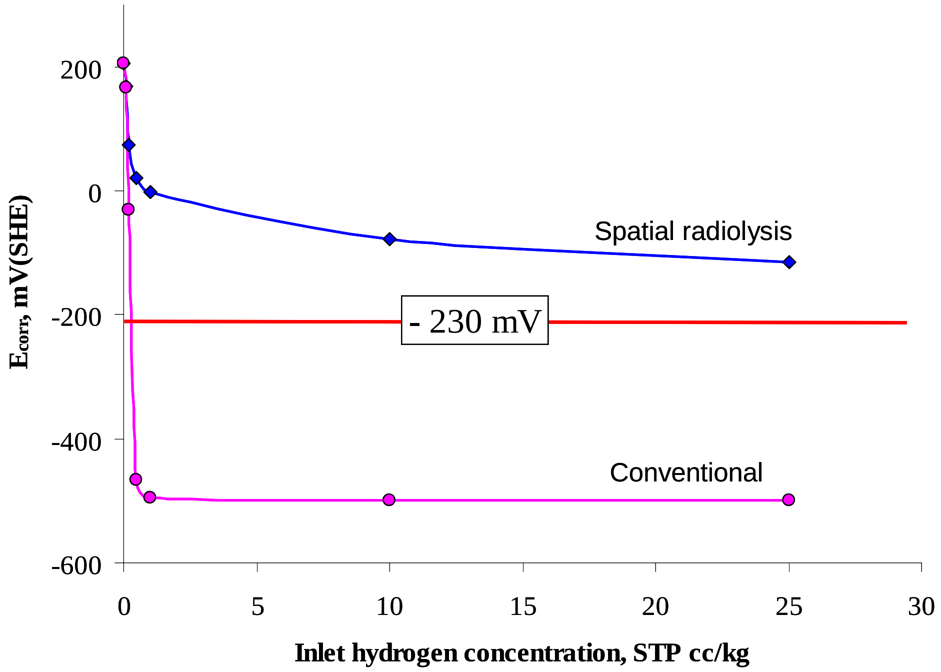
The reduced-reaction-set radiolysis has been implemented in two finite element codes: CMPH (COMSOL MULTIPHYSICS) and MIOTRAS (Multi IOn TRAnSport). The results of bulk composition calculations are in good agreement with those calculated by the original radiolysis codes.

We used the developed finite element framework for the investigation of radiolysis effects for a number of cases. One of it is a simulation of the electrochemistry in a crevice geometry. The left picture shows that the concentration of hydrogen peroxide generated by radiolysis decreases in the crevice due to its consumption by electrochemical reactions on the metal crevice surface and due to limited mass transport. Addition of hydrogen to the feed water significantly suppresses the production of oxidising species due to radiolysis. The right picture shows that the addition of 1 cm³/kg dissolved hydrogen to the feed water reduces the concentration of hydrogen peroxide in the bulk water from about 16500 to 3.8 ppb.



Distribution of hydrogen peroxide in crevice geometry for two different concentrations of dissolved hydrogen in the feed water: (a)0.01 cm³/kg; (b) 1 cm³/kg

Radiolysis in the diffusion layer close to the metallic surface plays a significant role in the accuracy of the prediction of the corrosion potential. The difference is illustrated in the picture below. One can see that conventional radiolysis predicts the corrosion potential for a creviced structure below -230 mV (SHE) (usually taken as the threshold for SCC) for an inlet dissolved hydrogen concentration less then 0.3 cm^3/kg in the feed water. The corrosion potentials calculated by spatial radiolysis are higher than the threshold potential for all the calculated feed water concentrations of dissolved hydrogen. Further simulations, taking into account decomposition of hydrogen peroxide on the metallic surface are to be performed to confirm the latter.



The corrosion potential of a crevice structure as function of the dissolved hydrogen concentration in the feed water. The red line represents the conventional threshold potential for SCC.
blue curve – corrosion potential calculated with spatial radiolysis model (developed at SCK•CEN).
red curve – corrosion potential calculated by conventional approach with imposing of calculated radiolytic species concentration in the bulk only.

The activity is sponsored by the European Community integrated project PERFECT (fp6perfect.net) and the Convention SCK•CEN-Electrabel.

Future developments

The major aim of our work is to predict the electrochemistry in occluded regions inside a nuclear reactor e.g. in a baffle bolt crevice. In order to do that temperature transients during reactor operations and temperature gradients inside of the crevice should be taken into account. Therefore coupling of mass transfer and heat transfer phenomena should be the next step in the developing of our numerical simulations.

Main contact persons

Serguei Gavrilov, serguei.gavrilov@sckcen.be; Marc Vankeerberghen marc.vankeerberghen@sckcen.be

Main reference

M. Vankeerberghen, S. Gavrilov, Radioelectrochemistry calculations in respect of stress corrosion cracking, Part I - Implementation of radiolysis models in finite element codes, Restricted Contract Report R-4334, Convention Electrabel.