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# F.E. modelling of hydrogen migration and blister formation in PHWR coolant channels

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ABSTRACT: The formation of a cold spot in pressure tube due to its contact with calandria tube of a PHWR coolant channel results in the migration of Hydrogen in pressure tube towards contact zone from its surrounding material. A 3-D finite element code SPARSH is developed to model the hydrogen redistribution and consequent hydride blister formation due to thermal and Hydrogen concentration gradients. In the present paper, the details and performance of this code are presented..

## 1 INTRODUCTION

Hydrogen (The term Hydrogen includes Deuterium) migrates under the influence of concentration and thermal gradients in Zirconium alloys. The pressure tubes of coolant channels of a PHWR (Pressurised Heavy Water Reactor) are made of Zirconium alloys. In the event of a cold spot arising out of pressure tube coming in contact with calandria tube, the consequences of hydrogen migration in these coolant channels can be severe as,it may result in the formation of brittle Zirconium hydride blisters at the cold spot. This mechanism was indeed found to cause the failure of pressure tube in Pickering-2 reactor in 1983. The events leading to such failure start with a localised contact between pressure tube and calandria tube at some points along the channel length. These contacts are primarily caused by the movement of garter springs and the effect of irradiation on creep sag of coolant channels. This leads to a cold spot at these contact points. Hydrogen migrates towards these locations. When the hydrogen concentration exceeds its terminal solid solubility (TSS), Hydrogen precipitates in the cold regions eventually forming blisters of brittle Zirconium hydride, under appropriate conditions. These blisters may act as initiation sites for cracks which would propagate by delayed hydride cracking mechanism. To analyse this phenomenon, it is important to study the migration of hydrogen in pressure tube of coolant channels. This requires the solution of governing diffusion equation of hydrogen migration in Zirconium alloys. In the past few decades, Finite element method has been proved to be a very versatile method for solution of continuum problems formulated by differential

equations. A 3-D finite element code SPARSH is developed for solving the governing equation for Hydrogen diffusion using finite element method. In the present paper the formulation and implementation details of this code and results obtained for a typical 220 MWe PHWR coolant channel are presented.

## 2 FORMULATION

The governing equation for hydrogen migration under the influence of thermal and concentration gradients is given by Sawatzky et al (1963).

$$\frac{\partial C}{\partial t} = \nabla\, V_s\, D \left\{ \nabla C_s + \frac{C_s\, Q^*}{R\, T^2} \nabla T \right\} + q \qquad (1)$$

Where
$C$ = total Hydrogen isotope concentration ; $T$ = temperature
$C_s$ = concentration in solid solution ; $t$ = time
$D$ = diffusion coefficient ; $R$ = gas constant
$q$ = Hydrogen ingress from operating environment
$V_s$ = volume fraction of isotope in solid solution
$Q^*$ = heat of transport

The diffusion coefficient is related to temperatures by the following relation

$$D = D_o\, e^{-Q/RT} \qquad (2)$$

Where $D_o$ is frequency factor and $Q$ is activation energy.
Defining normalised concentrations $S$ and $S_s$ as

$$S = \frac{C}{e^{[Q^*/RT]}} \quad \& \quad S_s = \frac{C_s}{e^{[Q^*/RT]}} \qquad (3)$$

The governing diffusion equation (1) can be modified to the following form (Byrne et al, 1985) with the help of relations (2) and (3) as

$$e^{Q^*/RT} \frac{\partial S}{\partial t} = \sum_{i=1}^{3} \frac{\partial}{\partial x_i} \left[ V_s\, D_{xi}\, e^{Q^*/RT} \frac{\partial S_s}{\partial T} \right] + q \qquad (4)$$

The determination of Hydrogen redistribution in a coolant channel involves the solution of the above equation subjected to specified initial and boundary conditions. It is important to note from equations (1) and (4) that only the Hydrogen available in solution form can take part in migration. The amount of Hydrogen in solution form is limited to its terminal solid solubility (TSS). All the Hydrogen in excess to this TSS will be available in precipitate form. Also, as the Hydrogen concentration in a hydride precipitate is assumed to be 16000 ppm, the maximum concentration at any point is limited to this value. Hence while solving equation (4), the following conditions must be satisfied.

$C_s = C$ for single phase (Hydrogen in solution form only)
$C_s$ = TSS for two phase (hydrogen in solution + hydride forms)

$C_{max}$ = 16000 ppm

The terminal solid solubility is a function of temperature according to the following relation.

$$TSS = C_T \, e^{-H/RT} \qquad (5)$$

Where $C_T$ is a constant and H is heat of mixing. It has been observed (Slattery, 1967) that the terminal solid solubility values for common Zirconium based alloys are quite different for precipitation and dissolution processes. For a given Hydrogen concentration, hydride precipitation occurs at a lower temperature than hydride dissolution. A comprehensive modelling of Hydrogen migration should consider this hysteresis effect properly.

## 3 DESCRIPTION OF FINITE ELEMENT MODEL AND SOLUTION SCHEME

Petrov-Galerkin method has been used to convert the governing hydrogen diffusion equation (4) into a set of simultaneous equations forming finite element characteristic matrices. These three characteristic matrices [Ce], [Ke] and {Pe} are related by the following expression

$$[C_e] \{\partial S/\partial t\} = [K_e]\{S_s\} + \{P_e\} \qquad (6)$$

The matrix [Ce] is obtained through the term attached to the derivative of the vector {S} in Eqn. (4). The matrix [Ke] is associated with the concentration and thermal gradients. The vector {Pe} is a load vector to consider the Hydrogen ingress at the surfaces.

These characteristic matrices and their relationship given by (6) are analogous to those obtained while solving transient heat conduction equation using finite element method. Whereas in transient temperature analysis, the elements of vectors {S} and {Ss} refer to single entity of nodal temperatures, in the present analysis these two vectors do not always refer to single hydrogen concentration value. Depending on the concentration of Hydrogen isotope at the nodal points of a particular element, one of the following three situations will arise.

1) For an element with its nodes containing single phase hydrogen, the vectors {S} and {Ss} are the same because all the Hydrogen is available in solution form. This results in a situation identical to transient temperature calculations. The characteristic matrices [Ce] and [Ke] will be symmetric . Direct integration techniques used for solving this transient problem requires a particular combination of these two matrices. The resulting coefficient matrices will be symmetric.

2) For an element with its nodes containing two phase hydrogen, the entities in {Ss} will be equal to the terminal solid solubilities corresponding to the nodal temperatures and hence are predetermined. Hence the product [Ke]{Ss} will yield a known vector, similar to load vector. Since the matrix [Ce] is a symmetric matrix, the resulting coefficient matrix for transient analysis is also symmetric.

3) For an element with some of its nodes containing single phase Hydrogen and rest of the nodes two phase Hydrogen, a complex situation arises. The vector {Ss} consists of known entities corresponding to nodes containing two phase Hydrogen and unknown entities corresponding

to nodes containing single phase hydrogen. Here the product of characteristic matrix [Ke] and vector {Ss} on the right hand side of Eqn.(6) is split into a load vector corresponding to known entities of {Ss} and an unsymmetric characteristic matrix corresponding to single phase nodes. The combination of this matrix along with [Ce] results in unsymmetric coefficient matrix for this particular case.

## 4 FINITE ELEMENT CODE 'SPARSH'

A finite element code SPARSH is developed for modelling hydrogen migration and blister formation based on the above principles. The organisation of this code runs on the familiar lines of a typical finite element program where, at first, characteristic matrices are formed, assembled and then solved for unknowns. This code can analyse hydrogen redistribution in plane, axisymmetric and 3-D structures. Linear and higher order Isoparametric elements such as 4/8 noded 2-D elements and 8/20 noded 3-D brick elements have been implemented in this code. Since unsymmetric coefficient matrices are generated for the elements in single phase/ two phase transition zone, the conventional banded solver has been suitably modified for this purpose. The other important features of this code are

1) Use of direct integration techniques for solving transient equations (Cook, 1988).
2) Consideration of Dirichlette and Neumann conditions such as Hydrogen flux and concentration boundary conditions.
3) Use of upwinding algorithm for suppressing boundary fluctuations in the case of application of forced boundary conditions.
4) Consideration of separate terminal solid solubilities of Hydrogen for precipitation and dissolution.
5) This code has got a separate module for 3-D temperature calculations. This facilitates simultaneous iterative calculations of temperature and hydrogen distributions.

## 5 CASE STUDY

The code SPARSH has been benchmarked against published literature (Sawatzky,1985). The code is then used to study the Hydrogen redistribution and consequent blister formation in a typical 220 MWe Indian PHWR coolant channel assuming pressure tube - calandria tube contact. The temperature distributions for different thermal boundary conditions and contact geometries in the pressure tube and calandria tube have been reported elsewhere (Bhasin et al, 1994). One temperature distribution corresponding to a 4 mm contact spot diameter, 275 $^{\circ}$C bulk coolant temperature and contact conductance of 0.068 W/mm$^2$-K has been used in the present analysis. The initial Hydrogen concentration in the pressure tube is taken as 60 ppm. Since the contact area for this case is circular, the temperature distribution is symmetric about the pressure tube thickness at the contact spot. Hence the pressure tube is initially analysed as an axisymmetric case. The results obtained for this case are shown in Fig.1. The Y-axis represents the thickness direction of pressure tube. This Figure shows the change in Hydrogen concentration and growth of blister near pressure tube / calandria tube contact up to nine months of hot operation. The four quadrants of this figure numbered as 1,2,3 & 4 show the concentration profiles after 2.25, 4.5,

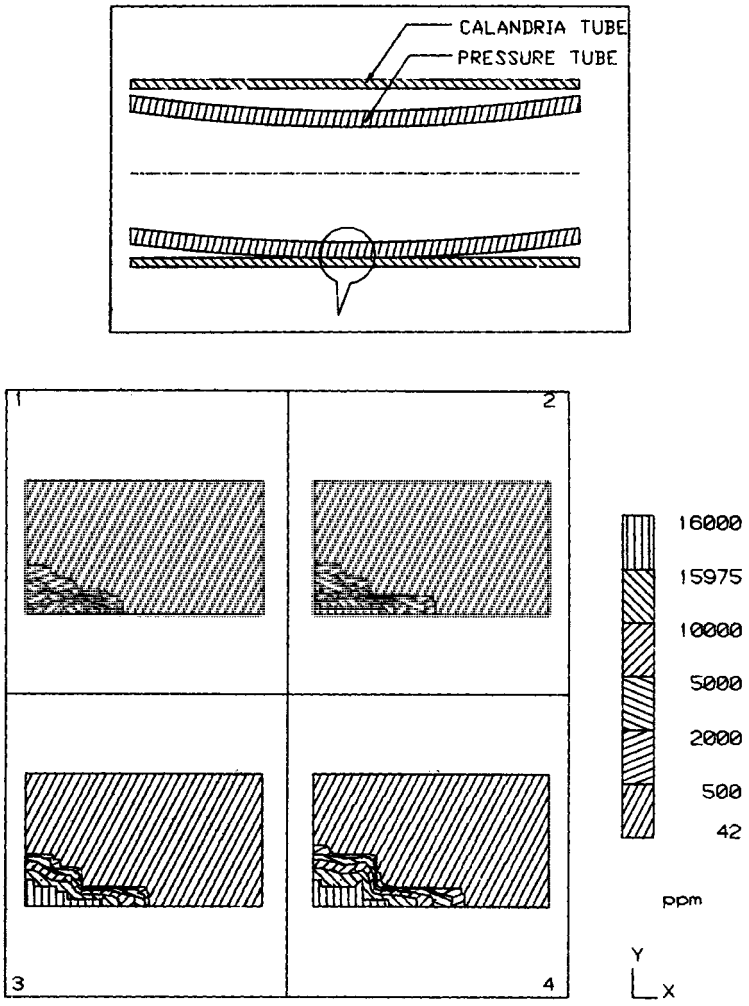


FIG. 1 2D HYDROGEN CONCENTRATION DISTRIBUTION IN PRESSURE TUBE AFTER 2.25, 4.5, 6.75, & 9 MONTHS OF REACTOR OPERATION

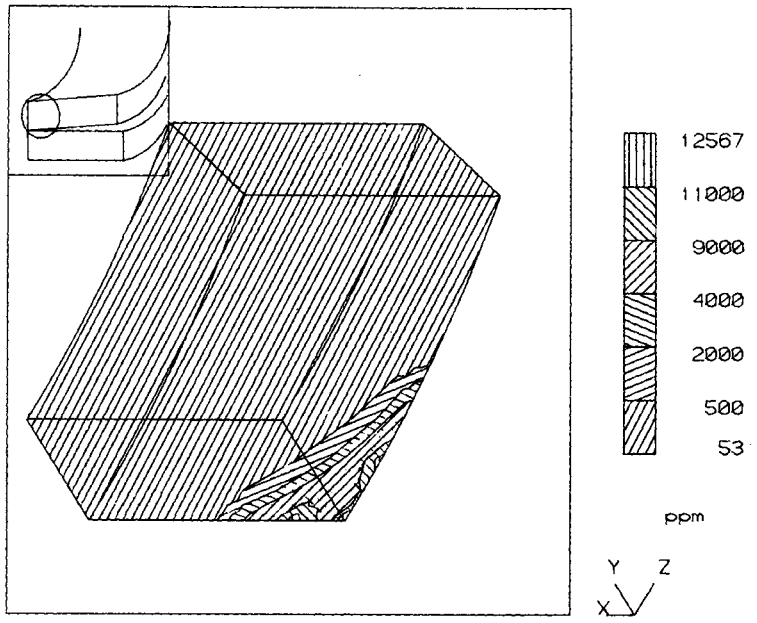


FIG.2 3D HYDROGEN CONCENTRATION DISTRIBUTION IN PRESSURE TUBE ON THE SURFACE OF CONTACT AFTER 60 DAYS OF REACTOR OPERATION

6.75 and 9 months of hot reactor operation respectively. The same problem has later been solved as a 3-D case by the present code to illustrate the 3-D capabilities. Figure.2 shows the Hydrogen concentration profiles at the end of 60 days of reactor operation. The symmetricity in the concentration profiles in this Figure justifies the adequacy of 2-D axisymmetric analysis for circular contact conditions. However, for non-circular contact geometries, 3-D analysis needs to be performed and the code SPARSH can be used for this purpose effectively.

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