

# EFFECT OF STRESS STATE AND TEXTURE ON DELAYED HYDRIDE CRACKING KINETICS IN REACTOR PRESSURE TUBES

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An experimental study based on the acoustic emission technique was carried out to examine how delayed hydride cracking (DHC) propagation kinetics was influenced by stress states using constant K specimens machined in two orientations from Zr-2.5 wt% Nb reactor pressure tube alloy, of 2 mm, 4 mm and 8 mm thickness.

DHC velocity was reduced by changing the stress state from plane strain to plane stress at a constant stress intensity of 15 MPa $\sqrt{m}$  and a hydrogen level of 50 ppm. Changing orientation of the zirconium hydride habit plane dictated whether DHC occurred or not.

An explanation of how the observations made on specimens can be related to observations made on pressure tube is presented.

## INTRODUCTION

The incidence of crack tunnelling by delayed hydride cracking (DHC), is observed in the Zr-2.5% Nb (Zr-Nb) alloy used for pressure tubes in CANDU (CANada Deuterium Uranium) power reactors. DHC can propagate from the inside surface of the tubes by a tunnelling mechanism and grow until significant leaking occurs. Only after removal and examination of the tube can the true extent of this DHC by tunnelling be determined.

The present work is an examination of how the state of stress at an advancing DHC front can influence the extent of this crack tunnelling, with the aim of discouraging DHC tunnelling and promoting leak-before-break. The influence of crystallographic texture on DHC is also considered in this highly anisotropic material.

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DHC AND STRESS STATE

DHC in Zr-Nb is considered as a diffusion-controlled fracture mechanism, such that the flux of hydrogen to a notch tip is governed by the hydrostatic component of the stress  $\sigma_{ii}$  (1,2,3). Propagation of DHC is by the precipitation or reprecipitation of hydride ahead of a notch, accompanied by cracking of this hydride under the action of the applied stress and failure of the intervening matrix by a ductile fracture process. The mechanism is sustained by the flux of hydrogen along the stress gradient.

In plane strain,

$$\frac{\sigma_{ii}}{3} = \frac{\sigma_{11} + \sigma_{22} + \sigma_{33}}{3}$$

and in plane stress,

$$\frac{\sigma_{ii}}{3} = \frac{\sigma_{11} + \sigma_{22} + (\sigma_{33} = 0)}{3}$$

Also, in plane strain,

$$\sigma_{11} > \sigma_{33} > \sigma_{22}$$

and in plane stress,

$$\sigma_{11} > \sigma_{22} > \sigma_{33} (= 0)$$

Further, the Tresca yield criterion gives, in plane strain,

$$\sigma_{11} - \sigma_{22} = \sigma_y$$

$$\sigma_{11} + \sigma_y = \sigma_{22}$$

and in plane stress,

$$\sigma_{11} - \sigma_{33} (= 0) = \sigma_y$$

$$\sigma_{11} = \sigma_y$$

Thus  $\sigma_{ii}$  is always greater in plane strain than in plane stress. Changing the axiality of the stress state should then lead to changes in the shape of the advancing DHC front and changes in DHC velocity.

## DHC AND CRYSTALLOGRAPHIC TEXTURE

In extruded pressure tubes, about 80% of grains have their base plane normals in the circumferential direction of the tube (4). The habit plane for hydrides precipitated during DHC is almost parallel to the (0001) in the Zr-Nb (4). The hydrides present from extrusion are orientated along the extrusion direction (4). It is therefore relatively easy for hydrides which are taken into solution at reactor operating temperatures to precipitate, on cooling, on their habit planes to provide an easy path through the tube wall for DHC propagation.

## MODEL MATERIAL

In order to examine the influence of stress state and texture, it was thought that sheet with the same microstructure as tube would provide the ideal model material to study. Accordingly, sheet was produced under the same thermomechanical conditions as tube with the exception that plane strain deformation by rolling was employed rather than the mixed plane strain and shear due to extrusion. In this way, sheets of 2 mm, 4 mm and 8 mm thicknesses were produced which exhibited an almost identical microstructure to that of extruded tube, i.e. the phase nature and distribution were almost the same and the basal plane normals were concentrated in the transverse direction of the plate, akin to their concentration in the circumferential direction of extruded tube.

## EXPERIMENTAL PROCEDURE

Constant K specimens (CKS), with  $W = 17\text{mm}$  (5) were machined from the 2 mm, 4 mm and 8 mm thick sheets. Specimens were produced such that the notches were parallel to the rolling direction. Loading was therefore in the "hard" direction of this highly anisotropic material (4).

Some specimens were also manufactured with notches transverse to the rolling direction.

The rigs used for the experiments were essentially standard stress rupture machines with grips modified to accommodate the CKS. A flat was ground on the pull rods to allow a transducer to be mounted for crack monitoring by acoustic emission (4).

The specimens were hydrided to about 50 ppm by heating in hydrogen gas, quenching and redistributing the formed hydride by subsequent heating at 673 K for three days. Each specimen was then loaded into the rig, heated to 593 K for 1 h, cooled to 498 K for 1 h and then loaded to a stress intensity of 15 MPa $\sqrt{\text{m}}$ . This procedure allowed dissolution of the hydride, diffusion of hydrogen along the stress gradient and precipitation of the hydride at the notch tip to take place. Crack monitoring was by acoustic emission and interruption of the test to measure the surface crack length, where possible, and note the time taken for a given amount of crack extension. The heating and loading sequence was repeated each time. Final separation of the surfaces was achieved by increased loading in the DHC rig. The fractured surfaces were examined by scanning electron microscopy (SEM) to measure the crack extension and determine the shape of the crack fronts.

Sections were cut through the centre of the notch, in some specimens, and examined by optical microscopy.

EXPERIMENTAL RESULTS

DHC velocities were determined using the method described previously by Coleman (4) and the following results obtained from specimens machined with their notches parallel to the rolling direction.

1. 8 mm thick specimens: average DHC velocity  $1.51 \text{ E-}08 \text{ m/s}$ .
2. 4 mm thick specimens: average DHC velocity  $1.82 \text{ E-}08 \text{ m/s}$ .
3. 2 mm thick specimens: average DHC velocity  $1.41 \text{ E-}09 \text{ m/s}$ .

In the 4 mm and 8 mm thick specimens, conditions of plane strain prevailed such that the shape of the advancing DHC crack front was approximately 90% square fracture in each case, Figure 1.

In the 2 mm thick specimens, conditions of plane stress prevailed such that the shape of the DHC front was about 50% square fracture, Figure 1.

Examination of the fracture surfaces by SEM revealed regions where propagation had occurred by DHC and these areas were discreet from the areas where separation had occurred by overloading at the end of a test, Figure 2. Tests performed on specimens machined with notches transverse to the rolling direction did not crack, at least not after 960 h, even when the test temperature was lowered to 423 K. However, in these tests, the 4 mm and 8 mm thick specimens did exhibit some reorientation of hydrides in the vicinity of the notch tip, Figure 3.

DISCUSSION

In the specimens machined with notches transverse to the rolling direction, DHC did not take place because the habit plane for hydride reprecipitation was not favourably orientated, demonstrating the potent effect crystallographic texture had on this mechanism. Therefore, in pressure tubes, if the texture was such that about 80% of the grains had their basal planes in the circumferential direction, then DHC should prove extremely difficult to initiate.

When DHC took place, higher crack velocities were observed when  $\sigma_{II}$  was greater, ie: under conditions of plane strain. Decreasing the axiality of the stress state decreased the crack velocity, but gave rise to a situation where DHC advanced by crack tunnelling in these specimens, under conditions of plane stress.

This latter observation is apparently at variance to what is observed when DHC takes place in pressure tubes. In that case, DHC spreads from the initiation point in a lenticular-shaped manner, appearing to slow down near the tube surface, but tunnelling outward in the central regions of the tube cross-section.

In an attempt to explain this apparently anomalous behaviour, an 8 mm thick specimen was allowed to fail by DHC until after plastic collapse of the remaining ligament of the specimen occurred. The specimen was subsequently loaded to failure and the fracture surfaces examined. In this specimen, DHC had advanced by 90% square fracture as before, until plastic collapse took place. At this point DHC began to advance an order of magnitude slower and by 50% square fracture. As plastic collapse occurred the state of stress at the crack tip changed from plane strain to plane stress.

Thus tunnelling observed in pressure tubes in CANDU power reactors can be explained in terms of stress state at the advancing DHC front. DHC will proceed through the tube wall until plastic collapse occurs and the conditions at the crack tip are those of plane stress. The onset of plastic collapse may be encouraged by sufficiently high tensile residual stresses. That is, general yield will occur under normal tube operating conditions at lower applied stresses than if tensile residual stress were not present.

Therefore, for a given applied stress and increasing residual tensile stresses, the width of the ligament sustaining the general yield load will increase. This has been observed in practice (6). Further, DHC will tunnel in the direction of the hydrogen flux driving force, that is to where  $\sigma_H$  is greater, away from the surface. Also, DHC has been reported to initiate at points of high tensile residual stress on the inside surface of tubes (4). Under these circumstances, the residual stress redistributes as soon as initiation has taken place and DHC will grow away from this initiation surface as before, giving rise to the lenticular appearance of DHC tunnelling as observed in pressure tubes.

In order to reduce DHC tunnelling, steps should be taken to increase the yield strength of pressure tubes at reactor operating temperatures and to reduce or eliminate tensile residual stresses at or near the tube surfaces. This action should therefore promote leak-before-break (7) and discourage DHC tunnelling resulting in an improved tube for CANDU reactors.

#### CONCLUSIONS

A description of DHC tunnelling observed in pressure tubes has been derived in terms of the stress states prevailing during DHC propagation in a model material.

DHC tunnelling can be discouraged by higher yield strength tubes and by reducing tensile residual stresses at tube surfaces. In these circumstances DHC propagation giving rise to leak-before-break is promoted.

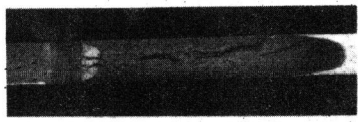
Further, DHC in pressure tube material can be prevented if the hydride habit planes are unfavourably orientated to a potential starter-notch tip.

#### ACKNOWLEDGEMENTS

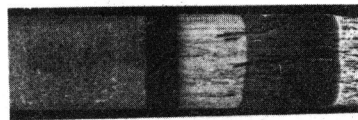
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(a)



(b)

Figure 1  
(a) DHC under plane stress crack front 50% square. 2mm thick specimen.  
(b) DHC under plane strain crack front 90% square. 4mm thick specimen.

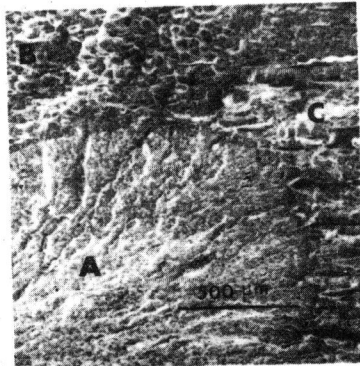


Figure 2  
Area A-DHC propagation,  
Area B-Shear lip denuded in hydride,  
Area C-Separation by overloading  
-note concentration of hydrides.

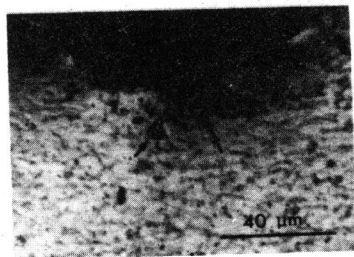


Figure 3  
Arrows show reorientation of hydrides in vicinity of notch tip