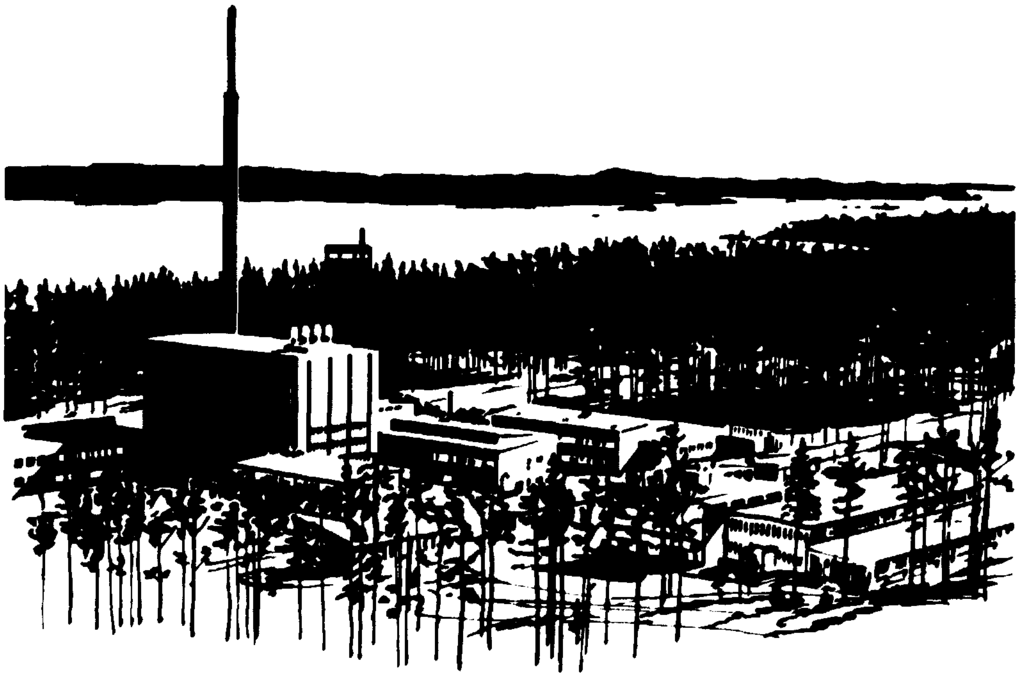
Studsvik Report

STUDSVIK/K4-78/12

STRESS CORROSION CRACK GROWTH IN UNIRRADIATED ZIRCALOY

Kjell Pettersson

SE7900158



STUDSVIK ENERGITEKNIK AB

STUDSVIK/K4-78/12

1978-10-20

1065091 Sprickutbredning AE

Kjell Pettersson

Nämnden för Energiproduktions-
forskning

STRESS CORROSION CRACK GROWTH IN UNIRRADIATED ZIRCALOY

ABSTRACT

Experimental techniques suitable for the determination of stress corrosion crack growth rates in irradiated Zircaloy tube have been developed. The techniques have been tested on unirradiated. Zircaloy and it was found that the results were in good agreemant with the results of other investigations. Some of the results were obtained at very low stress intensities and the crack growth rates observed, gave no indication of the existance of a K_{ISCC} for iodine induced stress corrosion cracking in Zircaloy. This is of importance both for fuel rod behavior after a power ramp and for long term storage of spent Zircaloy-clad fuel.

Approved by

Kjell Pettersson

LIST OF CONTENTS

1. INTRODUCTION

Ever since it was understood that iodine induced stress corrosion cracking (SCC) of Zircaloy could be the cause of pellet-clad-interaction (PCI) failures in water reactor fuel, such cracking has been the subject of numerous investigations. A recent review (1) shows that these investigatins have been concerned with the effects of different metallurgical factors such as cold work, texture, residual stresses, structure and irradiation. Other factors include temperature, iodine concentration and strain rate. The results from the many different test methods can usually not be compared with each other. It is particularly difficult to use the results in mathematical models for fuel behaviour. What is lacking is a failure criterion, which can predict when the fuel pin fails by the SCC mechanism during the operation of the reactor.

Criteria have been proposed such that when a critical stress, or a critical plastic strain, or both are exceeded the tube will fail due to the SCC mechanism. However, it is in all these cases possible to find laboratory test results which are at variance with the proposed criterion or criteria. Therefore it is likely that for the treatment of SCC in fuel rod modelling a detailed understanding is needed of the cracking process in Zircaloy under the influence of stress and fission products.

A simple model of the cracking process would be to divide it into three stages; crack initiation, crack growth and final failure. The crack initiation and growth are caused by the combined effects of stress and environment, whilst the final failure only requires the stress. The problem is to treat the different stages quantitatively. There are currently no theories available for the treatment of the initiation stage. For the growth stage it can be assumed that the crack growth rate is determined by the stress state at the crack tip (under constant environmental conditions at the crack tip). This approach has been successful in studies of SCC in several materials (2). Three recent studies of crack growth in unirradiated Zircaloy have shown that the approach is valid also for iodine induced SCC crack growth in Zircaloy (3 - 5). It has also been shown that it is possible to analyse stress-rupture data of Zircaloy tube pressurized with an argon-iodine mixture, on the basis of a stress intensity dependent crack growth rate (6).

Observations that the SCC behaviour of irradiated Zircaloy is sometimes very different from that of unirradiated Zircaloy (7) also indicate that the crack growth stage will be different in irradiated material. It is therefore necessary to measure crack growth rates in irradiated Zircaloy in order to obtain relevant crack growth data for fuel rod modelling.

The present report covers the first phase of a project aimed at the determination of SCC crack growth rates in irradiated Zircaloy as a function of the stress state at the crack tip.

The first phase includes methods of pre-cracking specimens by fatigue, and measurements of SCC crack growth rates on an irradiated material.

2. GENERAL DESCRIPTION OF METHOD

A method of measuring SCC crack growth rates in Zircaloy tube has been developed by Videm and Lunde (4).

In order to eliminate the crack initiation stage from the tests, the SCC crack growth is started from fatigue cracks introduced before the testing in iodine. The testing under a constant internal pressure of an argon-iodine mixture is carried out for a certain time before stopping the test. After the test the fracture surfaces of the fatigue-SCC cracks are examined in a scanning electron microscope. It is relatively simple to distinguish between the fatigue and SCC parts of the cracks, and to measure the extent of crack growth which has taken place during the SCC-test. The average crack growth rate can be related to the stress intensity factor of the crack, evaluated as (6)

$$K_I = 2.5 \, \sigma \sqrt{a} \qquad \text{(Eq 1)}$$

where σ is the hoop stress and a the crack depth. This formula is valid if the crack depth is about 0.1 of the length of the crack, and less than about 0.4 of the wall thickness of the tube. The validity of the formula is also limited to the case when the plastic zone is smaller than the remaining wall thickness under the crack tip. The size of the plastic zone is given by (8).

$$r_p = \frac{1}{6\pi} \left(\frac{K_I}{\sigma_s}\right)^2 \qquad \text{(Eq 2)}$$

With r_p equal to half the ligament or about 0.2 mm and σ_s = 180 MPa at 300°C, K_I = 11 MPa $\sqrt{m}$.

With a crack depth of 0.3 mm this value of K_I corresponds to a stress of 250 MPa. This means that general yielding takes place before the plastic zone becomes too large.

3. METHODS OF FATIGUE CRACKING

3.1 Notches

The best way of controlling the initiation of a fatigue crack is to let it start at a notch. The notch acts as a stress raiser, and the fatigue crack initiation can occur easily at a relatively low stress level. The original intention was to use notches made by electric discharge machining. However, for some unknown reason, these notches were not effective as crack initiators.

Of the different machined notches tried, a notch made by a scalpel blade turned out to be the best. With the scalpel blade mounted in a special rig it was possible to make notches of about 0.1 mm depth and 3 mm length reproducibly. It is also possible to design a rig for making such notches, which could be handled with manipulators in a hot cell.

3.2 Fatigue cracking by internal pressure variations

The first method of producing fatigue cracks was by subjecting the notched tubular specimens to a varying internal pressure at room temperature. The frequency was limited to 3 Hz. In order to get sufficiently high crack propagation rates the pressure was varied between 60 and 260 MPa. With a crack depth of 0.1 mm this gives a stress intensity range of 4 MPa $\sqrt{m}$ (6). Published crack growth data (2) gives a fatigue crack growth rate of about $2\cdot10^{-6}$ mm/cycle at 4 MPa $\sqrt{m}$. In order to get a 0.1 mm deep fatigue crack it is therefore necessary to cycle the specimen for 50 000 cycles, taking 4.5 h.

The practical tests showed that about 0.2 % plastic deformation was obtained in the specimen due to the fatigue cycling. This plastic deformation is undesirable because it may cause strain hardening which can affect the SCC properties of the material. It was judged to be impractical to reduce the stress range for fatigue cycling, since this would result in excessively long fatigue cycling times, according to the published fatigue crack growth data.

Another disadvantage with using internal pressure variations for fatigue cracking is that, since the specimen is already closed before the fatigue phase, it is difficult to load the iodine into the specimen for SCC testing.

Apart from the above mentioned difficulties, it was possible to obtain reproducible fatigue cracks by varying the internal pressure, when the notches were reproducible.

To summarize: it was judged that the disadvantages of fatigue cracking with internal pressure were too great. Other methods had to be sought.

3.3 Fatigue cracking by point loading

After some experimenting with the use of bending stresses for propagation of fatigue cracks, a rig for point loading on the specimen was constructed, as shown in Figure 1. The specimen holder and loading device can be mounted in an Instron 1271 dynamic testing machine. The machine can be run under load control so that the load on the specimen is varied between constant maximum and minimum values.

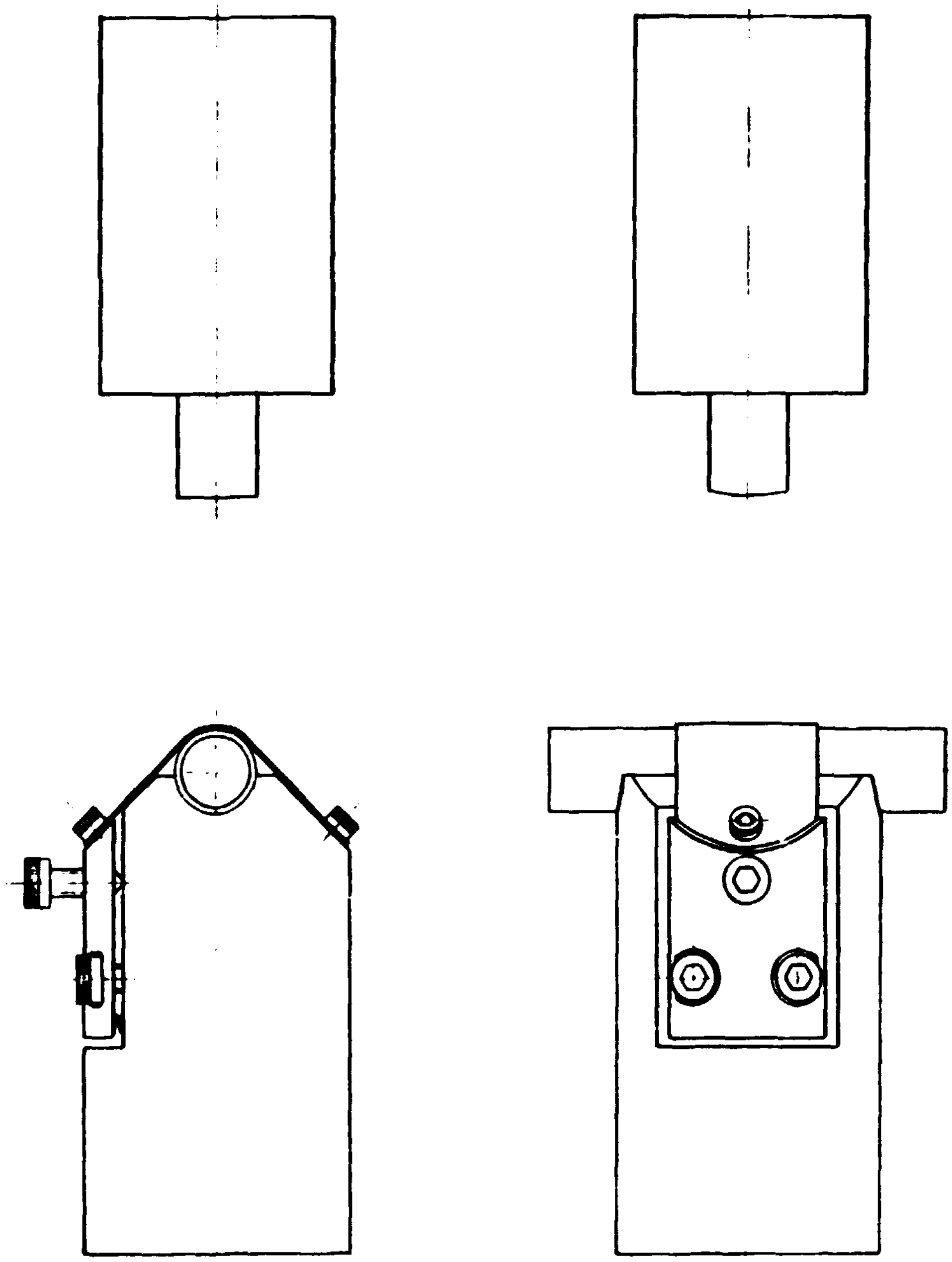


Figure 1.

Rig for point loading on tubular specimen used for fatigue cracking

Mainly by trial and error it was possible to find a suitable load amplitude at 100 Hz which gave reproducible fatigue cracks without any permanent deformation of the specimen. A typical loading time was 30 min, corresponding to 180 000 cycles, or a crack growth rate of about 10^{-6} mm/cycle, which corresponds to a stress intensity range of about 3 MPa $\sqrt{m}$. With a specimen length of 75 mm it is practical to have 3 cracks on each specimen.

Another advantage with the point loading device is that it will be relatively easy to design a radiation shield around it, so that irradiated specimens can be fatigue-cracked.

4. MEASUREMENTS OF CRACK GROWTH

4.1 SCC-testing

Before testing, leak-tight end fittings were mounted on the 75 mm long specimens. An internal mandrel in the specimen serves as support to the end fittings and keeps the specimen straight, and the length of the specimen constant during testing. One of the end fittings is connected to a tube which ends in a valve. The mounting of end fittings with tube and valve and the loading of iodine into the specimen is done in a glove box in an inert atmosphere. The valve is closed and not opened until it has been connected to the pressurizing system. The amount of iodine was 0.6 mg/cm^3 of gas volume in the specimen and tube connection.

The SCC testing was carried out at 300°C. The specimen was pressurized with purified helium. Four specimens with three fatigue cracks in each were tested. The pressures were chosen so that it would be possible to detect the change-over from intergranular to transgranular crack propagation according to the data obtained by Videm and Lunde (4). For two of the specimens the desired stress was 212 MPa and for the two others the desired stress was 142 MPa. Only one of the specimens was completely free of leaks. For two of the specimens, those with the lower stress, the leak rates were so low that they could be refilled with gas to the desired pressure after regular time intervals, whilst the leak rate in the fourth specimen was so high that it was impossible to maintain any pressure in the specimen.

The specimen with the highest stress failed in one of the pre-cracks after 1.6 h whilst the two specimens at the lower stess were tested for 167 h without failure.

4.2 Evaluation of crack growth

After testing, all the pre-cracks were examined in a scanning electron microscope (SEM). The examination was carried out on the fracture surfaces obtained by breaking up the specimens. In a few cases the specimens were sectioned through a defect so that a part of the crack was retained for metallography.

It was easy to distinguish between the parts of the fracture surface obtained during the different crack propagation stages: fatigue, SCC and final ductile failure during breaking up of the specimen. In all nine defects SCC growth could be seen. In the boundary between the fatigue fracture surface and the SCC surface there was a narrow zone where some evidence of ductile crack propagation was detected.

In the through-the-wall defect of the failed specimen the ductile ligament was only 0.06 mm, which is considerably less than would be expected from a limit-load-analysis. The limit load is given by (9)

$$\sigma = \sigma_y^1 \frac{(t - a)}{t} \qquad \text{(Eq 3)}$$

where σ_y^1 is the yield stress in the closed end burst test, t the wall thickness and a the crack depth. Part of the explanation to the stability against ductile failure can be that strain hardening stabilizes the material in the ligament, however, with σ = 212 MPa and t = 0.8 mm and t-a = 0.06 mm, σ_y^1 = 2 800 MPa which seems a little high.

The result of the SEM examination is summarized in the following table together with calculated K-values and the crack growth rates. For the specimens with varying stress, an effective average stress was determined from the recorded stress-time histories, with the assumption that the crack growth follows a law of the type

$$\frac{da}{dt} = C\, K_I^n \qquad \text{(Eq 4)}$$

with n = 10.

The crack growth rates are the average rates with no corrections for the variation of K during crack extension. K was evaluated at the start of the SCC crack growth.

The results have also been plotted in Figure 2 together with the best fit line to the results obtained by Videm and Lunde at 340°C (4). This line follows the equation

$$\frac{da}{dt} = 7.5 \cdot 10^{-14}\, K_I^{9.7} \quad (\text{mm/s},\ [K_I] = \text{MPa}\sqrt{\text{m}})$$

and the present data have been fitted to

$$\frac{da}{dt} = 7.3 \cdot 10^{-13}\, K_I^{7.2} \quad (\text{mm/s},\ [K_I] = \text{MPa}\sqrt{\text{m}})$$

Table 1.

Test temperature 300^{o}C. The wall thickness of the specimens was 0.79 mm.

Specimen/Defect	Notch mm	Fatigue crack mm	SCC crack mm	σ MPa	$K_I = 2.5\ \sigma\sqrt{a}$ MPa$\sqrt{m}$	$\frac{da}{dt}$ mms^{-1}	SCC crack type
A/1	0.12	0.26	0.07	212	10.4	1.2×10^{-5}	transgranular
A/2	0.09	0.26	0.09	212	10	1.56×10^{-5}	"-
A/3	0.15	0.28	0.30	212	11	5.2×10^{-5}	"-
C/1	0.135	0.226	0.203	118	5.6	3.4×10^{-5}	mixed inter
C/2	0.132	0.094	0.038	118	4.4	6.3×10^{-8}	and trans-
C/3	0.117	0.192	0.128	118	5.2	2.1×10^{-7}	granular
D/1	0.128	0.214	0.085	137	6.3	1.4×10^{-7}	
D/2	0.099	0.247	0.115	137	6.4	1.9×10^{-7}	"-
D/3	0.085	0.263	0.094	137	6.4	1.6×10^{-7}	

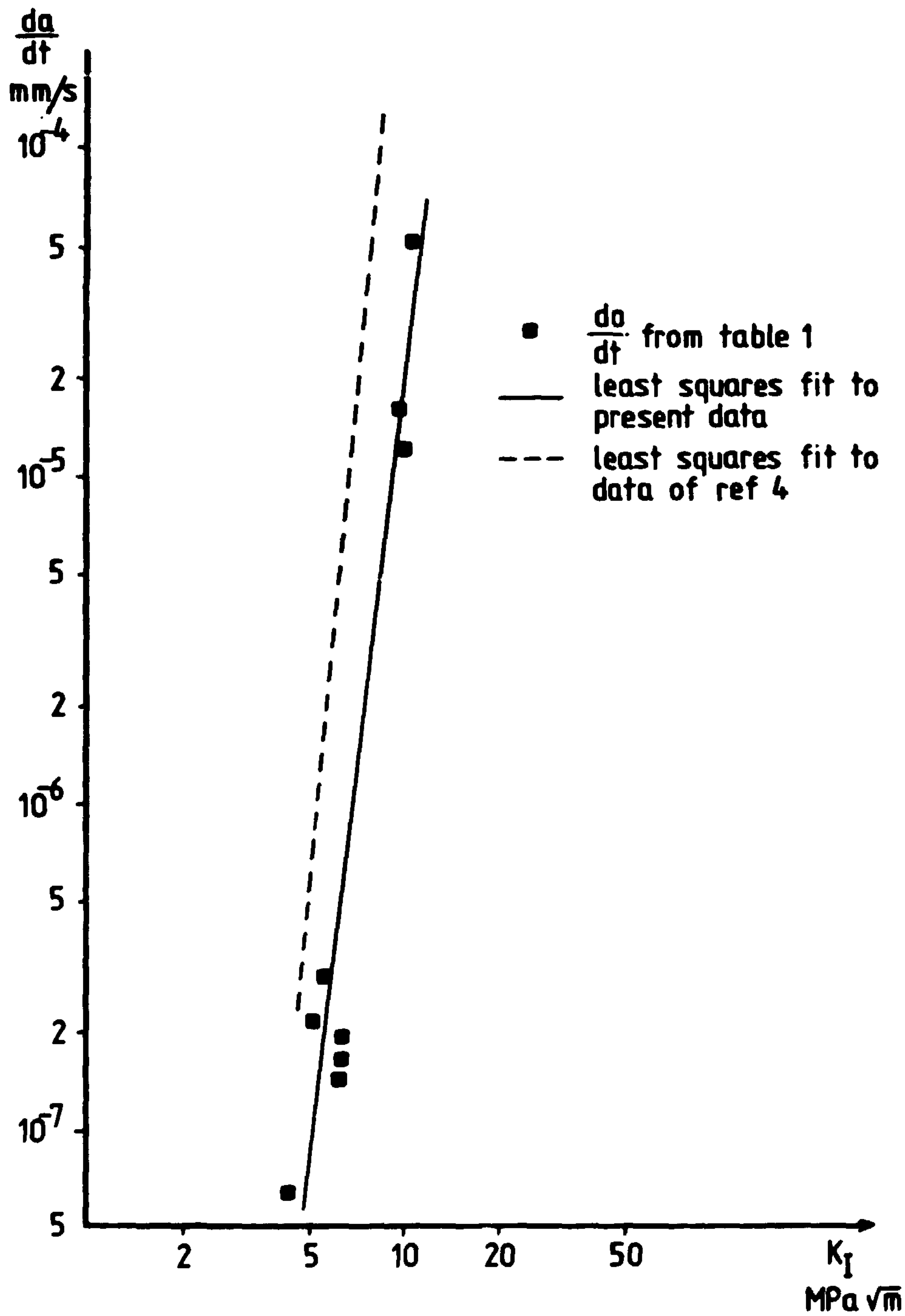


Figure 2.

Crack growth rate as a function of stress intensity factor K_I

5. DISCUSSION

The present data can be compared to those obtained by Videm and Lunde (4), the only difference in the experiments being the test temperature and the amount of iodine. Videm and Lunde carried out their tests at 340°C with an iodine concentration of 3 mg/cm^3. The effect of the temperature difference can be estimated from an activation energy determination by Kreyns et al (6) which gives the constant C in the crack growth law:

$$\frac{da}{dt} = C\ K_I^n \qquad \text{(Eq 5)}$$

an Arrhenius-type temperature dependence with

$$C = C_o \exp - \frac{Q}{RT} \qquad \text{(Eq 6)}$$

where Q was determined as 42.9 kcal/mol for recrystallized Zircaloy. This value of Q gives a rate increase with a factor of 11 from 300 to 340°C. Correcting the present data by a factor of 11 would result in the two best fit lines crossing each other.

It is not expected that the different iodine concentrations would give significantly different crack growth rates, since they are both above the limit ((1 - 5) $\cdot 10^{-4}$ g/ml) where iodine readily gives SCC of Zircaloy in short term tests (1). The agreement between the two investigations must, therefore, be regarded as extremely good.

One interesting difference between the two investigations is the mode of cracking. At the highest crack growth rates in the present investigation the cracking was transgranular, whilst Videm and Lunde observed mixed inter- and transgranular crack growth at these rates. For the lower crack growth rates Videm and Lunde observed purely intergranular crack growth, whilst in the present investigation the cracking was mixed inter- and transgranular. This difference could perhaps be a consequence of the higher iodine concentrations in the tests of Videm and Lunde, which may help grain boundary dissolution. It would on the other hand be expected that a difference in the cracking mechanism would be accompanied by a significant difference in stress dependence, and this was not observed. Clearly much more work is required for an understanding of all details of iodine induced SCC in Zircaloy.

The point at $6.3 \cdot 10^{-8}$ mm/s is the lowest crack growth rate measured so far, and it is interesting to note there is no sign that a K_{ISCC} for SCC exists. This has implications both for normal operation of Zircaloy clad fuel and long term storage of spent fuel.

If, as is suspected, crack growth rates are significantly higher in irradiated Zircaloy, it is conceivable that SCC cracks may grow for a long time following a power ramp, driven only by the low stress remaining after relaxation of the stresses in the cladding and fuel. For storage of spent fuel, where storage 40 years or more is envisaged, it must be ascertained that SCC crack growth at low stress intensities cannot endanger the cladding integrity. It is true that the activation energy of 42.9 kcal/mol predicts growth rates $5.4 \cdot 10^{-12}$ lower at $80^{o}C$ than at $340^{o}C$ but recent data on SCC crack growth in unirradiated Zircaloy shows that the growth rates are only about 10^{-5} lower (10). Therefore there is a clear incentive also to study crack growth at low temperature on irradiated Zircaloy, particularly Zircaloy with hydrogen contents typical of high burn-up fuel rods, since there is a risk of an interaction between delayed hydride cracking and SCC.

6. ACKNOWLEDGEMENTS

The author wishes to acknowledge the experimental assistance of H Bergqvist and Å Nylund. Special thanks are due to Y Haag for the design of the rig for fatigue cracking of tube.

REFERENCES

1. COX, B and WOOD, J C
Iodine induced cracking of Zircaloy fuel caldding - a review.
Electrochemical Society Meeting, New York, Oct 15 - 16, 1974.

2. SPEIDEL, M O and STAEHLE R W eds.
ARPA-Handbook on Stress Corrosion Cracking.
To be published.

3. WOOD, J C
Factors affecting stress corrosion cracking of Zircaloy in iodine vapour.
J. Nuclear Material 45 (1972/73) p 105 - 122.

4. VIDEM, K and LUNDE, L
Stress corrosion crack initiation and growth in Zircaloy.
Enlarged Halden Programme Group Meeting, Sanderstölen, Norway, 1977.

5. COX, B and WOOD, J C
The mechanism of SCC of Zirconium alloys in halogens.
Proc. Int Conf on Mechanisms of Environment sensitive cracking of Materials, Guildford, UK, 1977.
Eds. P R Swann et al, The Metals Society, London 1977. p 520 - 530.

6. KREYNS, P H, SPAHR, G L and McCAULEY, J E
An analysis of iodine stress corrosion cracking of Zircaloy-4 tubing.
J. Nuclear Materials 61 (1976) p 203 - 212.

7. LEE, D and ADAMSON, R B
Modeling of localized deformation in neutron irradiated Zircaloy.
ASTM-STM-633, p 385 - 401, ASTM 1977.

8. CARLSSON, J
Brottmckanik.
Ingenjörsförlaget, Stockholm 1976.

9. PETTERSON, K, VESTERLUND, G and ANDERSSON, T
The effect to irradiation on the strength, ductility and defect sensitivity of fully recrystallized Zircaloy tube.
Submitted for publication.

10. VIDEM, K and LUNDE, L
Stress corrosion crack initiation and growth and the formation of pellet-clad interaction defects.
Submitted for publication.

STUDSVIK/K4–78/12 . STRESS CORROSION CRACK GROWTH IN UNIRRADIATED ZIRCALOY . Kjell Pettersson

Studsvik

Studsvik Energiteknik AB S-611 82 NYKÖPING
SWEDEN Telefon 0155-800 00 Telex 64013 studs s