INFLUENCE OF IRRADIATION ON IODINE-INDUCED STRESS CORROSION CRACKING BEHAVIOUR OF ZIRCALOY 4

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1. INTRODUCTION

The first fractures by Pellet-Cladding Interaction (PCI) were observed during the 60s in BWRs or CANDU-type reactors, and in the mid-70s in PWRs [1]. The fracture mechanism was quickly identified as being iodine-induced Stress Corrosion Cracking (SCC) [2]. This conclusion was reached because PCI fractures appear in the absence of any general cladding deformation, they occur for relatively high burn-ups during or after the power ramp, and the fracture micrographs show the same aspect as that obtained in the laboratory with iodine-induced Stress Corrosion Cracking [3].

As a result, the CEA and Framatome decided to join forces to develop I-SCC laboratory tests. Ten years of study has given us a global vision of phenomena which lead a cladding submitted to SCC conditions to rupture. This vision allows us to propose a several-stages description of the clad life, expressed according to the data which rule it, versus the parameters which influence it [4]. This description organizes all the available and the future data. A PCI thermomechanical model which uses our description is expounded elsewhere [5].

This article purpose is rather to expose recent experimental results. To that effect, the main experimental device used for internal pressurisation is presented, and also the recorded data. The SEM fracture micrograph is used as a basis for describing the fundamental phenomena and to express them in terms of behavioural laws. The results required to apply these laws will be described for a stress-relieved Zircaloy cladding, both non-irradiated and irradiated for one PWR cycle.

The variation in parameter values as a function of irradiation dose and of other metallurgical parameters will be described. The discussion will close with a description of the behavioural similarities between new material and irradiated material.

2. EXPERIMENTAL DEVICE

The experimental device is designed to exert internal pressure at 350°C on a 135mm length of cladding tube [6]. A diagram of this device is given in figure 1. The pressure applied varies from that leading to immediate rupture down to that not causing rupture after a test period of 100 hours. The cladding dimensions and assembly are such that the circumferential stress is proportional to the longitudinal stress (σc/σl = 2) and that the radial stress σr is negligible compared to σc and σl. The results will therefore be expressed as a function of the circumferential stress.
The reference tests are carried out by pressurising the tubes with argon alone. For the tests in an iodine atmosphere, approximately 60 mg of iodine is introduced in the tube before pressurisation, giving a surface concentration of 1.5 mg.cm⁻².

The pressure is applied to samples of sound tubes and tubes pre-cracked by a fatigue process. In the first case, the sample lifetime includes crack initiation and growth stages. In the second case, only crack growth takes place. After the tests, the crack growth path during the test piece lifetime is measured and compared to the initial pre-crack size. This gives the crack growth speed and a fracture mechanics calculation gives the initial value of $K_T$, the stress intensity factor.

The main advantages of this technic are the following:

- the induced defect is exactly a crack; fracture mechanics laws can be applied, and complete $(da/dt, K)$ curve can be designed, which allows an accurate graphical determination of $K_{th}$,
- precracked tubes can be irradiated in research reactor, and recorded data can be transposed to tubes irradiated in power reactor with the same fast neutrons dose. Indeed, fast neutrons are the ones which induce microstructural defects in the material. Moreover, fission products, which modify in power reactor the clad internal surface, do not influence the crack propagation. This technic allows therefore to determine propagation threshold on irradiated materials, while post-irradiation defect machining as crack simulation is difficult and has been given up in a lot of laboratories [7].

The parameters chosen to characterise the cladding behaviour are therefore:

- the lifetime $t_p$ as a function of stress $\sigma_c$ for sound tubes tested in an inert atmosphere and in an iodine atmosphere,
- the crack growth rate $da/dt$ of the SCC mechanism as a function of the stress intensity factor $K$, measured on precracked tubes,
- the stress intensity factor threshold $K_{th}$ enabling these cracks to grow.

3. FUNDAMENTAL SCC FRACTURE MECHANISMS

Post-test observations of SEM fracture micrographs, of samples of non-irradiated cladding and cladding irradiated for one power reactor cycle, allow to identify the fundamental fracture mechanisms. The study of these mechanisms leads to a fine description of the several-stages life of a clad submitted to SCC conditions, and results in a better interpretation of the macroscopic data measured during the tests [5].

The fracture facies of the samples ruptured under SCC (figure 2) show:

- an intergranular corrosion zone surrounding the crack initiation site,
- a sudden transition to a transgranular brittle fracture zone.

When plastic instability is reached, a cupular facies characteristic of ductile fracture is observed.

The size of the intergranular zone $a_{ig}$ depends on the applied stress and can be expressed by the relation $a_{ig} \sqrt{\pi a} = K_{th}$ where $K_{th}$ is a constant characteristic of the SCC behaviour of the material. It is defined as the stress intensity factor required for SCC growth. $a$ is a shape factor generally close to 1 [8].

The crack initiation and intergranular corrosion stage is slow, being little affected by the stress level, and may represent up to 80% of the cladding lifetime.

Brittle transgranular growth takes place by pseudo-cleavage on the basal plane [9]. This is a fast-moving stage where the crack growth rate is directly dependent on the stress intensity factor at the crack tip.

Experiments and observations made by the CEA have shown that these mechanisms were identical in a new material and a material irradiated with doses of up to $3.7\times10^{21}$ n.cm⁻² (figure 3).

By identifying these mechanisms, it is possible to propose a single model for new and irradiated material. This modelisation integrates coherently physical failure mechanisms.
description and the whole experimental data. The cladding lifetime under SCC is then as follows [4]:

\[ t_f(\sigma) = t_i() + t_{grth}(\sigma) = t_i + \int_{a_i}^{a_r} \frac{da}{da/dt(K)} \]

where:
- \( t_f(\sigma) \) is the time to rupture, as a function of applied stress, measured experimentally by internal pressurisation of sound tube samples,
- \( t_i() \) is the duration of the crack initiation stage,
- \( t_{grth}(\sigma) \) is the duration of the crack growth stage,
- \( a_i \) is the size of the intergranular zone or the size that the crack must attain for brittle growth to become possible, i.e., so that \( K_{th} \) is reached,
- \( a_r \) is the crack depth on rupture, given by the thickness of the ductile ligament,
- \( da/dt \) is the transgranular brittle growth rate, determined experimentally by internal pressurisation of pre-cracked tubes.

This relation could be used to calculate potential damage to cladding subjected to SCC conditions, as described elsewhere [10].

4. IRRADIATION EFFECTS

The results were obtained on stress-relieved Zircaloy cladding. The lifetime of the irradiated material was measured on samples taken from power reactor fuel rod cladding. Their total fluence is 3.7x10^{21}n.cm^{-2}. Crack growth rates in the irradiated material were measured on cladding sections pre-cracked by a fatigue process, then irradiated in an experimental reactor with a fluence of 1.3x10^{21}n.cm^{-2}.

The sample lifetime curves and the crack growth rates are given in figure 4.

Graphs showing sample lifetime in an inert environment for new and irradiated materials show the extent of irradiation-induced hardening. While the slope of the \((\sigma, \log(t_f))\) straight line is approximately the same in both cases, the stress leading to rupture after one hour is 20% higher in the case of an irradiated sample compared to a non-irradiated sample.

The lifetime of irradiated and non-irradiated samples in an iodine atmosphere is approximately the same for stresses between 400 and 600 MPa. Although the lack of data on irradiated material lifetime means that it is not possible to determine the non-rupture limit in an iodine environment as accurately as that for the non-irradiated material, it may nonetheless be assumed that these limit stresses will not be significantly different: 280 MPa for new material and about 300 MPa for irradiated material. Other work [11] instances a non-rupture limit at 200 MPa for an irradiated stress relieved material, but the non-irradiated material behaviour has not been studied. A non-irradiated close material exhibits a non-rupture limit at 300 MPa.

When the behaviour in an iodine environment is compared to that in an inert environment it is therefore clear that the irradiated material is more sensitive to SCC than the non-irradiated material.

The stress intensity factor threshold, \( K_{th} \), required to trigger brittle crack growth is considerably modified by irradiation with a fluence of 1.3x10^{21}n.cm^{-2}: it is divided by 2.6 with respect to the new material, dropping from 3.2 to 1.5 MPa.m^{1/2}.

It is difficult to compare our \( K_{th} \) values to some from other authors, first because there is not a lot of published ones relative to irradiated materials, then because all the metallurgical parameters, for instance texture, are not always specified. Yet texture has a great influence on the threshold values [12] as detailed forward. However, it could be refered to YAGGEE data [7]. He has studied failure of post-irradiation notched tubes submitted to increasing stress levels. It arises from tests run on several irradiated materials \( K_{th} \) values between 2.2 and 3.7 MPa.m^{1/2}, while
values between 3.9 and 6 MPa m\(^{1/2}\) are measured on non-irradiated materials. Although non-irradiated materials are not the same than irradiated ones, these measurements show clearly a drastic decrease of \(K_{th}\) with irradiation, of a near 2 factor.

By integrating the \(da/dt = f(K)\) relationship over the entire crack growth stage, it is possible to calculate \(t_{grth}\) and, by subtraction from the total lifetime \(t_p\), to determine the initiation time \(t_i\). This calculation shows that the initiation part of the lifetime is shorter for the irradiated material than for the non-irradiated material. This difference is not due to a high sensitivity to crack initiation and intergranular corrosion, but is justified purely by the reduction in \(K_{th}\) and the crack growth rate after the threshold.

5. EFFECT OF VARIOUS METALLURGICAL PARAMETERS

The effect of other metallurgical parameters was explored in the laboratory.

Comparative effect of microstructure and texture

Specific experiments performed at the CEA have shown that the main factor controlling the crack growth stage in SCC is material texture [13]. Varying the chemical composition within the ASTM specification limits for Zircaloy 4 has very little effect on this stage. The same can be said for metallurgical state, mechanical properties or microstructure. This may be explained by the fact that the growth threshold \(K_{th}\), which marks the initiation stage, is highly dependent on texture [12-13].

On the other hand, microstructure can be expected to have a major effect on the initiation stage [14] which, as previously mentioned, may represent up to 80% of the total lifetime of the material.

The effect of texture on crack growth in irradiated materials subject to SCC is currently being studied. The change in microstructure as a result of irradiation may also lead to changes in SCC resistance properties.

Effect of neutron dose received

The exposed results and some other preliminary tests would seem to indicate that resistance to SCC changes with the irradiation dose. There are several possible reasons for this evolution [13]:

- change in the microstructure of the material under irradiation (amorphisation and/or resolution of precipitates, segregation of alloying elements) [15],
- change in deformation possibilities by the introduction of a large number of point defects during irradiation (occurrence of \(<c>\) loops [16] in addition to the hardening observed after one irradiation cycle),
- internal surface damage to the cladding by implantation of recoil fission products [17].

Specific experiments are currently in progress to determine the importance of each of these factors on the SCC behaviour.

6. CONCLUSIONS

Internal pressurisation tests at 350°C were performed to study the SCC behaviour of a stress-relieved Zry 4 non-irradiated cladding and the same cladding irradiated for one cycle.

Fracture micrographs have shown that the same mechanisms govern the various stages in the lifetime of an irradiated and non-irradiated cladding under iodine-induced SCC conditions.

For applied stresses between 400 and 600 MPa, the lifetime of the cladding irradiated up to 3.7x10\(^{21}\)n.cm\(^{-2}\) is approximately the same for both inert and iodine atmospheres. However, the irradiation-induced hardening which increases the strength of the material in an inert atmosphere means that the irradiated material is more sensitive to SCC than the non-irradiated material. This
gives rise to a considerable reduction in stress intensity factor threshold, $K_{th}$, required for crack growth under SCC conditions.

By understanding the SCC rupture mechanisms through laboratory tests and post-test fracture micrograph examination, it was possible to propose a model for cladding lifetime.

REFERENCES AND FIGURES


Figure 2: SCC fracture surface. Sharp transition from intergranular corrosion to transgranular propagation.

Figure 3: SCC fracture surface of unirradiated sample (a) and of irradiated sample (b).

Figure 4: Life time (a) and crack growth rate (b) of unirradiated and irradiated stress relieved Zircaloy 4 cladding.