

INFLUENCE OF THE THERMAL TREATMENT ON THE HARDNESS OF HIGHLY IRRADIATED CLADDING SAMPLES

E. H. Toscano and W. Goll***

* European Commission, Joint Research Centre, Institute for Transuranium Elements, PO Box 2340, 76125 Karlsruhe, Germany

** Framatome ANP GmbH, PO Box 3220, 91050 Erlangen, Germany

ABSTRACT

The straightforward Vickers hardness test, which can be easily performed under remote handling conditions, was chosen to analyse the recovery behaviour of different cladding materials under various thermal conditions. Samples cut from irradiated fuel rods were defuelled by using a small lathe installed in the hot cell. A special sample holder, able to withstand temperatures up to 600°C, was developed, allowing the annealing of the samples together with the sample holder. This facilitates the execution of the subsequent hardness test. A small furnace, electrically heated, was used to carry out the heat treatment under controlled inert gas atmosphere. In the present paper the experimental details (sample holder, furnace and sample preparation) are described and preliminary results presented.

INTRODUCTION

During irradiation in reactors, cladding materials are subjected to an intense fast neutron flux causing hardening due to the introduction of damage zones in the lattice. After irradiation, during fuel assembly transportation or interim dry storage, high temperatures may arise leading to recovery of the mechanical properties of the cladding materials. The amount of recovery is mainly dependent upon temperature but may also be affected by the irradiation history and the material properties.

Short time and long-term creep and rupture tests have been performed to assess the strain potential of cladding of high burnt fuel rods under conditions of dry storage^[1]. The tests were conducted at temperatures expected during intermediate storage (around 400 °C). At these temperatures and assuming that the creep kinetics is dominated by dislocations glide, the analysis of the internal stresses shows that the steady strain rate ($d\dot{\epsilon}/dt$) is determined by the ratio between two contributions: the static recovery, r_s , at the test temperature and the strain hardening, h , caused by the creep deformation^[2]:

$$d\dot{\epsilon}/dt = r_s/h$$

Whereas during creep both parameters play a role, the degree of recovery and recrystallization, r_s , can be assessed by means of static micro-hardness testing. This technique is widely used to characterise mechanical properties of materials and the most widespread method is the Vickers indentation.

Hence, in the framework of the program established to assess materials for interim storage, an assessment of the recovery and recrystallization properties has now been considered of principal importance for the better understanding of creep properties. In the present paper the technical scheme for the thermal treatment and the infrastructure for the hardness measurement is presented.

1. Annealing processes

Annealing phenomena may be relevant during fuel transportation and under dry storage conditions. Temperatures exceeding 400°C may be achieved during hours and days whereas 350°C may occur during months or years in interim dry cask storage. On the other hand, the irradiation in Light Water Reactors (LWRs) takes place at lower temperatures (around 300°C in BWRs and 320 up to 350°C in PWRs).

During in-service irradiation, defects have been found uniformly in the cladding material for fluences as low as 1×10^{24} n/m². They include point defects and dislocation loops. The softening of materials is caused by the dissolution of the point defects, rearrangement of dislocation loops and recrystallization processes. Large defects are relatively stable whereas small ones may be annealed at temperatures slightly higher than the irradiation ones.

For a given temperature, the evolution of the microstructure (F_R) versus time (t) may be described by the classical Johnson-Mehl-Avrami's law^[3]:

$$F_R = 1 - \exp [-(kt)^n]$$

Where the parameter k is correlated to the thermal activation energy of the process, Q , by the Arrhenius law:

$$k = k_0 \exp (-Q/RT)$$

The parameters involved can be evaluated by means of the micro-hardness technique.

2. Equipment

2.1. Sample holder

A sample holder to polish the samples and able to withstand the annealing temperatures (400 to 550°C) had to be developed to allow sample indentation after various annealing periods. In Fig. 1 the solution chosen is depicted. The material selected to build the sample holder was stainless steel AISI 304. The sample is supported internally and fixed by means of 3 floating screws, tightened in such a way as to avoid that any vibration during the polishing procedure can unfasten the sample.

2.2. Furnace

An electrical resistance furnace is used to perform the thermal treatments. The oven can achieve temperatures up to 1200°C. A commercial furnace was provided with two end caps, allowing the inlet and the outlet of the gas (Ar), which provide the inert atmosphere. The gas outlet takes place through a 40 mm water column, creating a slight overpressure inside the kiln, assuring that the surface quality remains constant after each heat treatment (oxidation free).

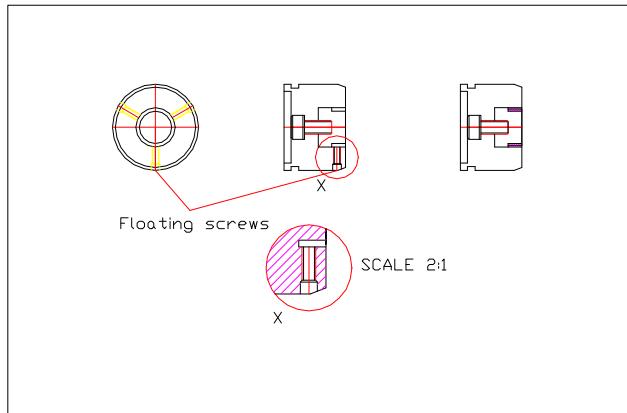


Fig. 1: Sample holder

2.3. Sample preparation

Rings having about 5 mm in length are cut from the irradiated fuel rods. Previously, most of the fuel is retrieved by means of a small lathe installed in the hot cell. Afterwards and in order to avoid any damage of the microscope through fuel particles, the rest of the fuel was dissolved in nitric acid. Subsequently, the de-fuelled samples were polished up to $\frac{1}{4}$ μm diamond paste.

2. Preliminary tests

In order to check the proper functioning of the furnace, in particular the influence of the atmosphere, measurements were performed on as-received Zircaloy 2 material.

The thermal treatments were performed under flowing Ar at 500 °C.

In Table 1 the Vickers' Hardness of the original material in the as-received form and after 4

Measurement	Original Material		After 4 h /500 °C		After 8 h /500 °C		Mean Value	Vickers Hardness
1								
2	92,76	93,27					93,02	
3	89,86	93,73					91,80	
							92,41	173,7
4			99,61	94,6			97,11	
5			95,65	99,13			97,39	
							97,25	156,9
6					97,95	94,43	96,19	
7					96,23	92,97	94,60	
8					95,59	93,09	94,34	
							95,04	164,2

Table 1: Vickers Hardness of Zircaloy-2 as received and after thermal treatments.

The applied load was 200 g and, in Fig.2, the appearance of the indentation marks is shown. In the second thermal treatment, an interruption of the gas flow caused a slightly oxidation of the sample surface. This explains the minor increase in the average hardness after 8 hours at 500°C. The surface appearance of the slightly oxidised material can be seen in Fig. 2 d.

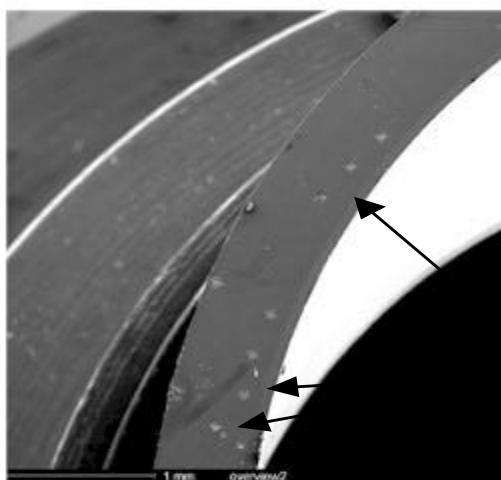


Fig.2 a:
Positioning of the Vickers' indentation marks
(arrows)

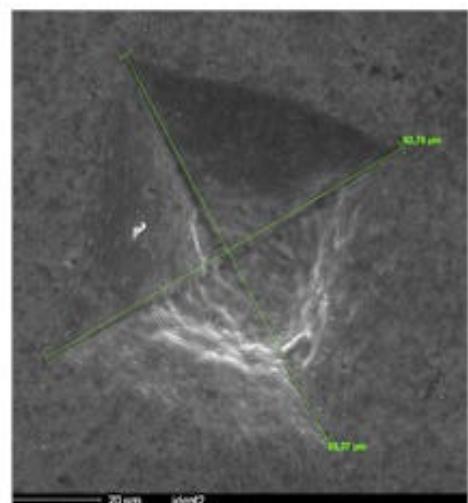


Fig.2 b:
Indentation mark of the as-received material

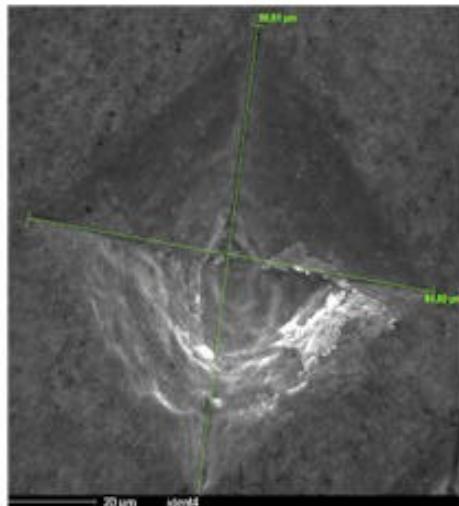


Fig.2 c:
Indentation mark on the material after 4 h at 500 °C

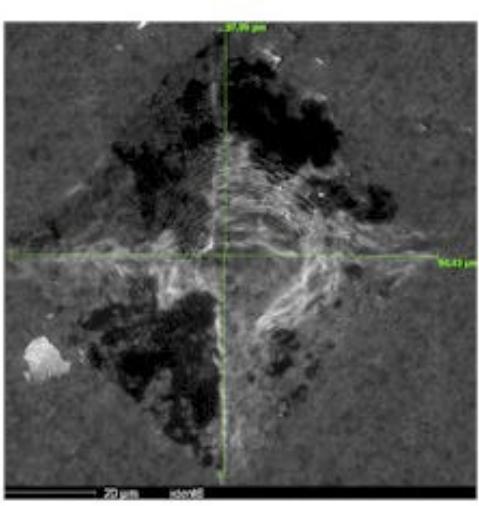


Fig.2 d:
Indentation mark on the material after 8 h

CONCLUSIONS

The infrastructure to prepare and to anneal irradiated samples for subsequent Vickers indentation tests has been developed.

Preliminary results obtained using non-irradiated material have demonstrated the sensitive role of the atmosphere during the thermal treatments.

References:

1. *W. Goll, H. P. Fuchs, and A. C. Leger, Int. Conference on Storage of Spent Fuel from Power Reactors, IAEA, Vienna, Austria, 2-6 June 2003*
2. *P. Bouffoux and L. Legras, ANS, Park City, Utah, April 10-13, 2000.*
3. *E. Steinberg et al., TopFuel 97, p.265.*
4. *T. Torimaru et al., J. Nucl. Mater. 238 (1996) 169*