Proceedings of the 15th International Symposium on the Packaging and Transportation of Radioactive Materials
PATRAM 2007
October 21-26, 2007, Miami, Florida, USA

MECHANICAL CHARACTERISTICS OF FUEL RODS IN TRANSPORT CONDITIONS

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ABSTRACT

A synthesis on the mechanical characteristics of unirradiated and irradiated fuel rod claddings was performed by IRSN in order to have reference data for the assessment of the safety demonstrations in normal and accident conditions of transport required by the procedure of package licensing. Indeed, the transport conditions correspond to a range of cladding temperatures (200°C to 550°C) which is only partly covered by the data acquired within the framework of the safety demonstration relative to the reactor normal operating conditions, especially beyond 400°C.

This work mainly concerned Zircaloy-4 cladding material (Zy-4). Data about mechanical properties (elastic and ductile properties, creep behaviour), oxidation (in reactor and under air during transport), hydrides, fracture toughness and fatigue behaviour have been collected and synthesized. The laws presented in the document can be used to obtain orders of magnitude of the oxide thickness, the hydrogen contents and the creep deformation rate.

The following phenomena which could influence the mechanical behaviour of the cladding were more particularly studied:

- oxidation which could become very important during transport in case of cladding temperatures of about 500°C,
- creep for which only a few data around 500°C are available and which depends in particular on the internal pressure of the rods, the cladding oxidation and the presence of the hydrides,
- recrystallization of Zy-4 at about 500°C, which could have consequences on the mechanical properties of the cladding after cooling during the storage.

For other topics of interest for the study of the mechanical behaviour of the cladding, such as the fracture toughness for example, it was identified that the data available is scarce.

This paper presents the main results obtained for Zy-4 and for M5™.

INTRODUCTION

Some hypotheses on the geometry of unirradiated and irradiated fuel assemblies or fuel rods transported in packages need to be considered in the containment and criticality analyses to demonstrate the safety of such transports. In the containment study, the activity release in normal and accident conditions of transport depends on the leaktightness of the fuel rods. In the criticality analysis, the geometry of the fissile material has a strong impact on the reactivity of the package. In this framework, it is useful to be able to predict the potential changes in the structure of the fuel assemblies in normal and accident conditions of transport, such as widening of the fissile section (birdcaging), slipping of the fuel rods, cracking and ruptures of the claddings.

Various cladding materials are used for LWR fuel rods, all of them are zirconium alloys, for example: Zy-4 used in PWR fuel assemblies, Zy-2 used in BWR fuel assemblies, M5™ developed by Areva for PWR use, Zirlo™ developed by Westinghouse also for PWR use. Some fuel claddings consist of two or three layers which can include pure zirconium. All these alloys have slightly different chemical compositions and thus may have different behaviours. Subsequent irradiation, heating and hydrogen pickup change the material properties.

The objective of the study performed by IRSN was to have reference data on the mechanical behaviour of fuel rod claddings for the assessment of the safety analyses provided to support the applications for package design approval. This work mainly concerned Zy-4 and M5™, which are the cladding materials mostly used in French PWR reactors. Data about mechanical properties (elastic and ductile properties, creep behaviour), oxidation, hydrogen pickup and fracture toughness, which could be necessary to study the mechanical behaviour of fuel rods in transport conditions are presented in this paper. Where experimental data are not available, it was necessary to make assumptions to define reference values.

METALLURGY OF ZY-4 AND M5™

Zy-4 and M5™ are two zirconium alloys used in nuclear industry. They are composed mainly of zirconium (more than 95 %) and contain also additives to improve their mechanical behaviour.

The chemical composition of Zy-4 is specified by ASTM standard B353. The main additives are iron (Fe) and chromium (Cr), which improve the resistance to corrosion, and tin (Sn), which improves the resistance to creep and the mechanical properties (increase of the yield strength and decrease of ultimate elongation). Some alloy grades of Zy-4 may contain a few ppm of oxygen (O) to improve the mechanical properties and slow down the creep rate, but in a controlled quantity since oxygen tends to reduce the resistance to corrosion.

M5™ is an advanced zirconium alloy developed by Areva. It is a 1% niobium zirconium alloy, containing also sulphur (S) and oxygen (O). Niobium (Nb) improves the resistance to corrosion, in a more effective way than tin for Zy-4, and it decreases creep rate for low stress level. Sulfur improves the creep behaviour without reducing the resistance to corrosion. In consequence, M5™ claddings oxidize less than Zy-4 claddings and creep more slowly for low stress level. However, for high stress level, M5™ claddings creep faster than Zy-4 claddings.

The Zy-4 used for fuel rod claddings could be in a cold worked stress relieved state or in a recrystallized state, whereas M5™ is always in a recrystallized state. Those two metallurgical states of the material are obtained by different thermal treatments applied after forming:

- for the cold worked stress relieved state (CW), the material is exposed at about 480°C during approximately 4 hours, the corresponding microstructure consists in stretched out grains,
- for the recrystallized state (RX), the material is exposed at about 550°C during approximately 4 hours, the corresponding microstructure consists in small equiaxed grains.

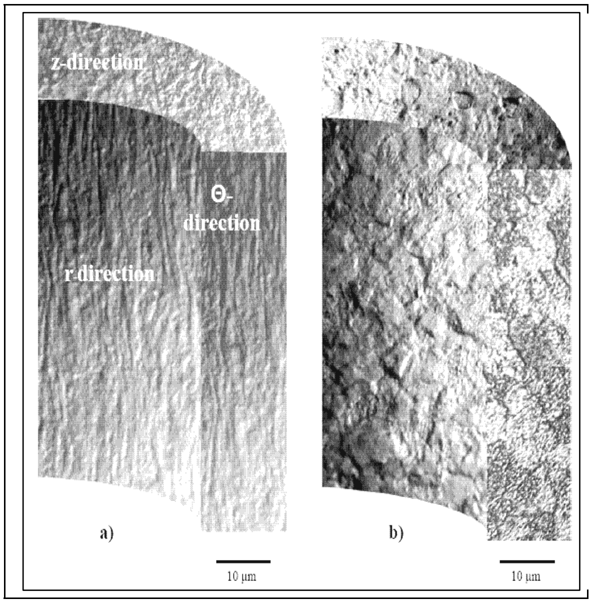

Figure 1. Metallographic aspects of CW Zy-4 (a) and RX Zy-4 (b) ([1]).

The main difference between those two states is the dislocation density, which is higher for the CW material than for the RX material. In consequence, the yield strength and ultimate tensile stress of the RX material are lower than that of the CW material.

MECHANICAL PROPERTIES OF ZY-4 AND M5^{TM}

The elastic and ductile properties of the cladding materials are entry data for the study of the mechanical behaviour of fuel rods.

Elastic properties

The elastic properties, Young modulus (E) and Poisson's ratio (ν), given hereafter apply to unirradiated and irradiated material as they are not significantly influenced by irradiation.
The Young modulus of Zy-4 is comprised between 97 GPa at 20°C and 62 GPa at 600°C. In this range of temperature, it could be estimated by the following relationship, issued from [2], E (MPa) = 114800 – 59.9 x T (K). The Young modulus of recrystallized zirconium alloy materials, such as M5^{TM}, is usually slightly lower than the one at the cold work stress-relieved state. For both materials, the Poisson's ratio is comprised between 0,3 and 0,4.

Ductile properties

The ductile properties, ultimate tensile stress (UTS), yield stress (YS) and rupture elongation (UE), unlike the elastic properties, are influenced by irradiation. Irradiation has a hardening effect on the material: UTS and YS increase while UE decreases. This effect can be considered saturated above a burn-up of about 5 GWd/tHM (burn-up reached during the first cycle of irradiation). Thus, in transport conditions, the cladding material can be considered either unirradiated or irradiated, without consideration of the burn-up or fluence.
A comparison of the ductile properties of Zy-4 and M5^{TM}, irradiated or not, made on the basis of literature data extracted from the PROMETRA program [3], is presented in table 1 hereafter. For the irradiated case, the data are issued from ring tensile tests made on Zy-4 samples irradiated up to 65 GWd/tHM with an oxide thickness less than 130 µm and on M5^{TM} samples irradiated up to 58 GWd/tHM with an oxide thickness less than 30 µm. Nevertheless, due to the saturation of the above mentioned effect of irradiation, the results should apply to higher burn-up values.

Table 1. Ductile properties of CW Zy-4 and M5^{TM}

<table>
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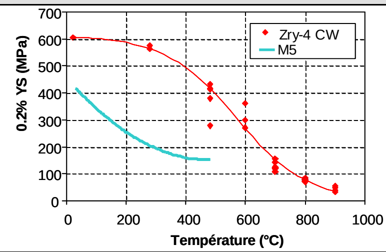
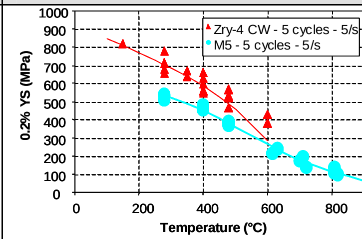
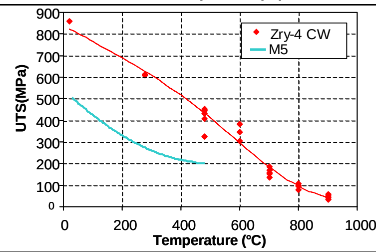
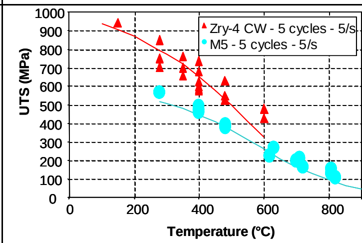
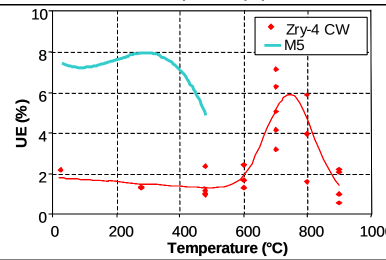
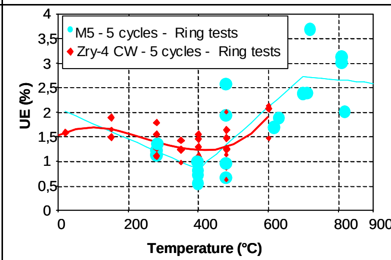
<tr><td>0.2% YS* (MPa)</td><td></td><td></td></tr>
<tr><td>UTS (MPa)</td><td></td><td></td></tr>
<tr><td>UE (%)</td><td></td><td></td></tr>
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* 0.2% YS is the engineering yield stress corresponding to 0.2 % plastic strain

CREEP IN NORMAL CONDITIONS OF TRANSPORT

A large set of data about creep behaviour of irradiated and unirradiated Zy-4 claddings is available, on the basis of which it is possible to elaborate evolution laws ([4], [5]). This is not the case for M5^{TM} claddings, for which only tendency could be found.

The results of creep tests available for Zy-4 show a high dispersion of the failure elongation.

For the unirradiated material, at temperatures lower than 500°C and hoop stresses lower than 150 MPa, which are typical of normal conditions of transport, RX Zy-4 tends to creep slower than CW Zy-4 and to reach a higher failure elongation ([6], [7]). The same tendency has been observed on unirradiated M5^{TM} claddings ([8]).

Up to 500°C, irradiation tends to reduce the creep rate and the failure elongation ([7]). It can be assumed that the rupture time is shorter for the irradiated material than for the unirradiated one.

Discussion about creep phenomenon during transport

The creep phenomenon occurring during transport has not been much studied in the range of temperatures from 400°C to 600°C, which is a domain between the reactor normal operating conditions (< 400°C) and the reactor accident conditions (> 600°C). Moreover, the study of creep characteristics requires a complex analysis since parameters such as internal pressure of the rods, thickness of the oxide layer, hydrogen content and orientation of the subsequent hydrides have to be taken into account for an accurate analysis.

Nevertheless, in a first approximation, it is possible to consider only the internal pressure to estimate the deformation and time to rupture. In this case, simple calculations show that creep is a self-limiting phenomenon. Indeed, creep induces an expansion of the cladding volume leading to a decrease in the internal pressure and consequently in the stress in the cladding; creep stops when the stress becomes sufficiently low. However, in the mechanical analysis, it must be checked that there is no interaction between rods and structural components of the fuel assembly (such as grids) which could induce additional shear stresses at the interface.

OXIDATION BEFORE AND DURING TRANSPORT UNDER AIR

During a transport of fuel assemblies in a package cavity filled with air, the cladding of the fuel rods will be oxidized by oxygen. The oxidation rate is influenced by the already present oxide thickness of the cladding; this oxide thickness increases during in-reactor irradiation.

The growth of the oxide layer (ZrO_2) is characterized experimentally by the weight gain per unit surface exposed (due to oxygen pickup). Part of the zirconium cladding is consumed during the zirconia (ZrO_2) growth. This leads to a decrease of the cladding metallic thickness (e) compared to its initial thickness (e_0), which has to be taken into account in the mechanical analysis of the rods behaviour.

The Pilling-Bedworth ratio of zirconia, defined as the ratio of the volume of the metal oxide to the volume of the consumed metal, is 1.54. Thus, the order of magnitude of the remaining cladding metallic thickness (resistant thickness), e, could be estimated by the following relationship: $e = e_0 - e_{ZrO2} / 1.54$, with e_0 the initial cladding thickness and e_{ZrO2} the oxide thickness. Besides, loss of the oxide layer ("spallation") could be observed when the oxide thickness exceeds 90 µm.

Oxidation in reactor normal operating conditions (LWR)

Measurements made on PWR rods with low tin Zy-4 cladding alloy (AREVA Zy-4), irradiated in EDF reactors show that the oxide thickness could sometimes exceed 100 µm at 60 GWd/tHM ([11]), corresponding to a ratio e/e0 ≈ 0.89 for an initial cladding thickness e_0 = 0.57 mm). Due to chemical composition and fabrication process, M5^{TM} cladding-alloy is more resistant to water-side corrosion. The oxide thicknesses measured on M5^{TM} fuel rods irradiated up to 80 GWd/tHM do not exceed 40 µm ([11], [12]), corresponding to a ratio e/e0 ≈ 0.95 (e_0 = 0.57 mm).

Oxidation during transport under air environment

Tests have been performed on unirradiated Zy-4 cladding samples (non pre-oxidized) to estimate the oxide growth for exposures comprised between 38 days at 500°C and 420 days at 350°C ([9]). The results show that the evolution is close to linear, thus an extrapolation to exposures up to 365 days, maximal duration of a transport, could be reasonably made.

For irradiated Zy-4 and unirradiated or irradiated M5^{TM} claddings, data are available for exposures reaching 1000 hours (41 days) at 300°C and 600 hours (25 days) at 500°C ([10]). The evolution seems rather parabolic, and extrapolation to longer duration is not appropriate.

From these data, laws could be elaborated to give an order of magnitude of the additional oxide thickness created during transportation. Comparisons could be made between Zy-4 and M5^{TM} claddings and between irradiated or unirradiated claddings.

For example, for a 41 day transportation at 500°C, the oxide thickness created is estimated to:

- around 46 µm on unirradiated CW Zy-4 claddings (e/e_0 ≈ 0.95 for e_0 = 0.57 mm)
- around 44 µm on unirradiated M5^{TM} claddings (e/e_0 ≈ 0.95 for e_0 = 0.57 mm),
- around 50 µm on CW Zy-4 claddings irradiated up to 50 GWd/tU (e/e0 ≈ 0.88, for e_0 = 0.57 mm and with 53 µm of oxide created during in-reactor irradiation),
- around 36 µm on M5^{TM} claddings irradiated up to 50 GWd/tU (e/e0 ≈ 0.94, for e_0 = 0.57 mm and with 20 µm of oxide created during in-reactor irradiation).

Discussion about the oxidation during transport under air

The available data show that the oxide thickness growth could become very important for cladding temperatures of about 500°C, which is the present temperature limit admitted for LWR fuel rods in normal conditions of transport. Thus, any mechanical analysis of irradiated fuel assemblies should take into account the decrease of the cladding metallic thickness (resistant thickness) due to oxidation.

HYDROGEN PICKUP OF ZY-4 AND M5™ CLADDINGS

Hydrogen pickup is a consequence of irradiation and oxidation of the cladding by the water of the coolant in LWR reactor. The reaction between the zirconium of the cladding and the water of the coolant is: $Zr + 2\ H_2O \rightarrow ZrO_2 + 4\ H$

The hydrogen produced is only partly absorbed by the cladding, this fraction, noted F(%), is called « hydrogen pickup ». The order of magnitude of the hydrogen pickup is 15% for CW Zy-4 PWR rods [13] and only 7.3% for M5™ PWR rods [14].

The hydrogen contained in the cladding is either dissolved in the crystal lattice of zirconium, and is then invisible, or precipitated as hydrides platelet, which can be observed through metallography. The maximum content in hydrides, Cs, is called hydrogen solubility and depends on temperature and its evolution (heating or cooling).

Hydride precipitates form preferentially near the outer (cooler) surface of the cladding:

- either in uniform distribution,
- or in the form of hydride *rim* (continuous layer containing a high concentration of discrete hydrided particles),
- or, under conditions where oxide spallation, occurs, in the form of hydride *blisters*, often lens shaped (typically a few mm in major dimension) consisting in a very high hydride concentration or in a solid hydride.

Blisters and rim induce a significant loss of ductility and recent studies suggest that a ductile-to-brittle transition occurs with increasing hydride rim thickness. The orientation of the hydrides, radial or circumferential, is an important parameter. The radial hydrides are the most harmful as they reduce the mechanical resistance of the cladding, which has to be taken into account in the mechanical analysis.

Only circumferential hydrides are observed in irradiated stress relieved Zy-4 claddings. In the case of Zy-4 claddings with high hydrogen concentration, a thermal dissolution of the hydrides is possible at the temperatures reached during transport (beyond 400°C). After transport, if the fuel rods are immersed in water for cooling, a precipitation of radial hydrides in the cladding could occur if it is subjected to tangential stresses.

In M5™ claddings, due to the low hydrogen content, all the hydrogen is in solution at reactor operating temperature (~350°C). At room temperature, only uniform distributions of hydrides are observed up to 75 GWd/tHM. Rim and blisters, observed in Zy-4 claddings, were not observed in M5™. On the other hand, a reorientation of the hydrides in the radial direction could be observed in M5™ cladding for a lower stress level than in CW Zy-4, which is about 75 to 90 MPa. This stress level could be reached during transport, but the temperature is too high to induce a precipitation of radial hydrides. However, if the fuel rods are immersed in water for cooling after transport, this precipitation could occur due to the decrease of cladding temperature.

FRACTURE TOUGHNESS

Fracture toughness, K_{IC}, is an intrinsic macroscopic material property that quantifies the resistance to crack extension. It is strongly influenced by the hydrogen content of the cladding,

and thus by irradiation: a decrease in the fracture toughness is observed as the hydrogen content increases.
A few data are available for CW Zy-4, as presented on the figure hereafter.

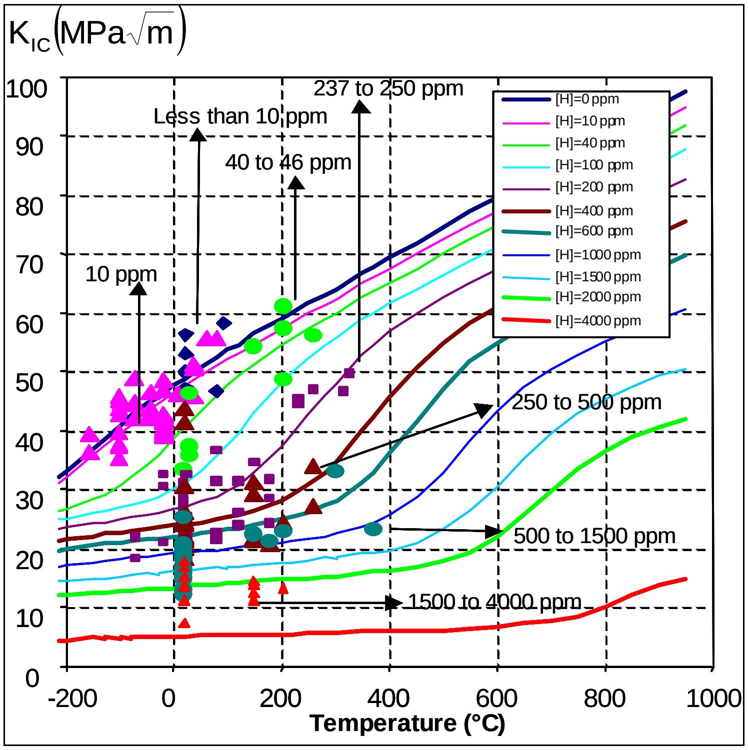

Figure 2. Fracture toughness law of irradiated CW Zy-4 and experimental data [16]

There are no data available on fracture toughness for M5^{TM}. However, impact toughness of RX material, such as M5^{TM}, tends to be greater than the one of CW material. Thus, fracture toughness of M5^{TM} should be higher than that of Zy-4.However, considering the large impact on safety that may have brittleness of cladding material, more information and analyses are needed.

CONCLUSIONS

A survey on physical and mechanical properties of the main zirconium cladding alloys used in the nuclear industry, and more specifically in fuel rod technology, has been performed. Creep properties as well as in-pile or air oxidation properties have been collected in order to simulate cladding creep during transportation. The high strain rate mechanical properties have been collected as well to address accident conditions. All these data, collected in the literature or coming from industry research programs, have been derived as correlations and finally implemented in a software. This tool may be used to provide the data needed when analysing the regulatory transport situations or when assessing these analyses.

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