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DEVELOPMENT AND VALIDATION OF A NEW EXPERIMENTAL DEVICE FOR STUDIES OF IODINE STRESS CORROSION CRACKING OF ZIRCONIUM ALLOYS

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Zirconium alloys are widely used in the nuclear industry as cladding material due to their good thermo-mechanical, chemical and neutronic properties as well as an excellent general corrosion resistance in many environments. Nonetheless, Iodine-induced Stress Corrosion Cracking (ISCC) is an identified failure mechanism that can occur in nuclear fuel cladding as a result of Pellet Cladding Interaction (PCI) during incidental power transients in the reactor. In this study a new experimental device allowing studies of ISCC in a controlled chemical environment is presented. The experiments are performed in an environment that is free of contaminants. First results show that the time to rupture of the specimen is inversely related to the iodine partial pressure. The lowest tested iodine partial pressure, 150 Pa, resulted in an ISCC crack. Post-Mortem microstructural observations provide evidence of iodine corrosion of Zircaloy-4.

I. INTRODUCTION

Zirconium-based alloys are widely used as fuel cladding for nuclear reactors due to their low thermal neutron cross-section and their resistance to corrosion under reactor operating conditions. ¹

Soon after the start of operation of the first civil boiling water nuclear reactors, failures of nuclear fuel cladding caused by pellet cladding interaction (PCI) were observed. PCI is considered as one of the main reasons for failures of zirconium fuel cladding, especially under power transient conditions. ² Since the identification of the problem in the 1960s, a great deal of research has been focused on understanding PCI, which has led to the identification of preventive measures, such as the control of power ramps. Currently, with the increased use of renewable energy there is strong interest to better understand

the PCI phenomena and ease the stringent operational restrictions.

One of the phenomena classified as PCI is Iodine Stress Corrosion Cracking (ISCC). During a power transient and the resulting thermal load, the fuel pellet expands and exerts a high stress on the fuel cladding. Due to this expansion, the fuel partitions and exhibits radial cracks that grant a path for the migration of corrosive fission gases, including iodine, towards the cladding. There are many factors that affect ISCC, e.g. iodine concentration, oxygen concentration, irradiation, composition, microstructure. ^{3, 4, 5}

Many experiments have been conducted to study ISCC in zirconium alloys. However, most experimental devices have contained materials that are not found in the fuel cladding in the reactor, such as stainless steel, copper, aluminum etc.. Because iodine reacts readily with all of these materials, iodides can form that affect the chemistry of the experiments and influence the conclusions drawn from the studies. For example, ISCC crack sites are found near high local impurity concentrations, which are believed to accelerate ISCC. ⁶ The presence of foreign materials, such as silicone or stainless steel have been shown to influence the rate of formation of cracks. ⁷ Thus, there is a need to develop a new experimental device that will allow studies of ISCC in a controlled chemical environment without any reactive foreign materials that can affect the experiments.

II. EXPERIMENTAL SET-UP

II.A. Objective

The goal of this work was to design an experimental device that will allow studies of the chemical aspects of ISCC, in particular the effect of iodine partial pressure, the effect of oxygen partial pressure, the role of ZrI_x compounds, and the incubation time. The study of other factors, such as applied stress and surface conditions, is possible as well.

II.B. Material

The samples used in this study consisted of as-received, unirradiated Cold Worked Stress Relieved (CWSR) Zircaloy-4 cladding material. The dimensions of a cladding tube are as follows: external diameter equal to 9.5 mm, thickness equal to 0.57 mm. The composition of the material is listed in Table 1. Each specimen was manufactured in the form of a C-ring of 10 mm width with a slit opening of 3.8 mm. After carefully considering several candidates, the C-ring geometry was chosen because it can be loaded with sufficiently high levels of hoop stress (approximately 550 MPa or more) on the inner surface with use of relatively small loads (around 100 N).⁸

TABLE 1. Chemical composition of the as-received Zircaloy-4.

Element	Sn	Fe	Cr	O	Zr
Content (wt. %)	1.3	0.22	0.12	0.12	Bal.

II.C. Scheme of experimental set-up

The experimental setup is presented in Figure 1. It was designed to study ISCC of Zirconium alloys caused by iodine vapor in an open system, where the desired flow of the gas is assured by use of a flowmeter. Argon is used as a carrier gas that passes through an oxygen ion pump that controls the oxygen partial pressure. Subsequently, the gas passes through the iodine source that controls the iodine partial pressure. The gas arrives to the surface of the Zircaloy-4 specimen placed in the test chamber, where the temperature of the experiment is controlled by a heating system, and the specimen is subjected to a constant tensile load.

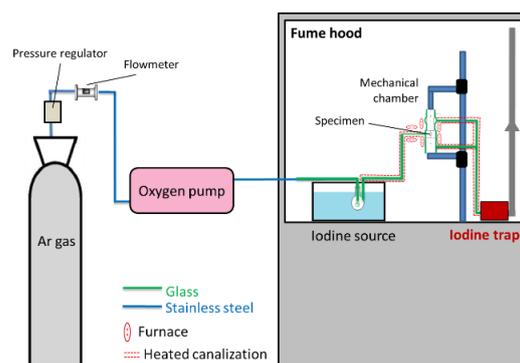


Fig. 1. Experimental setup used to study ISCC.

II.D. Oxygen partial pressure

The oxygen partial pressure is controlled by the use of an oxygen ion pump (*GEN'AIR Oxygen pump-gauge* manufactured by SETNAG), which applies a voltage to a zirconia tube that the gas passes through, thereby adding or removing oxygen molecules as a function of the applied voltage. The detailed functioning of the pump is described by Zhang et al. 2015.⁹ It has been concluded that the minimum controllable oxygen concentration obtained in the current setup is 0.1 Pa.

II.E. Iodine partial pressure

The iodine partial pressure is controlled by regulating the temperature of an iodine sublimation reaction. A glass container containing solid iodine beads is submerged in a water bath with a specific temperature, and this temperature fixes the partial pressure of the iodine. Figure 2 shows the relation between the temperature and the resulting partial pressure of iodine. The data used to create this relationship were calculated using FACTSAGE software by DEC - Département d'Etudes des Combustibles (CEA Cadarache, France)¹⁰ from the data coming from OECD's project "Thermodynamics of Advanced Fuels – International Database (TAF-ID) Project".¹¹

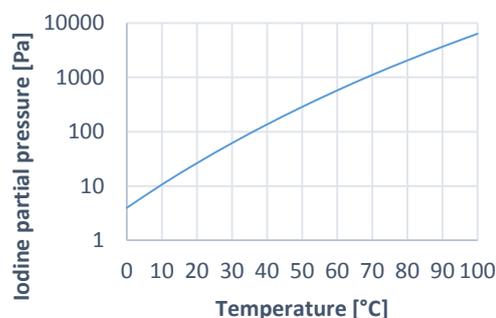


Fig. 2 Theoretical iodine partial pressure as a function of temperature.¹¹

II.E. Instrumentation

Due to the fact that iodine can react quickly with many materials, including most metals, the experimental device was built with use of inert materials. The parts of the experimental device that are in direct contact with the iodine vapor are made of either glass or PTFE.

III. RESULTS

In order to validate the experimental set-up and study ISCC, constant load creep tests in an iodine vapor atmosphere were performed. The temperature of tests was set as 380°C. Figure 4 presents the deformation of the specimen and the force applied to the specimen as a function of time. The deformation (depicted on Figure 3) is defined as:

$$Deformation[mm] = H - H_0 \quad (1)$$

Where:

H- Height of deformed specimen.

H₀ - Initial height of the specimen.

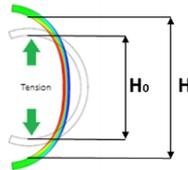


Fig. 3 Definition of the applied force and of the deformation of the specimen during the experiment.

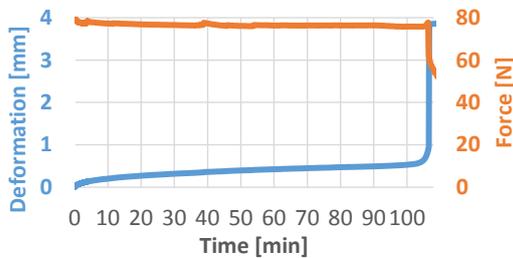


Fig. 4 Deformation of the specimen and the force applied to the specimen as a function of time.

In Figure 4, one can observe the evolution of the deformation of the specimen, where a relatively quick primary creep phase is followed by a longer secondary creep phase. During this test the load of 78 N was applied, what corresponds to hoop stress on the inner surface of the specimen equal approximately to 550 MPa (immediately after applying the load) and relaxing down to 450 MPa (at the time of failure) due to visplastic behavior of the material.⁸ The failure of the specimen occurs at $t=105$ minutes, when a stress corrosion crack forms and causes the sample to open rapidly.

Figure 5 presents a comparison of creep tests performed with and without iodine vapor. The temperature of the specimen was set at 380°C. The test without iodine vapor was performed using a slightly higher load (83 N) than the test with iodine (76 N), which is why the strain rate is slightly higher. Nevertheless, the specimen without iodine did not fracture after 6 hours, whereas the specimen that was exposed to iodine fractured in less than 2 hours.

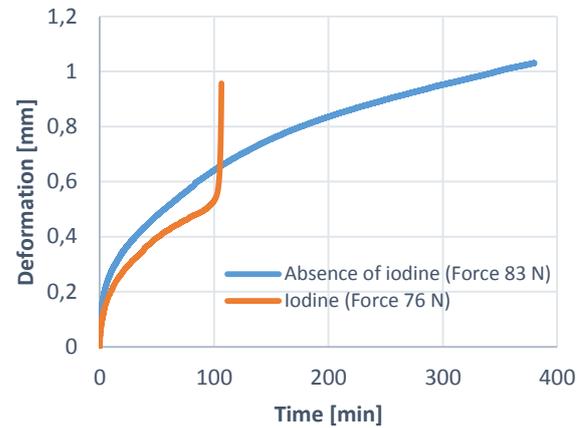


Fig. 5 – Deformation of specimens during creep tests performed with and without iodine vapor.

Figure 6 shows the relation between the time to failure of the specimen and the iodine partial pressure during constant load creep tests. Three values of iodine partial pressure were investigated: 150, 320 and 640 Pa. In all tested conditions rupture by ISCC occurred within less than 3 hours. One can observe a significant effect of the iodine partial pressure on the time to failure. An increase of iodine partial pressure causes a quicker failure by ISCC. Thus it promotes ISCC susceptibility of Zircaloy-4. These results are consistent with earlier studies, where the time to failure is inversely related to the iodine concentration.^{12, 13, 14}

Several researchers have suggested that there is a threshold for the iodine partial pressure, below which failure by ISCC does not occur.^{12, 13, 14} Peehs suggested that the critical partial pressure of iodine for ISCC is approximately 400 Pa. More recent studies by Anghel and Chen show that the threshold iodine partial pressure is lower than 60 Pa for irradiated Zircaloy-2 and 100 Pa for unirradiated Zircaloy-4 respectively. This study agrees with Anghel and Chen and supports the hypothesis that ISCC of zirconium alloys can occur under very low concentrations of iodine, as was proposed by Cubicciotti and Sidky.^{3, 15}

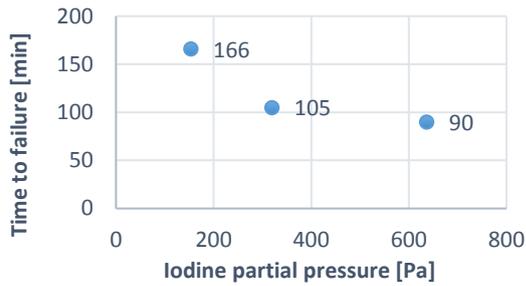


Fig. 6. Time to failure of the specimen as a function of iodine partial pressure.

Fracture morphology and qualitative identification of elements on the surface of the specimen were performed using SEM (JEOL IT300) and EDS analysis (OXFORD Instruments piloted by AZTEC software). The observations of the crack face indicate that the fracture is initially brittle and then becomes ductile [Figure 7]. The brittle region is characteristic of ISCC cracks observed by previous researchers, where corrosive iodine activity causes embrittlement of the material. The brittle crack propagates until the local stress becomes sufficient to pull the specimen apart in a ductile manner. Many corrosion pits can be observed on the surface of the specimen [Figures 7 and 8]. Those findings are consistent with the microstructural observations of ISCC cracks resulted from internal pressure test.^{16, 17}

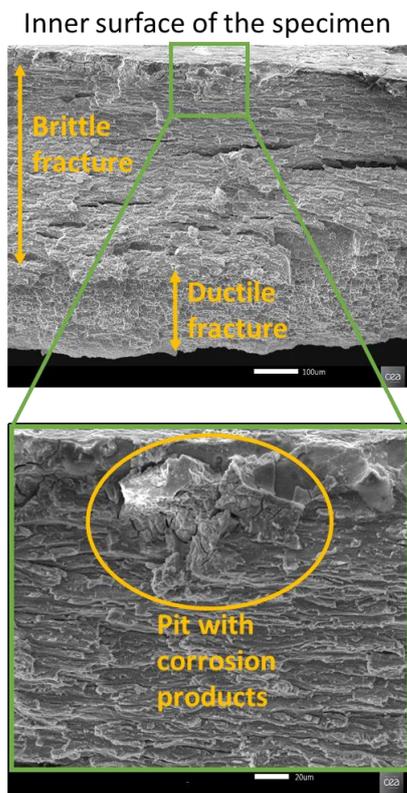


Fig. 7 – Microstructure of a crack face.

Iodine reacts quickly with zirconium creating zirconium iodides (ZrI_n with n from 1 to 4). At first, when iodine is in contact with Zr, it creates ZrI . Then, with more iodine coming, ZrI becomes ZrI_2 . In the same way ZrI_3 and ZrI_4 must form later, also in sequence. From all zirconium iodides only ZrI_4 is volatile at cladding temperatures.³ It is believed that the agglomeration of corrosion products in the pit presented on the Figure 7 is composed of a mixture of solid ZrI_x compounds. This hypothesis remains to be confirmed.

The Figure 8 presents the evidence of pitting corrosion caused by iodine. Qualitative EDS analysis was performed that showed the presence of iodine in the pits.

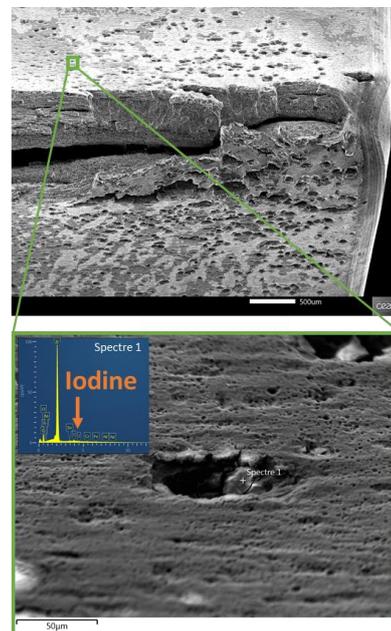


Fig. 8 – EDS analysis of a corrosion pit on a Zircaloy-4 surface.

IV. CONCLUSIONS

A new experimental device allowing studies of ISCC in a controlled chemical environment has been developed and validated. The experiments are performed in an environment that is free of contaminants, in which all components are chemically inert. Initial experiments prove the functionality of the set-up. Early results show that the time to rupture of the specimen is inversely related to the iodine partial pressure, and thus a greater iodine concentration promotes ISCC susceptibility of Zircaloy-4. This study supports the hypothesis that ISCC of zirconium alloys can occur under very low concentrations of iodine, with cracks observed at 150 Pa. The microstructural observations provide evidence of iodine corrosion of Zircaloy-4.

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