RESEARCH ARTICLE

Study on hydrogen permeation method of zirconium alloy tube

Du Hao
R & D Center, Department of Biotechnology, Dayananda Sagar College of Engineering, Bengaluru 560078, Karnataka, India

Abstract: Physical and Chemical Testing Center of North Nuclear Fuel Components Co., Ltd., Baotou, Inner Mongolia, 014035, China The determination process of hydride orientation factor of zirconium alloy mainly includes hydrogen permeation process and hydride display process. The key of hydride orientation factor measurement lies in controllable and effective hydrogen permeation and true and clear display of hydride. When a metallographic picture with appropriate number of hydride and clear display is obtained, the hydride orientation factor of sample can be accurately and quickly measured. By analyzing and testing the possible factors affecting the hydrogen permeation effect of zirconium alloy tube, the corrosion parameters such as temperature, pressure, type, concentration and holding time of corrosion solution were determined.
Keywords: Zirconium alloy; hydrogen permeation; hydride orientation factor.

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1 Introduction

The operating conditions of zirconium alloy tubes used as cladding materials of nuclear fuel elements in nuclear reactors determine that hydride generation is inevitable, in which the existence of radial hydride reduces the fracture toughness of zirconium alloy, and cracks spread through radial hydride, resulting in cladding tube rupture [1-2]. Among the technical conditions of cladding tubes for nuclear fuel elements, hydride orientation factor, as an important detection requirement, is listed in the domestic and foreign standards. The effect of hydrogen permeation directly affects the validity of hydride orientation factor detection results.

And accuracy. Therefore, we have analyzed and tested the possible influencing factors of hydrogen permeation effect of zirconium alloy tubes, hoping to obtain hydrogen permeation samples with appropriate hydrogen permeation amount and uniform hydride distribution through a controllable and effective hydrogen permeation process.

2 Test Materials and Test Design

2.1 Test Materials

The tube selected in the hydrogen permeation test is zirconium 4 alloy tube purchased from ZPI Company of Canada, and the state of mechanical addition and heat treatment is as follows: reducing the diameter, extruding, relieving stress and annealing.

2.2 Test Design

2.2.1 Selection of hydrogen permeation methods

At present, the mature hydrogen permeation methods mainly include autoclave hydrogen permeation and hydrogen furnace hydrogen permeation. Autoclave hydrogen permeation is that lithium hydroxide solution is added into the autoclave container as hydrogen permeation liquid [3], and hydrogen permeation is carried out at a certain temperature and pressure. Hydrogen furnace hydrogen permeation is that the temperature of the hydrogen furnace is controlled at a certain temperature, the flow rate and pressure of hydrogen are controlled in the hydrogen atmosphere, and zirconium alloy tubes are subjected to hydrogen permeation. The samples after hydrogen permeation must undergo vacuum desorption heat treatment. Considering the simplicity of operation and the universality of equipment, we choose autoclave for hydrogen permeation [4].

2.2.2 sample selection and sample treatment before hydrogen permeation

Considering the needs of hydrogen content measurement and metallographic sample making, Zr-4 alloy tube with a length of 15mm-20mm was selected as the experimental sample. In order to facilitate the post-processing of samples, try to intercept samples without oxide layer on the cross section and inner and outer surfaces.

According to the general requirements of autoclave corrosion experiment [4], before hydrogen permeation, the intercepted samples should be cleaned (to remove zirconium scraps and pollutants on the surface of the samples), ground (to polish the end face of the samples with diamond sandpaper to remove the cross-section deformation caused by cutting) and cleaned (to put the ground samples in mixed acid 3% ~ 7% hydrofluoric acid, 40% ~ 50% nitric acid and water for cleaning to remove zirconium -4 alloy tubes.

2.2.3 Design of hydrogen permeation parameters of autoclave

The principle of autoclave hydrogen permeation test is the same as that of autoclave water corrosion test. According to the conditions of autoclave water corrosion [4], the corrosion temperature is (360 3) ℃ and the corrosion pressure is (18.7±0.5)MPa. The amount of hydrogen permeation can be controlled by adjusting the concentration of lithium hydroxide solution and holding time. For this reason, we plan to conduct several groups of pre-experiments, and then design experiments with different concentrations of lithium hydroxide solution and different holding times according to the pre-experiments.

2.2.4 Test plan of samples after hydrogen permeation

We plan to test the weight gain, hydrogen content and metallographic picture of the sample after hydrogen permeation. On the one hand, we can measure the degree of hydrogen permeation of the sample in the autoclave; on the other hand, the measured results will be the main basis for judging whether the hydrogen permeation effect is suitable or not.

3 Test Results and Analysis

The temperature (360 3) ℃ and pressure (18.7±0.5)MPa are fixed in the experimental parameters, and the experimental parameters that affect the hydride display, weight gain and hydrogen content of the sample are determined, including the concentration of experimental medium solution and the holding time of hydrogen permeation experiment. In order to determine the best concentration of experimental medium and holding time, many experiments were carried out. Table 1 and Table 2 show the changes of weight gain and hydrogen content of samples with the increase of holding time when the concentration of experimental medium is 1mol/L, 1.5mol/L and 2.0mol/L.

Table 1 Weight increase unit of sample in experiment: mg

Concentration (mol/L) / Time (h)	1.0	1.5	2.0
five	9.6	10.7	12.2
six	9.9	10.9	12.5
seven	10.2	11.0	12.5

Table 2 Hydrogen content of samples in experiment unit: μg/g

Concentration (mol/L) / Time (h)	1.0	1.5	2.0
five	69.5	268.4	587.8
six	137.2	278.5	724.5
seven	169.4	342.7	698.0

It can be seen from the trend diagrams of weight gain and hydrogen content of samples in Table 1 and Table 2 that when the concentration of experimental medium changes from 1.0mol/L to 2.0mol/L, the weight gain and hydrogen content of samples change slowly, while when the concentration of experimental medium changes from 1.0mol/L to 1.5mol/L, the weight gain and hydrogen content of samples increase rapidly, which shows that when the concentration of experimental medium is greater than 1mol/L, the chemical reaction of experimental samples increases rapidly. Through the metallographic analysis of the samples with different concentrations and different times, it can be seen that when the concentration of experimental medium is 1.5mol/L and the experimental holding time is 6 hours, the hydride analysis effect is the best, and the hydrogen content of the samples can be controlled at (200 ~ 300) μ g/g.

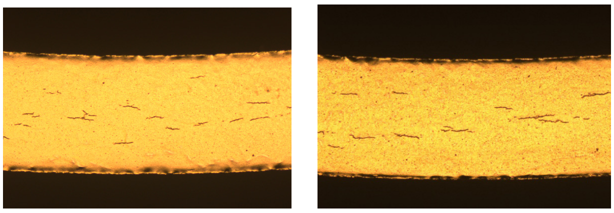

Fig. 1 Metallographic picture of sample with 100μg/g H content

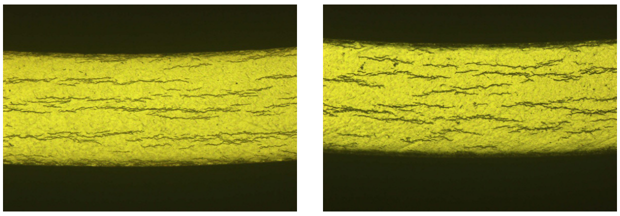

Fig. 2 Metallographic pictures of samples with 200μg/g-300μg/g H content

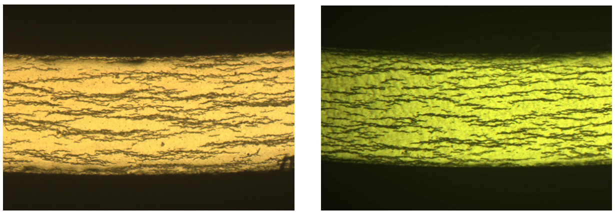

Fig. 3 Metallographic picture of the sample with the content of 500μ g/g-1000μ g/g.

4 Conclusion

(1) The hydrogen content of the sample is (200 ~ 300) μ g/g, which is beneficial to the measurement of hydride orientation factor;

(2) Under the conditions of autoclave water corrosion, that is, temperature (360 3)℃ and pressure (18.7±0.5)MPa, the hydrogen permeation parameters: the concentration of lithium hydroxide solution is 2.0mol/L, and the holding time is 6 hours, the expected hydrogen permeation effect can be obtained.

References

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