11th International Conference on Advanced Materials
Rio de Janeiro Brazil
September 20 - 25
ICAM2009

Kinetics Analysis of Zircaloy-4 Hydriding between 20 and 670 °C

I. S. Dupim(1), J. M. L. Moreira(1)*, S. L. Silva(2), C. C. G. e Silva(2), O. Nunes Jr.(2) and R. G. Gomide(2)

(1) CECS, Universidade Federal do ABC, Rua Santa Adélia 166, 09210-170 Santo André, SP, e-mail: joao.moreira@ufabc.edu.br
(2) Centro Tecnológico da Marinha em São Paulo, Av. Professor Lineu Prestes 2468, 05508-000 São Paulo, SP, e-mail: cecilia.chaves@ctmsp.mar.mil.br
* Corresponding author.

Abstract – Nuclear fuels based on composite materials such as dispersion of uranium compounds in a Zirconium alloy matrix are possible alternatives for advanced 4th generation nuclear technology. A necessary step for obtaining such fuels is producing Zr alloy powder for the metal matrix composite materials. This article presents some results from the kinetics of the Zircaloy-4 hydrogenation reaction with the purpose to embrittle and, subsequently, comminute the alloy. The data are obtained at low temperatures and pressures do reduce fabrication costs. The kinetics analysis showed that the reaction rate starts with a linear dependency on the H_2 partial pressure but, as the Hydrogen concentration in the Zircaloy-4 increases, the reaction rate decreases.

Nuclear fuels based on composite materials such as dispersion of uranium compounds in a metal matrix are possible alternatives for advanced 4th generation nuclear technology. Zr alloys are good options for a metal matrix due to their low thermal neutron absorption cross-section, good corrosion resistance and high thermal conductivity. A necessary step for obtaining such fuels is producing Zr alloy powder for the metal matrix composite materials. This article presents some results from the kinetics of the Zircaloy-4 hydrogenation reaction with the purpose to embrittle and, subsequently, comminute the alloy.

Several hydrogenation tests were performed and studied through thermal gravimetric analysis (TG). The Zircaloy-4 chips used were provided by INB (Indústria Nuclear do Brasil). The chips had an approximate rectangle cross-section shape with about 60 mm^2, and were cut into lengths of approximately 2 mm. The samples compositions were characterized with a Fluorescence X-ray EDS (HS EDX-800 Shimadzu). All had a chemical composition within the acceptable value for the Zircaloy-4 alloy, except for Fe with content 0.03% higher than in the literature.

The hydriding tests included H_2 pressures of 50 and 25 KPa and temperatures ranging from 20 to 670 °C. Figure 1 shows the mass variation due to H absorption for Test 5 which was run with a constant H_2 partial pressure of 25 KPa. The scattering in the data is due to fluctuations in the TG scale which is very sensitive. The samples undertook the following temperature profile: beginning with 18 °C which, kept in this value during the first 1,5 minutes; temperature increased with a constant rate of 20 °C/min; at the end a plateau was reached for isothermal analysis at 570 or 670 °C. The experiments yielded some kinetics information about the Zircaloy-4 hydriding reaction.

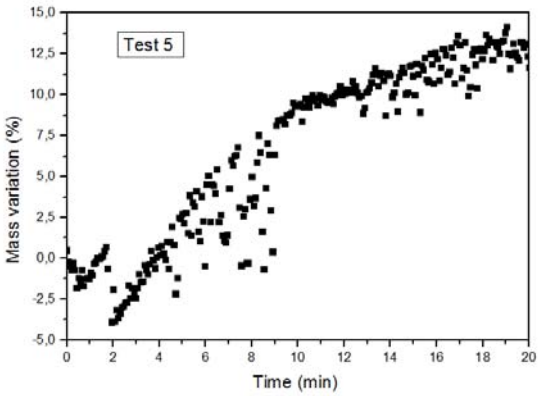

Figure 1: Mass variation due to Hydrogen absorption by the Zircaloy-4 as a function of time. H_2 partial pressure was 25 KPa and the heating rate was 20 °C/min.

Table 1: Sieverts constant for the Zircaloy-4 hydrogenation. Results obtained at equilibrium H concentration in the Test 9 sample.

H_2 pressure	25 KPa
Temperature	670 °C
Sieverts constant	0.15 $Pa^{-0.5}$

During the initial 2 minutes the sample appears to lose mass, about 3 %. This may be due to some physical phenomena or, simply, due to the scale stabilization. The Zircaloy-4 sample initial mass was 40.70 ± 0.014 mg. Then, as the temperature starts to increase steadily, the mass gain due to H absorption increases. It can be seen in Figure 1 that the rate is linear with time at the beginning but, as the H content in the sample increases, it slowly decreases.

The hydrogenation reaction is a reversible process with absorption and desorption of Hydrogen [1]. As the H concentration increases, the rate of H uptake decreases. Table 1 presents the Sieverts constant obtained for the Test 9 which accounts for absorption and desorption of H (0.15 $Pa^{-0.5}$). It was obtained at equilibrium conditions of Hydrogen uptake and release by Zircaloy-4 at 670 °C and H_2 pressure of 25 KPa.

[1] M. Steinbruck, Hydrogen absorption by zirconium alloys at high temperatures. J. Nucl. Mat. 334 (2004) 58.