Oxidation Behaviors and Its Kinetics of Zircaloy-4 in Air

Je Geon Bang, Sun Ki Kim, Tae Hyun Chun

Korea Atomic Energy Research Institute, Daedeok-daero 989, Yuseong-gu, Daejeon, Korea, 305-353
[*]Corresponding author: jbbang@kaeri.re.kr

1. Introduction

Behaviors of spent fuel in air environment in a spent fuel pool after LOCA has been discussed since Fukushima accident. It may be possible that exposure of spent fuel cladding to air during a hypothetic severe accidents such as loss of cooling and/or loss of coolant in spent fuel pool. It is well known that oxidation in air leads to a strong degradation of the cladding material and the barrier effect of the fuel cladding is lost much earlier than during accident transients with a steam atmosphere only[1-3].

In comparison with steam, chemical heat release during oxidation in air is higher by 80%, which may lead to a more rapid evolution of the accident[4-6]. Additionally, the oxidation kinetics in air is much faster than in steam due to the formation of non-protective oxide (nitride) scales[1]. From the safety point of view, the barrier effect of the cladding against release of fission products is lost much earlier in air compared to steam.

The aim of this study is to investigate the oxidation behaviors of nuclear fuel cladding and to generate its kinetic data during an accident in spent fuel pool. In this study, the oxidation behaviors and its kinetics of the Zircaloy-4 were investigated in air environment for various air ingress scenarios in the temperature range 600℃～1,400°C by thermo-gravimetric analysis.

2. Experimental details

2.1. Specimens

2 cm long Zircaloy-4 tube segments (9.50 mm outer diameter, 0.570 mm wall thickness) were cut from longer tubes, deburred, ground at both ends, and cleaned in an ultrasonic bath of acetone. The samples were positioned vertically in the furnace of the TGA. The specimens were open at the ends, as a result of which, the specimen were oxidized on both sides.

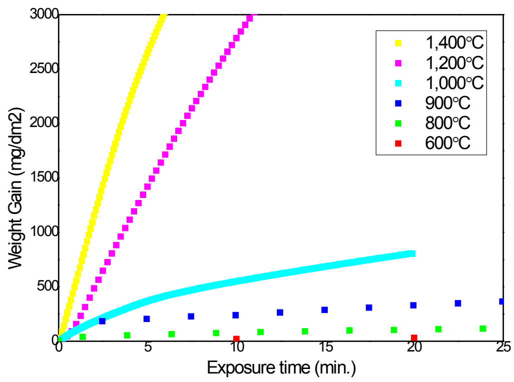
2.2. Test procedure

For isothermal tests, the specimens were heated to the desired temperature at a rate of 30 K/min in argon and thermally equilibrated for 10 min. Then, the air was injected with a flow rate of 50 sccm. The tests were finished by switching off the oxidizing gases(air) and cooling down the furnace as fast as possible with an argon atmosphere.

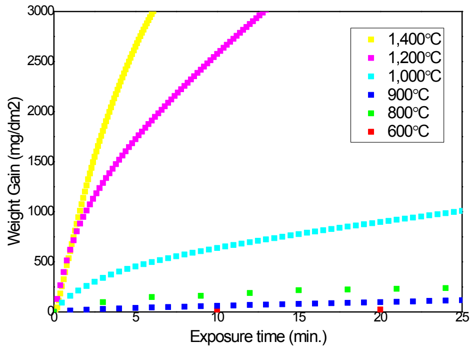
3. Results

Oxidation behaviors of Zircaloy-4 in air were shown in Fig.1. As shown in Fig.1, weight gain with exposure time was decreases with time, and the weight gain behavior follows parabolic kinetic law.

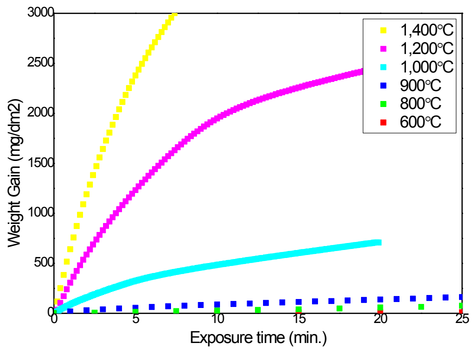
And also, based on the weight gain behavior with oxidation time, the rate constant were withdrawn at each temperature. The Arrehenius plot was presented in Fig. 2.



(a)



(b)



(c)

Fig. 1 Oxidation behaviors of Zircaloy-4 in air
(a) bare, (b) ~20 μ m pre-oxidized,
(c) ~55 μ m pre-oxidized,

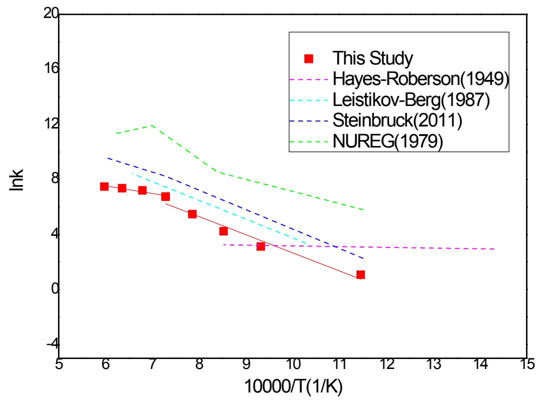


Fig. 2 Oxidation rate constant vs. temperature

The oxidation rate constant is as follows.

$k = 7.50\ x\ 102exp(-109,524/RT)\ (T<1373K)$
$k = 5.31exp(-46,365/RT)\ (T>1373K)$

4. Conclusions

In this study, the oxidation behaviors and kinetics of the Zircaloy-4 were investigated in air environment. And the oxidation rate constant were withdrawn as follows.

$k = 7.50\ x\ 102exp(-109,524/RT)\ (T<1373K)$
$k = 5.31exp(-46,365/RT)\ (T>1373K)$

ACKNOWLEDGEMENTS

This work has been carried out under the Nuclear Research and Development Program supported by the Ministry of Education, Science and Technology in the Republic of Korea.

REFERENCES

[1] M. Steinbrück, "Separate-effects tests on high temperature oxidation of zirconium alloys in various atmospheres", Workshop on Computational and Experimental Studies of the LWR fuel element behaviour under beyond design basis accidents and reflood conditions, IBRAE, Moscow, 27-28 July 2009.

[2] Martin Steinbrück, Nóra Vér, "High-Temperature Oxidation of Zircaloy-4 in Mixed Steam-Air and Steam-Nitrogen Atmospheres", Proceeding of ICAPP 2010, San Die 해, CA, USA, Jun3 13-17, 2010.

[3] Martin Steinbruck, et. al, "Experiments on air ingress during severe accidents in LWRS", Nucl. Eng. Des. 236 (2006) 1709-1719.

[4] Martin Steinbruck, "Prototypical experiments relating to air oxidation of Zircaloy-4 at high temperatures", J. Nuclear Materials 392 (2009) 531-544.

[5] M. Steinbruck, M. Bottcher, "Air oxidation of Zircaloy-4, M5 and ZIRLO™ cladding alloys at high temperatures", J. Nuclear Materials 414 (2011) 276-285.

[6] C. Duriez, T. Dupont, B. Schmet, F. Enoch, "Zircaloy-4 and M5 high temperature oxidation and nitriding in air", J, Nuclear Materials 380 (2008) 30-45.