A surface-to-volume model of UO_2 oxidation in air at 573 - 723K

Hyounggyu Park[a], Kwangheon Park[a*]

[a] *Department of Nuclear Engineering, Kyunghee University, Kyunggi-do, 446-701, Korea*

**Corresponding author: kpark@khu.ac.kr*

Green
Nuclear
Research
Laboratory

GNRL

Introduction

The mechanism of oxidation of uranium dioxide in the air condition can be described, $UO_2 \rightarrow U_4O_9/U_3O_7 \rightarrow U_3O_8$.

The formation of U_3O_8 leads to an approximately 23% density decrease and a 36% volume increase. The increased volume of U_3O_8 causes a safety problem of long-term storage. Because of the safety problem, more studies are required to estimate the behavior of a uranium oxidation.

In this study, a surface-to-volume model is proposed to describe the kinetic mechanism from UO_2 to U_3O_8. The purpose of this study is to describe the model of UO_2 oxidation mathematically.

Previous model

Johnson-Mehl-Avrami model has been applied to express the sigmoidal curve of the reaction mechanism, nucleation and growth, mathematically.

< Johnson-Mehl-Avrami model >

$$\alpha = 1 - \exp(-K\, t^n)$$

K and n are determined empirically. α is a fraction from UO_2 to U_3O_8 and t is time. The oxidation curve was not expressed by this model as seen in the Fig.1.

Table 1: The values of K and n at 573, 623,673 and 723K

	K	n
573 K	2.665E-8	1.7015
623 K	6.387E-7	1.7248
673 K	2.823E-6	1.7302
723 K	9.629E-5	1.3337

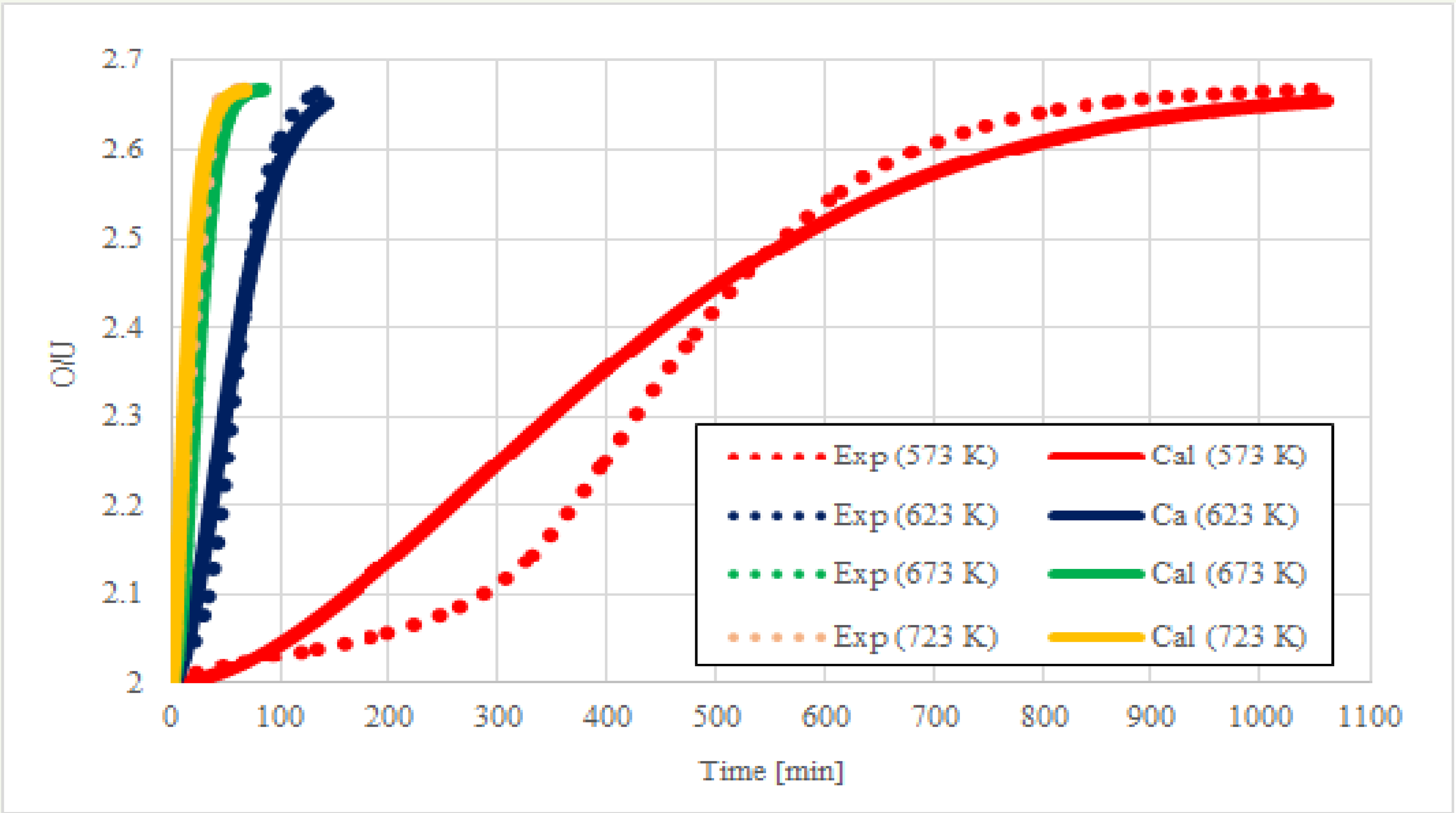

Fig. 1. Comparison of experimental data and calculation data

New model

Johnson-Mehl-Avrami model could not explain the effect of incubation time and cracking of uranium oxide. Then, a surface-to-volume model is proposed to consider the effect of incubation time and cracking. This model was suggested to explain the UO2 oxidation mechanism well.

< A surface-to-volume model >

$$\dot{x} = f(t) \cdot (x_{eq} - x)$$

$$f(t) = A_1\left(1 - \frac{A_3}{1 + A_2 \cdot e^{\alpha t}}\right)$$

$$\mathrm{x(t,T)} = \frac{8}{3} - \frac{2}{3}\left(\left(\frac{1 + A_2}{1 + A_2 e^{\alpha t}}\right)^{\frac{A_1 A_3}{\alpha}} \cdot e^{A_1(A_3 - 1)t}\right)$$

Table 2: The values of A_1, A_2, A_3 and α at 573, 623, 673 and 723 K

	A_1	A_2	A_3	α
573 K	1.413E-4	9.398E-3	0.9846	1.845E-4
623 K	9.095E-4	1.987E-2	0.9729	1.194E-3
673 K	1.855E-3	3.167E-2	0.8492	2.103E-3
723 K	3.584E-3	5.739E-2	0.9316	1.604E-3

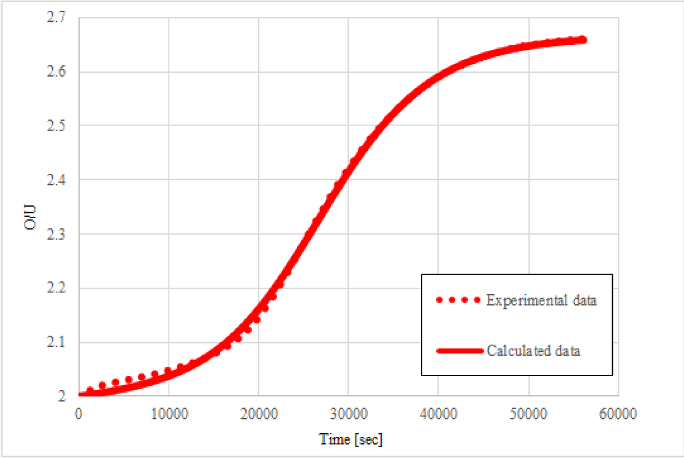

Fig. 2. 573 K

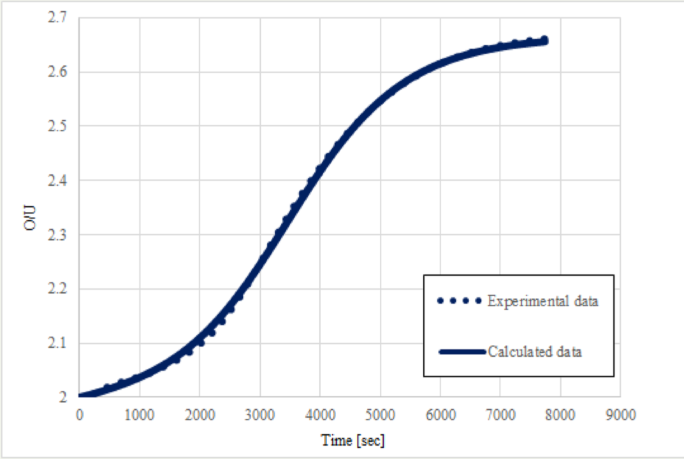

Fig. 3. 623 K

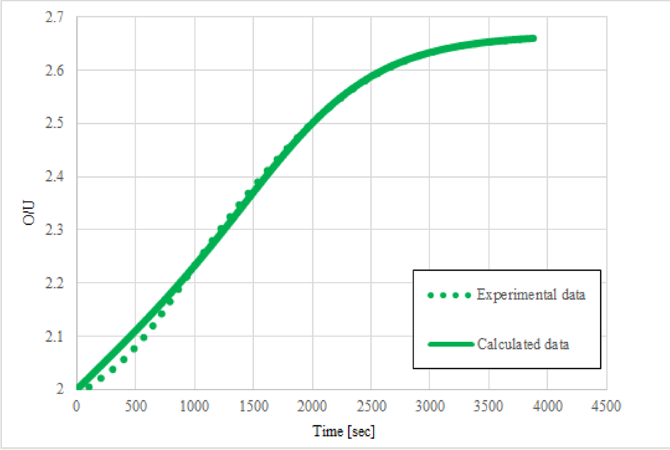

Fig. 4. 673 K

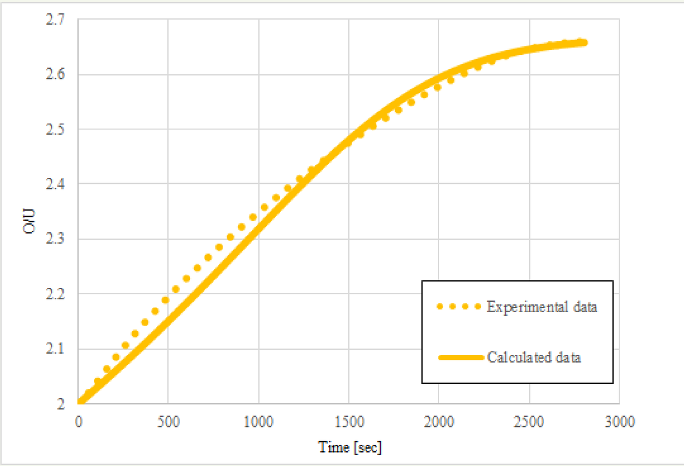

Fig. 5. 723 K

Conclusion

The surface-to-volume model was proposed to describes the sigmoidal curve of the UO_2 oxidation mechanism with time-dependent surface-to-volume ratio. This was found to explain UO_2 oxidation better than the Jonhson-Mehl-Avrami model with the experimental data at 573, 623, 673 and 723 K.