

Development of fission release model accounting for UO₂ oxidation under air atmosphere

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1. Introduction

In KAERI, nuclide management technology is being developed for reduction of disposal area required for spent fuel management and major fission products such as Cesium, Krypton, Xenon and Iodine are considered to be completely removed during thermal treatment under air atmosphere since the thermal treatment at powder state facilitates the release of fission products. For the development of optimal thermal treatment, fission release model applicable to the description of experimental fission product release is desirable allowing the minimization of the required experimental study. However, traditional fission release model is inapplicable to the present condition since the existing model does not account for the change of UO₂ pellet to U₃O₈ powders at voloxidation condition. In this study, two-stage diffusion model accounting for UO₂ oxidation behavior is developed to explain Cesium and Krypton release behavior undergoing pulverization.

2. Model developments

The developed model is based on the coupling of UO₂ oxidation model [1] to modified two-stage diffusion model [2]. Fuel fragment is considered to be approximated to 1D sphere uniformly divided into shells containing grains.

2.1 Oxidation model

A semi-mechanistic model for UO₂ pellet oxidation was developed to describe various oxidation behavior significantly affected by the factors such as fuel geometry, oxygen content and thermal conditions. For model development, the following assumption were made.

- All pellets are approximated to sphere having same ratio of surface to volume
- Cracking occurs due to the formation of U₃O₈ occurs in solid surface yielding to the increased reactive surface area
- Porosity of pellet changes as reaction proceeds
- Oxygen diffusion follows 1D spherical diffusion
- Sphere is equally divided into symmetrical shell

Oxidation model considers both diffusion and reaction representing UO₂ conversion and working equation considering both diffusion and reaction is as follows

$$\frac{\partial \varepsilon_i C_i^*}{\partial t} = \frac{1}{r_i^2} \frac{\partial}{\partial r_i} \left(r_i^2 \varepsilon_i^0 D \frac{\partial C_i^*}{\partial r_i} \right) - k(1 - X_i) C_i^* \theta \quad (1)$$

where X_i represents UO₂ conversion at i^{th} shell(or subvolume). The present oxidation model allows to evaluate oxidation degree of all grains at given temperature, oxidation potential and pellet geometries.

2.2 Modified Two-Stage Diffusion model

Hidaka [2] presented two-stage diffusion model for the release of cesium at helium atmosphere. The model is based on the assumptions that gaseous diffusion through an open pore follows the diffusion through a solid spherical fuel grain. In spherical fuel grain of i^{th} , diffusion is modeled by continuity equation as follows

$$\frac{\partial C_{S,R}(r_i,t)}{\partial t} = \frac{1}{r_i^2} \frac{\partial}{\partial r_i} \left(r^2 D_S \frac{\partial C_{S,R}(r_i,t)}{\partial r_i} \right) \quad (2)$$

where $C_{S,R}$ represent the concentration of fission product in a grain and D_S the diffusion coefficient in the grain. In diffusion through the open pores of a radial fuel pellet, diffusion is governed by the following equations

$$\alpha \frac{\partial C_P(R_i,t)}{\partial t} = \frac{1}{R_i^2} \frac{\partial}{\partial R_i} \left(R^2 \beta D_P \frac{\partial C_P(R_i,t)}{\partial R_i} \right) \quad (3)$$

where α and β represent void fraction of fuel and porosity correction factor and D_p means binary gas diffusion coefficients in open pores. In original context, α and β and D_S is considered as constants, however, these parameters should be modified to account for oxidation degree since in oxidation process void fraction of powder is increased compared to that of UO₂ particle, Therefore, we introduce dependency of oxidation degree into these parameters,

$$\alpha = \alpha_{UO_2} + (0.76 - \alpha_{UO_2}) f(X_i) \quad (4)$$

$$\beta = \beta_{UO_2} + (0.76 - \beta_{UO_2}) f(X_i)^3 \quad (5)$$

$$D_S = D_S(1 + \gamma X_i^3) \quad (6)$$

For the evaluation of solution of eqs (1), (2) and (3), finite volume method is applied with implicit method.

2. Results and Discussion

The developed model requires four model parameters and these parameters were determined by fitting to experimental fission product release data [3]. Although different fission release data are available, the data of HOX1-AR1 and HOX1-AR2 were employed since low-temperature oxidation interval(400°C in air) is included in thermal treatment condition. The thermal condition of

HOX1-AR1 and HOX1-AR2 as revealed in Figure 1 and 2 are similar except to starting temperature of thermal program. Both figures show that the proposed model can roughly represent experimental fission release behavior, especially in Krypton release behavior during low temperature oxidation. In both studies, calculated Cesium is also shown to be lately released compared to Krypton, consistent with experimental behavior.

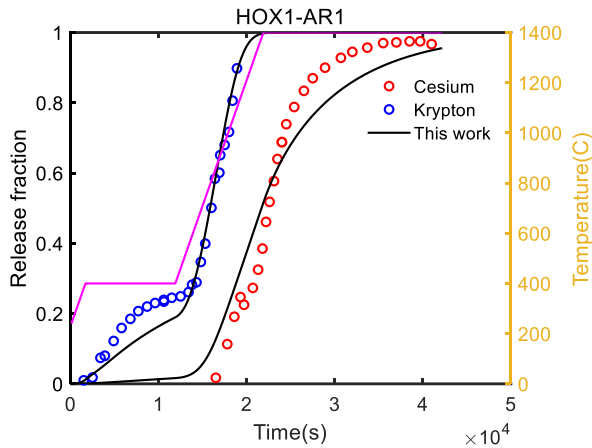


Fig. 1. Correlated Cesium and Krypton release in test AR1.

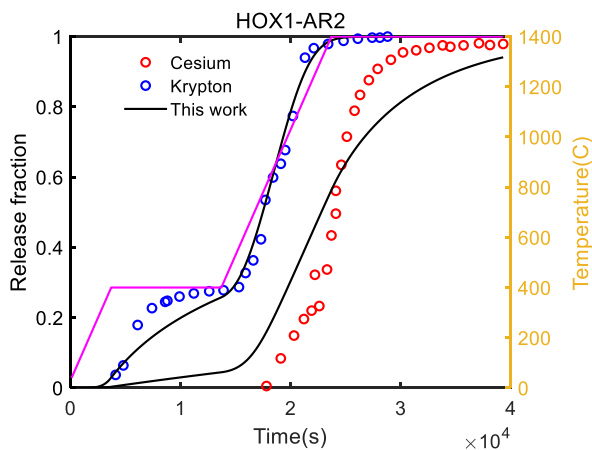


Fig. 2. Correlated Cesium and Krypton release in test AR2.

4. Conclusions

In this study, the two-stage diffusion release model coupled to UO_2 oxidation model was presented and found to reasonably exhibit Cesium release behavior and Krypton release behavior during low-temperature oxidation. The developed model will be applied to the induction of optimal thermal treatment condition for efficient fission product removal

Acknowledgement

This work was supported by the National Research Foundation of Korea (NRF) grant funded by the Korea government (MEST)(No. 2021M2E3A1040059)

REFERENCES

- [1] J. H. Lee, J. W. Lee, S. M. Hong and C. H. Lee and Y. Z. Cho, A Semi-mechanistic model for UO_2 pellet oxidation at 400, 500 and 700°C, Proceeding of the Korean Radioactive Waste Society Conference, Jun 2-4, 2021
- [2] A. Hidaka, T. Kudo, T. Ishigami, J. Ishikawa and T. Fuketa, Proposal of Simplified Model of Radionuclide Release from Fuel under Severe Accident Conditions Considering Pressure Effect, Journal of Nuclear Science and Technology, Vol. 41, p.1192, 2004.
- [3] Z. Liu, D. S. Cox, D. D. Semeniuk, R. D. Barrand, T. J. Kunkel, R. F. O'Connor, D. H. Rose, R.S. Dickson, P. H. Elder, D. F. Shields and G. W. Wood, Fission-Product Releases and Morphologies of Powders Produced by the OREOX Process: Report on the Hox1 Experiment, RC-1375, DUPIC-AR-FT-06, 1995.