Development of fission release model accounting for UO_2 oxidation under air atmosphere

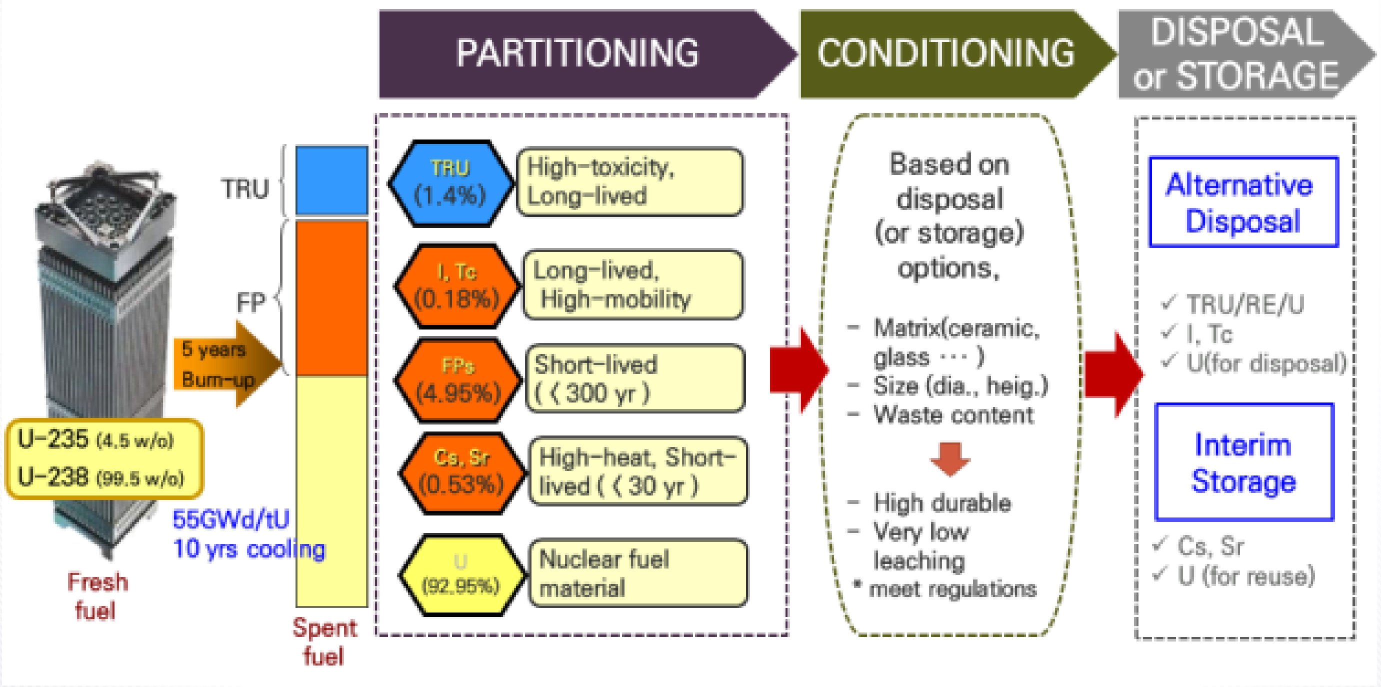
Ju Ho Lee, Yung Zun Cho, Chang Hwa Lee

Korea Atomic Energy Research Institute

KRS fall meeting, Changwon
2021.10.20-22

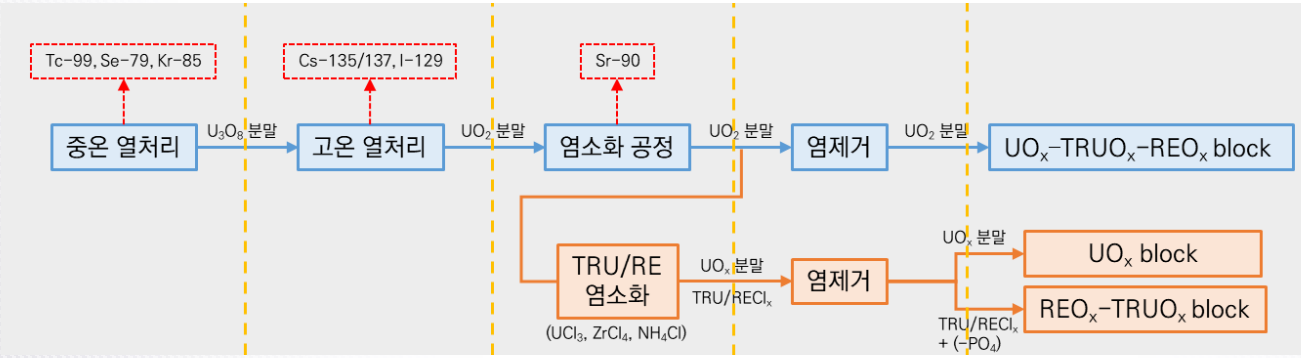
01 Research Scope

- In KAERI, Nuclide management technology(2021–) is being developed for reduction of disposal area required for spent fuel management
- The technology focuses on the release of fission products in UO_2 spent fuel such as Tc, Se, Kr, Cs, I using thermal treatment at oxidization condition improving fission release

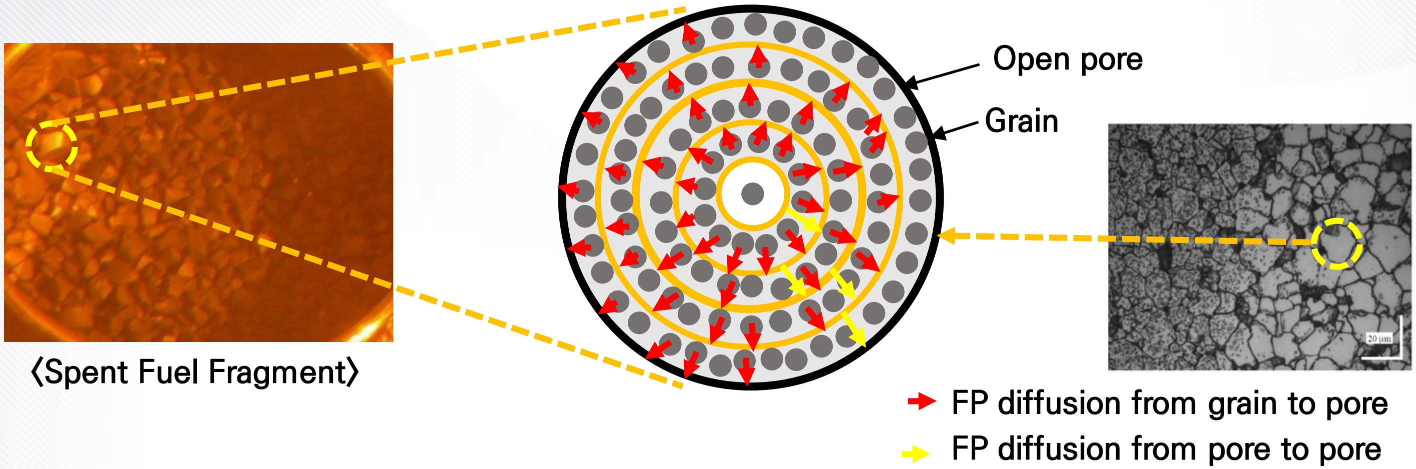


01 Research Scope

- However, the formation of $CsTcO_4$ should be avoided in thermal treatment since the compound is not easily volatilized leading to the reduced trapping efficiency of off-gas treatment process
- Therefore, thermal treatment should be divided into mid-temperature treatment mainly removing Tc, Se and Kr and high-temperature treatment removing Cs and I avoiding simultaneously release of Cs and Tc
- For the determination of such thermal treatment conditions, fission-release modeling is required, however, existing fission-release modeling cannot describe pulverization effect UO_2 ➔ U_3O_8 in oxidizing condition.
- Therefore, for the description of pulverization effect, fission-release modeling coupled to UO_2 oxidation model are developed and tested against Kr and Cs release data



02 Model Development



Model assumptions ::

- All fuel fragment are approximated to 1D sphere of mm size and grain to 1D sphere of μm size
- Fuel fragment is composed of equal shell representing grain and open pore at which fission products transport
- Fission products follows two stage diffusion, diffusion in solid spherical grain followed by diffusion in open pore
- UO_2 oxidation and fission product release separately occurs

03 Calculation Procedure

Gas composition(Inert, O_2 or Air)
Size of fuel fragment and grain
Heat treatment program

Input Conditions

UO_2 oxidation model

- Evaluation of average grain conversion from UO_2 to U_3O_8 and oxygen concentrations

FP Diffusion model

- Evaluation of FP concentration in grain/open pores and cumulative FP release amount through Finite-Volume method

04 Governing equations

UO_2 oxidation model

O_2 diffusion

$$\frac{\partial(\varepsilon_i C^*_{i,O_2})}{\partial t} = \frac{1}{r_i^2}\frac{\partial}{\partial r_i}\left(r_i^2 \varepsilon_i^0 D_p \frac{\partial C^*_{i,O_2}}{\partial r_i}\right) - (1-\varepsilon_i)k(T)(1-X_i)C^*_{i,O_2}\theta$$

UO_2 conversion

$$\frac{\partial(X_i)}{\partial t} = -3(1-\varepsilon_i)k(T)(1-X_i)C^*_{i,O_2}\theta$$

FP Diffusion model

FP diffusion between open pores

$$\frac{\partial(\varepsilon_i C^*_{i,FP})}{\partial t} = \frac{1}{R_i^2}\frac{\partial}{\partial R_i}\left(R_i^2 \varepsilon_i^0 D_P \frac{\partial C^*_{i,FP}}{\partial R_i}\right) + (1-\varepsilon_i)\frac{3}{a}\left(-D_s \frac{\partial C^{S*}_{i,FP}}{\partial r}\right)_{r=a}$$

FP diffusion from grain to open pores

$$\frac{\partial(C^{S*}_{i,FP})}{\partial t} = \frac{1}{r_i^2}\frac{\partial}{\partial r_i}\left(r_i^2 D_s \frac{\partial C^{S*}_{i,FP}}{\partial R_i}\right)$$

$$D_P = 0.001884\sqrt{\frac{2(RT)^3}{\pi}\left(\frac{1}{M_1}+\frac{1}{M_2}\right)}\frac{1}{N_A P \sigma_{12}^2 F}$$

$$D_S = A_{FP} exp\left(-\frac{B_{FP}}{RT}\right)(1 + CX + Dp_{O_2})$$

Solution strategy : Finite Volume method (FVM)

05 Results – Tested systems

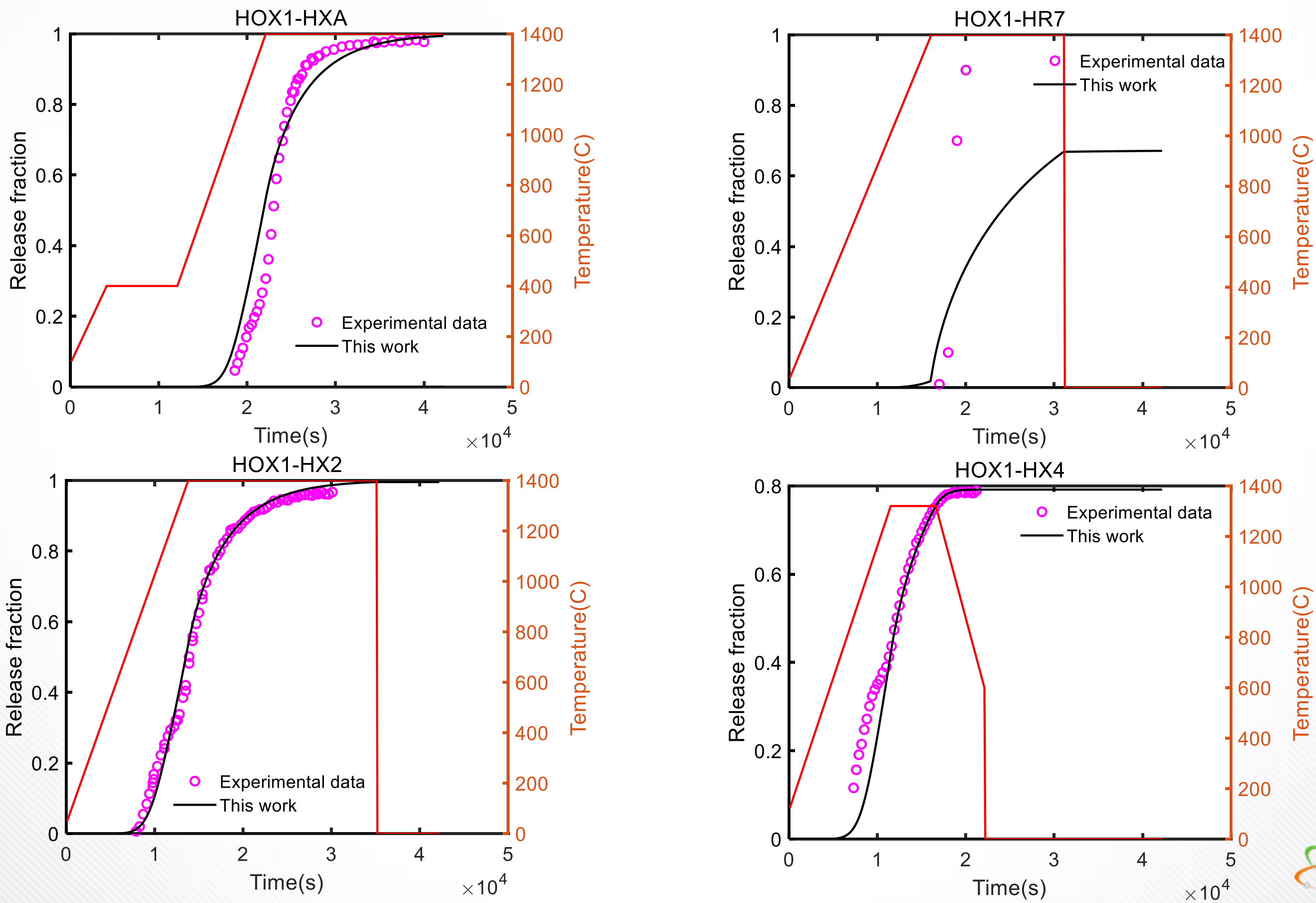
- Cummulative FP release data of AECL employed (DUPIC-AR-FT-06)

Test	Previous Treatment (starting material)	Tested condition
HOX1-HX1	None (UO_2 fuel fragment)	20℃ → 440℃(air)
HOX1-HX2	HX1 (U_3O_8 powder)	50℃ →1320℃(air)
HOX1-HX4	HX1 (U_3O_8 powder)	125℃ →1400℃(air)
HOX1-HXA	None (UO_2 fuel fragment)	20℃ → 400℃(Ar/4%H_2) → 400℃(air) →1400℃(air)
HOX1-HR7	None (UO_2 fuel fragment)	40℃ → 1400℃ (air)
HOX1-AR1	None (UO_2 fuel fragment)	40℃ → 400℃ (air) → 1400℃ (air)
HOX1-AR2	None (UO_2 fuel fragment)	240℃ → 400℃ (air) → 1400℃ (air)

05 Results – Low Burn-up Fuel

- LWR fuel, 2.55 % enriched U-235, 28 Mwd/kgU
- FP dependent Two parameters fitted to experimental release data

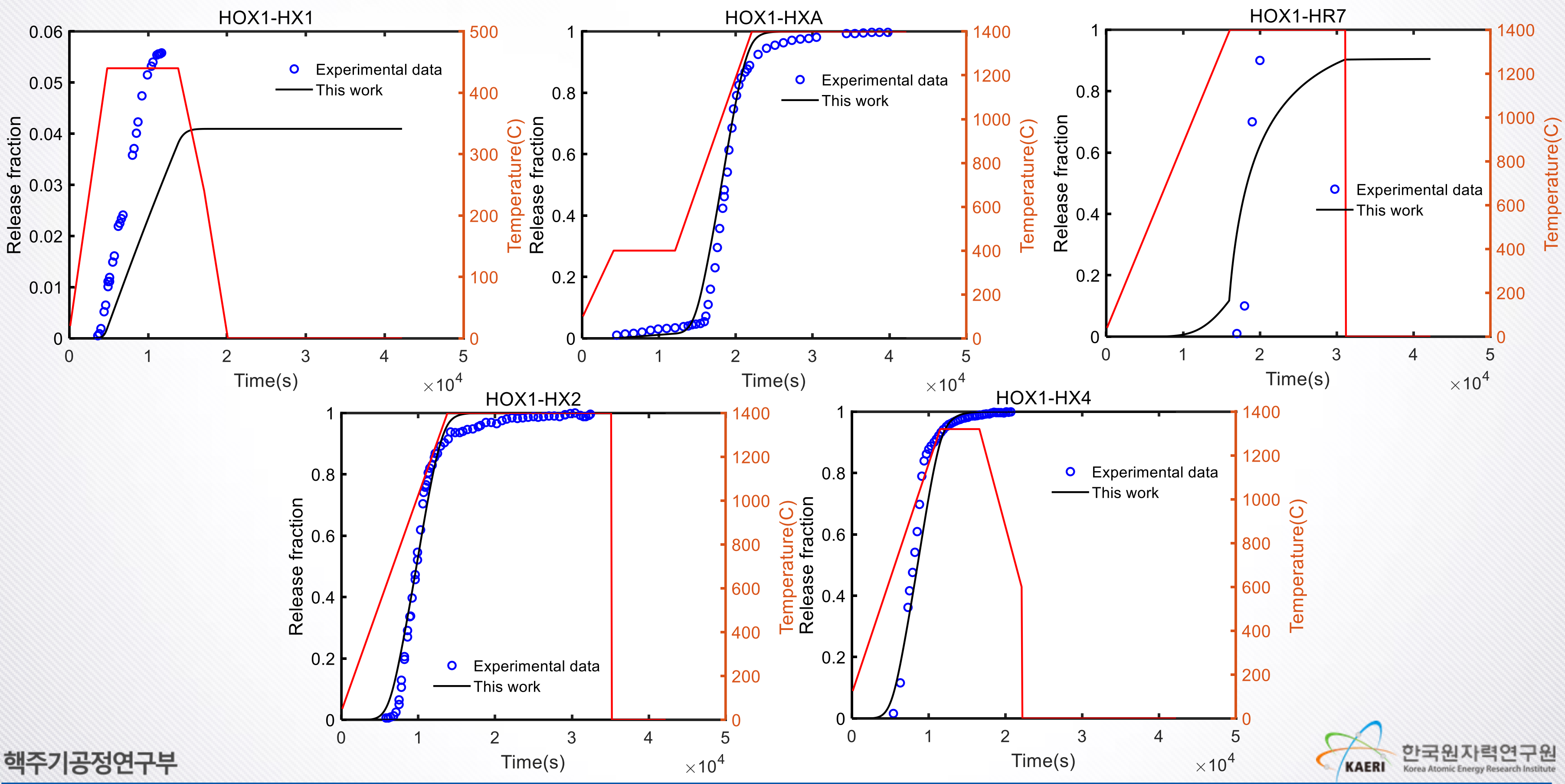
〈 Cesium 〉



05 Results – Low Burn-up Fuel

- LWR fuel, 2.55 % enriched U-235, 28 Mwd/kgU
- FP dependent Two parameters fitted to experimental release data

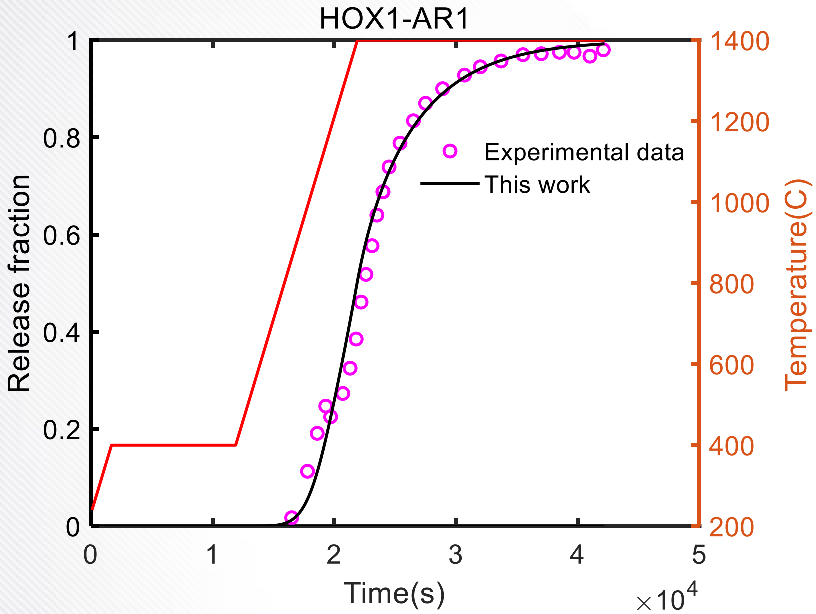
〈 Krypton 〉

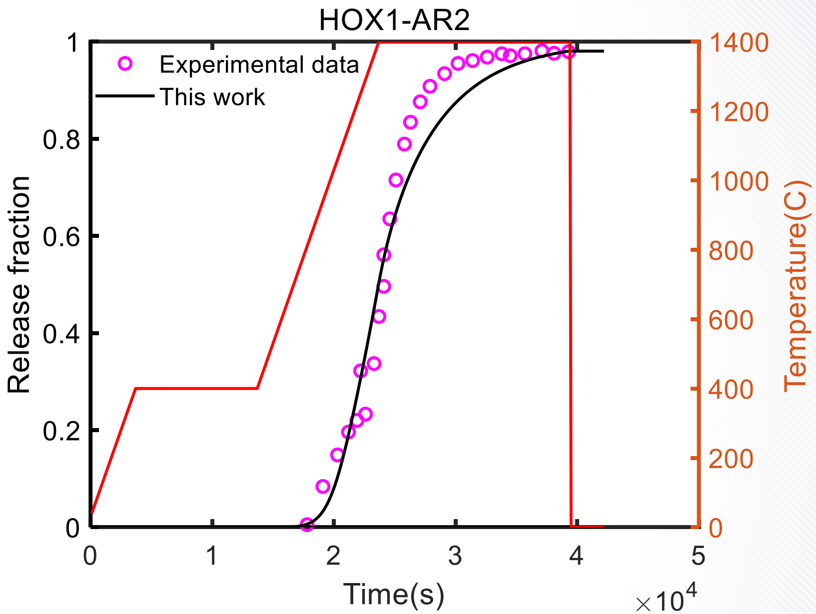


06 Results – High Burn-up Fuel

- LWR fuel, 3.5 % enriched U-235, 58 Mwd/kgU

〈 Cesium 〉

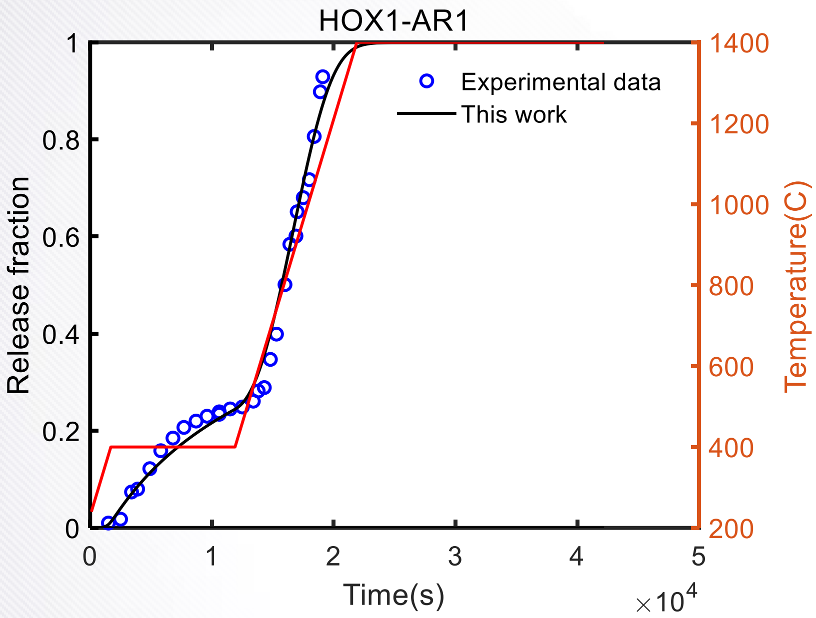


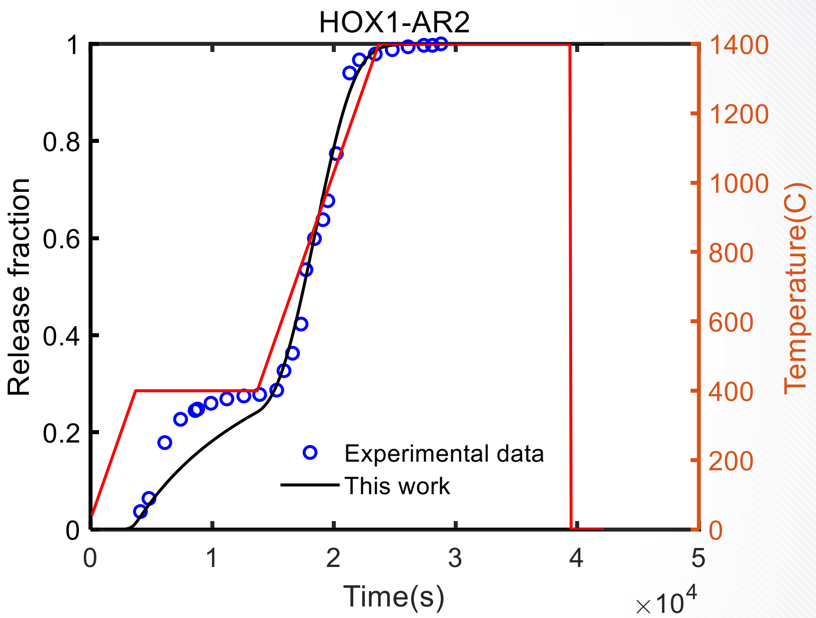


06 Results – High Burn-up Fuel

- LWR fuel, 3.5 % enriched U-235, 58 Mwd/kgU

〈 Krypton 〉





07 Conclusion

- Two-stage diffusion release model coupled to UO_2 oxidation model was developed accounting for the effect of oxygen content and pulverization effect

- A good agreement between experimental data and calculated fission release data : Close agreement in high burn-up fuel and reasonable results for low-burn-up fuel was obtained

- The developed model is expected to estimate Cs and Krypton release behavior at given scenario, enabling the derivation of optimal thermal treatment condition for efficient fission product removal

- In subsequent study, the present modelling is being extended to the release modeling of Technetium and Iodine

THANK YOU