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MODELING VOLATILE FISSION PRODUCTS TRANSPORT INTO THE GERMINAL V2 FUEL PERFORMANCE CODE BY COUPLING TO THERMOCHEMICAL SOFTWARE

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Contents

1/ Context

- Integration of thermochemical component into the PLEIADES simulation platform
- Chemical state of fission products in FBR (U,Pu)O₂ fuel
- Specific interest for GERMINAL: JOG and FCCI description

2/ Thermochemistry:

- Tools: ANGE (AdvaNced Gibbs Energy minimizer) and OpenCalphad (OC)
- Models + databases: TBASE and TAF-ID

3/ Coupling ANGE / OpenCalphad to GERMINAL V2

- Calculation scheme
- JOG evolution with time /burnup
- Volatile FPs radial distribution
- Oxygen radial redistribution

4/ Next steps

- Incorporation of the cladding material constituents Cr, Fe and Ni
- Oxygen quantity available for reacting into the gap

Context: the PLEIADES fuel simulation plateform – ceaden **the GERMINAL code**

R&D activities in support of ASTRID (French SFR prototype project)

- \rightarrow Need to update the fuel performance code for the design studies
	- With a validated modelling and calculation scheme for normal and off-normal conditions
- → Development of **GERMINAL V2** within **PLEIADES** simulation framework

PLEIADES: Federative framework for fuel performance codes Co-developed by CEA, EDF and FRAMATOME

- Multi-physic fuel coupling \Rightarrow taking into account the thermochemistry is needed in order to describe exp. observations performed during power transient
- Main goals:
- PWR: to simulate the cladding failure by SCI (Stress Induced Corrosion)
- SFR: to be representative of the chemical state of the FPs in our description of the JOG formation and of the FCCI

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General presentation of GERMINAL

1D½ calculation scheme

3 OCTOBRE 2018

- → Geometry assimilated to a **revolution cylinder**
- Fuel pin decomposed into **axial slices** Thermal evolutions linked by heat removal by coolant
- → **Radial resolution** of coupled physical processes **in each axial slice**

Thermal analysis, mechanics, fuel physics & chemistry

Special features of SFR fuel pins

- **High temperature conditions:**
	- \approx 2200°C at pellet center, 1000°C at periphery High temperature gradients : \approx 5000 $^{\circ}$ C / cm
- Radial migration of elements (O, U, Pu)
- Radial migration of porosity (central hole formation)
- **Preponderant gas release (80-90% at end-of-life)**
- \blacksquare Volatile FPs release \rightarrow "Joint Oxyde-Gaine" formation
- **Steel material for cladding ; important damage & swelling**

PHENIX pellet, initially solid Final burn-up : 13,3 at%

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FBR fuel behaviour

Very high thermal level \Rightarrow enhanced phenomena

Current description (into GERMINAL V2) : П

 \rightarrow Relatively empirical, deduced from qualitative PIE performed into the Phénix Reactor (in the 80's)

 \Leftrightarrow based on the similarity between the cesium and the (stable) fission gases retention

\blacksquare For the JOG : JOG = porous $\mathsf{Cs}_2\mathsf{MoO}_4$

- \checkmark λ (JOG) = f [λ (Cs₂MoO₄), p(JOG)] for low porosity
- \checkmark = f [λ (Cs₂MoO₄), λ (gas),p(JOG)] for high porosity

The ROG (FCCI) :

- $\frac{\partial e_{corr}}{\partial \tau}$ = +10µm/at% undeformed cladding
- $\frac{\partial e_{corr}}{\partial \tau}$ = +15µm/at% deformed cladding

Limitations:

- Real chemical composition of the JOG, more complex, is not considered
- Thermodynamic of the fuel is not considered (oxygen chemical potential - T°, p(O₂), FPs activities, presence of liquid phases)

 $\epsilon \in$ Goal \Rightarrow to develop a description of the JOG ROG on physical basis (namely thermodynamic + transport)

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INTERNATION CONTROLLER CONTROLLER CONTROLLER CONTROLLER CONTROLLER CONTROLLER CONTROLLER CONTROLLER CONTROLLER IA IIIA IIIA III. Waxaa kale dheer iyo dhamaan iyo dhamaan iyo dhamaan iyo dhamaan iyo dhamaan iyo dhamaan iyo **IPAN III Chemical state of fission products in** H **He** Li Be B C N O F Ne Na Mg IIIB IVB VB VIB VIIB VIII VIII VIII IB IIB Al Si P S Cl Ar

\checkmark Each fission creates 2 FPs: **Rb Sr Y Zr Nb Mo Tc Ru Rh Pd Ag Cd In Sn Sb Te I Xe Ce Pr Nd Pm Sm Eu Gd** Tb Dy Ho Er Tm Yb Lu $P(U)$ Dividence $Z + E_0$.

H **He**

Na Mg IIIB IVB VB VIB VIIB VIII VIII VIII IB IIB Al Si P S Cl Ar

⇒ Modification of physical and chemical properties of the irradiated material: thermal conductivity, creep, grain size, fission gas release...

 The chemical state of the fuel depends strongly of the oxygen chemical potential of the solid solution $(U_{1-y}Pu_y)O_{2-x}$:

 $\Delta \overline{GO_2}(x, y, T) = \mu(O_2) = RT Ln (p_{O_2}/\frac{1}{O_2}) \Big| \frac{(8.314 \text{J/mol/K})}{T \cdot \text{Temperature}}$

 $2'02'$ \rightarrow T. Temper *R: Perfect Gases Constant (8.314J/mol/K) T: Temperature (K) P(O²): oxygen partial pressure (bar) P°(O²): standard pressure (1 bar)*

Fission products creation in a Phénix fuel pin at 10 %FIMA

Thermochemical components available in PLEIADES: Raden **ANGE and OC* (OpenCalphad)**

Open source thermodynamic softwares:

- ANGE : AdvaNced Gibbs Energy minimizer, SOLGASMIX based code which has been implemented into the PLEIADES fuel simulation platform (partnership CEA and EDF)
- OC: OpenCalphad, developed by Bo Sundman (formerly KTH INSTN), one of the first developer of the Thermo-Calc software

Principle:

 \rightarrow Calculation at fixed T and P or V of the thermochemical equilibria of the different chemical constituents located in the different (solid, liquid and gaseous) phases, under the species balance constraint by **minimising the free enthalpy** (or *Gibbs free energy*) of the whole system.

Thermodynamic models for the mixture phases:

- Lindemer & Besmann *description* for the (U,Pu,PF)O_{2-x} solid solution in ANGE: neutral compounds with a given cation valency
- CEF (*Compound Energy Formalism*) with OC
	- \rightarrow CEF based on the CALPHAD method with several sub lattices (anionic, cationic, with vacancies)
	- \Rightarrow In order to reproduce the stoichiometry of the phases considered

Thermodynamic databases:

- \blacktriangleright \blacksquare (TBASE + SGTE) with ANGE
- Þ (TAF-ID) with OpenCalphad

** B. Sundmann, U.R. Kattner, M. Palumbo and S.G. Fries "OpenCalphad - a free thermodynamic software" Integrating Materials and Manufacturing Innovation, 4:1 (2015)*

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Comparison of the thermodynamic databases

ANGE

<u></u>* TBASE + Lindemer & Besmann's model

- Database from the 90's with regular thermodynamic functions updating
- 24 elements of the periodic table
- Lindemer & Besmann's description (associated species model) for the mixed oxide
- \rightarrow (sub-)regular solution of:

 $(U,Pu,Am,Ln)O_{2+x} =$ **(U1/3, UO² , U2O4.5, U3O⁷ , PuO² , Pu4/3O2 …)**

- *Stoichiometric compounds (100)*
- *Metallic precipitates description quite simplist: only one phase of « noble metals »*
	- \rightarrow Quite simple models that do not describe the non-stoichiometry of the phases as well as the liquid solutions

TAFID_V8 (beginning 2018)

*^{**} CEF + Thermo-Calc or OC (Open Calphad)*

- Database developped and maintained in the frame of the TAF-ID project (OECD/NEA)
- 41 elements of the periodic table
- Model CEF *sub-lattice description in relation with crystalline structure for the mixte oxide*
- \rightarrow non ideal solution of:

 $(U,Pu,Am,Ln)O_{2+v} =$ $(U^{+3}, U^{+4}, U^{+5}, Pu^{+3}, Pu^{+4}, Am^{+3}, Am^{+4}, Ln^{+3})$ ₁(O⁻²,Va)₂(O⁻²,Va)₁

- *Phases stoichiometry range (150) described*
- *Description of the mixture liquid phases, including the various metallic precipitated phases*
	- **→ Complete** model describing the non stoichiometry of the phases as well as the formation of liquid solutions
- More phases as well as more sophisticated models implemented into the TAF-ID
	- \rightarrow More representative of the phase transitions and of cristallographic defects
		- \rightarrow but still in test and validation state

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CCA OEN Coupling GERMINAL with a thermochemistry component

- **Thermodynamic equilibrium calculation in the** fuel pellet after local convergence loop
	- \Leftrightarrow System with 16 elements:

He, U, Pu, O, Cs, Te, I, Mo, Ba,

Zr, Ce, La, Gd, Eu, Ru, Pd

- \rightarrow Quantities of gaseous and liquid volatile fission products phases released into the gap
- **Fission products release: 4 options**
- **Options 1 and 2**: Total release of VFPs gaseous / (gaseous + liquid) phases formed into a radial mesh directly into the fuel-toclad gap
- **Options 3 and 4**: Release of VPFs gaseous/ (gaseous + liquid) phases homothetic to stable gases release
- **Thermodynamic equilibrium calculation in the** $gap \Leftrightarrow System$ with 6 or 7 elements :

Cs, Te, I, Mo, Ba, Zr, Pd, O

 \rightarrow Nature and quantities of solid and liquid phases contributing to the JOG formation

Difficulties

- Convergence of thermodynamic equilibrium
- **Calculation times**

Ceaden **Résultats GV2-ANGE / OpenCalphad : Composition chimique de la pastille combustible**

- Chemical composition of the JOG calculated at T°(gap) :
	- **With ANGE**: $Cs + Cs + Cs$ = calculated in the gap in irradiation conditions
	- **With OpenCalphad :** Mo + BaO + liquid (Cs,I,Te,O,Mo)
- Chemical composition of the JOG calculated at room $T^{\circ} \Leftrightarrow$ PIE conditions
	- **With ANGE :** phases identical to those obtained at T°(irr) ;

low $Cs₂MoO₄$ content

With OpenCalphad : $Mo + Cs + CsI + Cs₂Te + BaO + Cs₂MoO₄$

Nestor-3 : 13,3 %FIMA

Evolution versus burnup of the ratio of volatile FPS release between OC and ANGE:

- \rightarrow Similar values for Cs, Te and I
- \rightarrow Ba and Mo are not predicted similarly with GERMINAL + ANGE vs GERMINAL + OC: thermodynamic database specificities

Results GERMINAL V2 - ANGE: Evolution of JOG width (µm) versus time

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 $BOITX1 - 7,0$ %FIMA - e(JOG) = $f(t)$

- **Scheme operationnal with ANGE**

- **With OC: calculation is performed a posteriori in autonomous mode from the gap inventory**

RESULTS GERMINAL V2-ANGE / OC : JOG WIDTH VERSUS BURN-UP (1)

Final JOG widths calculated at $T = 298.15K$ (room temperature \Leftrightarrow PIE) at the PPN (Peak Power Node = maximum flux) for following experiments:

HADIX-1 , BOITIX-1, COUCOU-1, SPHINX-1, NESTOR-3 : | 3,8 %FIMA | 7,0 %FIMA | 9,0 %FIMA | 11,2 %FIMA | 13,3 %FIMA |

Case 100% VFP (volatile fission products) release

JOG width versus final Burn-up: Case 100% volatile fission products release

RESULTS GERMINAL V2-ANGE / OC : JOG WIDTH VERSUS BURN-UP (2)

Final JOG widths calculated at $T = 298.15K$ (room temperature \Leftrightarrow PIE) at the PPN (Peak Power Node = maximum flux) for following experiments:

HADIX-1 , BOITIX-1, COUCOU-1, SPHINX-1, NESTOR-3 : | 3,8 %FIMA | 7,0 %FIMA | 9,0 %FIMA | 11,2 %FIMA | 13,3 %FIMA |

Case: VFP release ∞ FG (fission gas) release

CALCULATION NESTOR-3 GV2-ANGE / OC: JOG WIDTH VERSUS AXIAL POSITION

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Comparison GERMINAL V2 - ANGE /OpenCalphad: ceaden **Radial distribution of Volatile Fission Products**

NESTOR-3 - 13.3 %FIMA : Radial distribution of cesium at PPN and at eol calculated by GERMINAL + ANGE - Case VFP release \propto FG release

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NESTOR-3 - 13.3 %FIMA : Radial distribution of cesium at PPN and at eol calculated by GERMINAL + ANGE - Case VFP release \propto FG release

Distance from periphery (um)

NESTOR-3 - 13.3 %FIMA : Radial distribution of iodine at PPN and at eol calculated by GERMINAL + ANGE - Case VFP release \propto FG release

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Calculation GERMINAL V2 - OpenCalphad: Cesium radial distribution

NESTOR-3 - 13.3 %FIMA : Radial distribution of cesium at PPN and at eol calculated by GERMINAL + ANGE - Case VFP release \propto FG release

Coucou1 - 9.0 %FIMA - Cesium radial evolution

 Results seem to be more consistent when we consider only partial (an not total) release of VPF = volatile fission products

O/M ratio evolution with burnup: Results GERMINAL V2 - ANGE

 \Rightarrow Local O/M ratio in the fuel pellet:

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Difference between Germinal law and thermochemistry evaluation with ANGE irradiation goes up with time..

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Conclusion - Outlooks

Inventory JOG-ROG:

Taking into account palladium (Pd) as volatile FP in the calculation procedure: \rightarrow can be found in metallic inclusions with Te (and Ag) on liquid state from the low temperatures (\sim 700 K)

- taking into account constituents of the cladding material Fe, Cr and Ni:
	- \rightarrow chemical composition of ROG phases to come
	- \rightarrow volatile FPs release law: description more phenomenological to develop

Oxygen key-role:

Coupling with the oxygen radial redistribution model:

 \rightarrow taking into account the average $(\overline{O/M})_{\text{slice}}$ ratio given by the thermochemistry as an input for the OXIRED model in order to have the radial redistributed $(\overline{O/M})_{local}$ ratio

to introduce into the coupling with OpenCalphad

Work /study on the oxygen available into the gap:

- \rightarrow Influence of the available oxygen on chemical equilibria:
- oxygen in excess in the fuel-to-clad gap ?
- oxygen in excess on the fuel pellet surface ?
- Taking into account the oxygen activity instead of the oxygen quantity?

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Complementary slides

Current thermodynamic description considered in ANGE

Thermochemical component ANGE (*AdvaNced Gibbs Energy minimizer***)**

SAGE (*SOLGASMIX Advanced Gibbs Energy***)**

Thermodynamic database + models:

PWR database:

22 elements 61 gaseous compounds 80 stoichiometric compounds 3 compounds for metallic precipitates 19 species in solid solution $(U,Pu,L^*)O_{2-x}$ $(L^* =$ Lanthanides)

SFR database:

24 elements 90 gaseous compounds 100 stoichiometric compounds 5 compounds for metallic precipitates 27 species in solid solution $(U,Pu,L^*)O_{2-x}$

K.

FPs in solution modeling:

 \Rightarrow Lindemer & Besmann (L&B) description:

- **<U1-yPuyO2-x> irradiated** = **(U,Pu,L*)O2-x** : avec L* = Gd, La, Nd.. mixture of chemical species PuO₂, Pu_{4/3}O₂, UO₂ et U₂O_{4.5} (or U₃O₇), UL*₂O₆ and L*_{4/3}O₂ representative of the valency state
- Description semi-empirical not representative of cristallographic defects

LA RECHERCHE À L'INDUSTRIE **TAF-ID Project (OECD/NEA) :** Ceaden *Thermodynamic Advanced Fuels-International Database*

Models

TAF-ID V8 database (2018) :

TAF-ID: Thermodynamics of Advanced Fuels - International Database

Introduction Models

Flements

Assessed binary

Assessed ternan

Higher order sys

Systems with Ag

B, Ba, C, Ca, C

Fe, Gd, H, He,

Mo, N, Nb, Nd,

Pd, Pu, Re, Rh

Ta, Tc, Te, Th W, Zr **Periodic table**

41 elements :

- **200 binary systems**
- **75 ternary systems**

Introduction of models for binary and ternary systems

Elements

Periodic table

Home

Assessed binary systems Assessed ternary systems

Introduction

Systems with Ag, Al, Am, Ar, B, Ba, C, Ca, Ce, Cr, Cs, Fe, Gd, H, He, I, La, Mg, Mo, N, Nb, Nd, Ni, Np, O, Pd. Pu. Re. Rh. Ru. Si. Sr. Ta, Tc, Te, Th, Ti, U, V, W, Zr

TAF-ID: Thermodynamics of Advanced Fuels - International Database

Introduction Thermodynamic models Phases described usual name database namprototype StrukturBericht table **Assessed systems**

Home

elements binary system: ternary systems periodic table

Introduction Models Phases Systems To support the development of Generation 4 reactors (SFR, SCWR, GFR, LFR, MSR, VHTR) and to contribute to lifetime extension, safety improvement and safety analysis for Generation 2 & 3 systems (PWR, BWR, PHWR), there is a need to make available a comprehensive, internationally recognized, and quality-assured thermodynamic database. For this reason, a joint Project between 9 organizations representing 6 member states coordinated by the OECD-Nuclear Energy Agency (NEA) was started in 2013 with an initial 3 years period. The objective of the project titled Thermodynamics of Advanced Fuels - International database (TAF-ID) is to develop a thermodynamic database using the Calphad method to perform thermodynamic calculations on different types of fuels (oxide, metallic, nitride, carbide) including minor actinides (Am, Np), fission products (Cs, I, Ba, Sr, Mo, Zr, lanthanides, metallic fission products) and structural materials (steel, Zr alloy, B4C, SiC, concrete). Thermodynamic properties of fuels versus temperature and composition (with fission products and minor actinides) will be provided. The inclusion of structural materials will allow the prediction of fuel/cladding chemical interactions under normal and off-normal conditions. The database will be generated and regularly updated by merging existing and developing databases from the various participating organizations. The database will be available in both Thermo-Calc and FACTSAGE usable formats. Members of the Programme Review Group

Consultant: N. Dupin, Calcul Thermodynamique, France

Coordinator: OECD-NEA (S. Massara)

Remarks and comments on this decumentation and on the thermodynamic database are welcome. We thank you to ask for the autorization
In case where you would like to communicate this documentation and/or the database to any o documentation cannot be modified. This documentation uses javascript, You must allow it in order to get it work properly. Current version: V6 2015 September 30th

TAF-ID: Thermodynamics of Advanced Fuels - International Database **Phases**

Systems

Thus over 10660 ternary systems. 72 systems are assessed in the current database.

PAGE 20