THE OECD-NEA THERMODYNAMICS OF ADVANCED FUELS – INTERNATIONAL DATABASE (TAF-ID) PROJECT

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1. Context and aim of the project
2. TAF-ID project
3. Calphad method
4. Existing databases
5. Thermochemical models
6. Progress of TAF-ID database development
7. Example of comparison of existing descriptions
8. Documentation
9. Conclusion

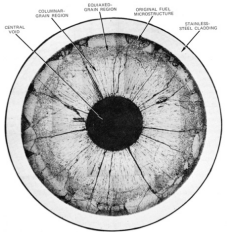
CONTEXT AND AIM OF THE TAF-ID PROJECT

AIM

- To develop a **thermodynamic database** as a **computational tool** to perform thermodynamic calculations on advanced fuel materials using the Calphad method
  ➡ **Phase diagrams + Thermodynamic properties of the phases**
- To **exchange** on the models, review of experimental data, softwares, assessments ….

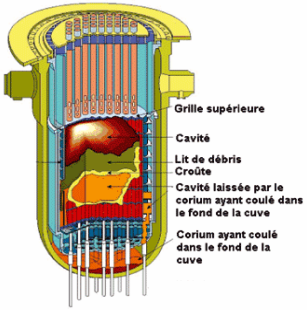
CONTEXT ➡ Fuel materials for Generation 2,3 & 4 reactors

- **Fuels**
  - UO_2, $(U,Pu,Am,Np)O_2$, $(U,Th)O_2$, (U,Pu,Zr,Am,Np), UN, (U,Pu)C
- **Fission products**
  - Ba, Sr, Mo, Zr, Lanthanides (Ce, La, Nd, Gd), metallic FPs (Pd, Ru, Rh, Te), Volatile (Cs, I, Te)
- **Structural materials**
  - Fe-Cr-Ni, Zr alloys, Fe-Cr-Al-Y, Concrete (SiO_2-CaO-Fe_xO_y-Al_2O_3-MgO), SiC, B_4C



APPLICATIONS

- **Fuel behaviour at high temperature under normal and off-normal conditions**
  - Influence of the **Minor Actinides** on the fuel thermodynamic properties
  - **Fission product** « chemistry »
  - **Solid/liquid** phase transition
  - **Vaporization**
- **Fuel / cladding chemical interaction at high temperature**
- **Fuel fabrication**



TAF-ID PROJECT

2013-2016: Thermodynamics of Advanced Fuels – International Database

➡ **The aim is to make available a comprehensive, internationally recognised and quality-assured thermodynamic database for advanced fuels**

- Coordinated by S. Massara from OECD/NEA➡ http://www.oecd-nea.org/science/taf-id

		Programme Review Group	
Country	**Signatory**	**Representative**	**Alternate Member**
Canada	AECL, RMCC, UOIT	D. Barber	E.C. Corcoran
France	CEA	C. Guéneau (Chair)	J.-C. Dumas
Japan	JAEA, CRIEPI	M. Kurata	T. Ogata
The Netherlands	NRG	R. Hania	G.-J. de Haas
Rep. Of Korea	KAERI	B.-O. Lee	J.-H. Kim
USA	DOE	T. Besmann (Vice-Ch)	P. Turchi

- The TAF-ID database is being built by **merging existing databases**
- N. Dupin (consultant) is responsible for the maintenance and documentation of the database

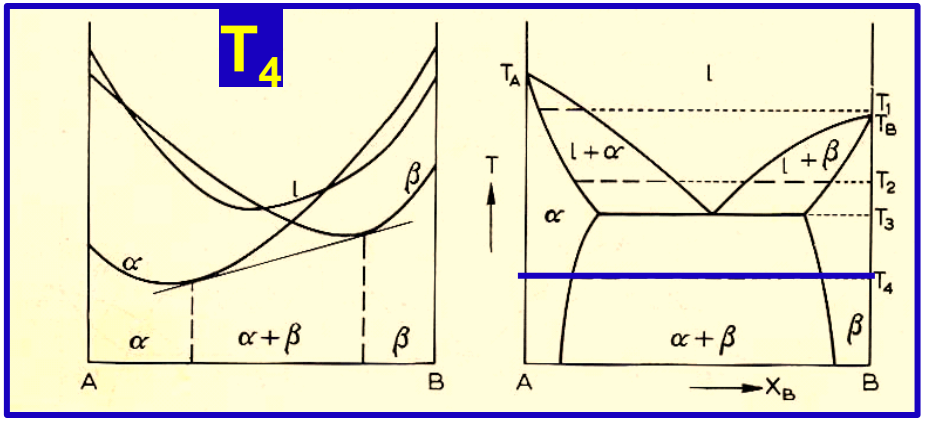
HOW TO CALCULATE THERMODYNAMIC EQUILIBRIUM ?
➡CALPHAD METHOD

- In multi-component systems, the thermodynamic equilibrium for given number of moles of components n_i, temperature T and pressure P is calculated by **minimization of the Gibbs energy** of the system

$$\min(G) = \min\left(\sum_{\alpha} m^{\alpha} G_m^{\alpha}(T, p, x_i^{\alpha} \text{ or } y_k^{(l,\alpha)})\right)$$

  - Softwares: Thermo-Calc, FACTSAGE, Pandat, Gemini, HSC Chemistry, OC …

- The Gibbs energy of the system G is a combination of the Gibbs energies of the phases G_m^{α}



$$G = \sum_{\alpha} m^{\alpha}.G_m^{\alpha}$$

$$x_i = \sum_{\alpha} m^{\alpha}.x_i^{\alpha}$$

- For a multi-component system ➡ Extrapolation from binary and ternary systems

➡ **It requires developement of databases containing the Gibbs energy functions for all the phases**

$$G_m^{\alpha}(T, p, x_i^{\alpha} \text{ or } y_k^{(l,\alpha)})$$

EXISTING DATABASES

Country	Software	Chemical system	Model
Canada AECL RMC UOIT	*FACTSAGE*	Fuel UO_2 + Np, Pu + Coolant H_2O FPs: (Kr), Rb, Sr, Y, Zr, Mo, Tc, Ru, Rh, Pd Te, I, Xe, Cs, Ba, La, Ce, Pr, Nd	302 comp. 17 solutions
France CEA	*Thermo-Calc*	Fuel UO_2, MOX+Am,Np,(U,Pu,Zr), (U,Pu)C, (U,Pu)N FPs: Ag, Ba, Cs, I, La, Mo, Nb, Ru, Sr, Te Materials: Concrete, B-C, Fe-Cr-Ni, SiC,Ta, W, V	Solutions 561 binaries 30 ternaries
Japan JAEA, CRIEPI	*Thermo-Calc*	Metallic fuel: Am-Cd-Ce-Fe-Gd-La-Nd-Np-Pr-Pu-U-Y-Zr Oxide fuel: Pu-Zr-O, Fe-B-C-O	Solutions
Netherlands NRG	*FACTSAGE*	Fuel: (U, Pu, Am, Np, Th)-(C, N, O) FPs: Ag, Ba, Br, Ce, Cs, Eu, I, La, Mo, Nb, Nd, Pd, Rb, Ru, Sb, Sn, Sr, Te, Y, Zr Materials: Fe, Ni, Si, Ti, Ta, W, Zn + Na, H, Pb	727 comp.
USA ORNL LLNL	*FACTSAGE* *Thermo-Calc*	UO_2 + RE: Pu-U-Gd-Ce-La-O Metallic fuel: Am-Pu, Mo-Pu,Mo-U,Nb-U,Nb-Zr,Pu-U,Ti-U,U-Zr Materials: Be-Cu-Fe-Nb-Ta-Ti-Zr	C1 solution solutions

- ➡ Different databases with different models.
- ➡ FACTSAGE and Thermo-Calc softwares are used.

THERMOCHEMICAL MODELS

- **The models are different from one database to another**

  - **Models with stoichiometric phases and a few solid solutions**

    TBASE+RMC+UOIT: many compounds are described

    ➡ Associate model for oxide solid solutions (A,AO_2,AO_3 …)

    ➡ The phase diagrams can not be calculated

    A solution model for (Mo,Pd,Rh,Ru,Tc)

  - **Model for MOX solid solution with Lanthanides**

    ORNL: to calculate oxygen potential versus burnup

    ➡ Ionic three sublattice model for MOX : Compatible with CEA database

    ➡ The phase diagrams can not be calculated

  - **Models with a full description of non stoichiometric phases** (solid & liquid)

    JAEA+CEA+LLNL ➡ The phase diagrams can be calculated

In case of no phase diagram data, the G functions of the compounds will be introduced.

The aims of our project are:
- to merge these different databases,
- to **agree on thermodynamic descriptions** for binary and ternary sub-systems
- to **propose experiments** to improve the descriptions

in order to provide a **consistent database.**

TAF-ID DATABASE

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

BINARY SYSTEMS

Ag-I Ag-Ti Ag-Zr
Al-Ca Al-Cr Al-Fe Al-Mg Al-O Al-Si
Am-Fe Am-Np Am-O* Am-Pu* Am-U Am-Zr
B-C B-Fe B-H B-I B-O B-Pu B-U
Ba-H Ba-I* Ba-La* Ba-Mo* Ba-N Ba-O Ba-Ti* Ba-V*
C-Cr C-Fe C-Mo C-N C-Nb C-Ni C-O C-Pu* C-Re C-Si C-Ta C-Ti C-U* C-V C-W C-Zr
Ca-Fe Ca-Mg Ca-O Ca-Si
Ce-O*
Cr-Cs* Cr-Fe Cr-H Cr-I Cr-La* Cr-Mo Cr-N Cr-Ni Cr-O Cr-Pu* Cr-Si Cr-Ti Cr-U* Cr-Zr
Cs-I* Cs-Mo* Cs-Nb* Cs-O Cs-Pu* Cs-Ta* Cs-Te* Cs-Ti* Cs-U* Cs-V* Cs-Zr*
Fe-Ni Fe-Np Fe-O Fe-Pu Fe-Si Fe-U Fe-Zr
H-I H-O H-Sr
I-Mo I-Sr+ I-Te
La-Mo* La-Nb* La-O La-Pu* La-Re* La-Ta* La-Te La-Ti* La-U* La-V* La-W*
Mg-O
Mo-N Mo-O Mo-Pd Mo-Pu* Mo-Re Mo-Rh* Mo-Ru Mo-Si Mo-Sr* Mo-Te* Mo-Ti Mo-U* Mo-Zr
N-O N-Pu* N-Si N-Ti N-U+ N-Zr
Nb-O Nb-Pu* Nb-Si Nb-U Nb-Zr
Nd-O* Nd-U*
Ni-O
Np-O* Np-Pu* Np-U Np-Zr
O-Pu* O-Ru O-Si O-Sr O-Te O-Th* O-Ti O-U O-Zr
Pd-Rh Pd-Ru* Pd-Te*
Pu-Re* Pu-Ru* Pu-Si* Pu-Ti* Pu-U+ Pu-W* Pu-Zr
Re-Si Re-U* Re-W
Rh-Ru* Rh-Te*
Ru-Te* Ru-U*
Si-Ta Si-Ti Si-U* Si-W Si-Zr
Sr-Ti* Sr-V*
Ta-U*
Ti-U* Ti-Zr
U-W* U-Zr

In red*: assessed within our project
In blue: coming from the literature

TERNARY SYSTEMS

Al-Ca-O Al-Cr-O Al-Fe-O
Al-Mg-O Al-O-Si
Am-O-Pu
B-C-Fe B-Pu-U
C-Mo-Re C-Mo-Si C-Mo-Ti
C-Mo-U C-N-Ti C-O-Pu C-O-U
C-Pu-U C-Pu-W C-Re-U
C-Re-W
C-Si-Ti C-Si-U C-U-W C-U-Zr
Ca-Fe-O Ca-Mg-O Ca-O-Si
Cr-Fe-O
Fe-O-Si
Mg-O-Si
Nd-O-U
Ni-O-Si
O-Pu-U O-Pu-Zr
Pu-U-Zr

TAF-ID DATABASE ➡ 2013 PROGRAMME OF WORK (1)

Application	Task	Work	Delivery
Metal Fuel **U-Pu-Zr-Am-Np-Fe, U-Ti**	1	Comparison of available descriptions (JAEA, CEA, LLNL)	Sept.
ThO_2 fuel **ThO_2**	2	Introduction of ThO_2 phase + gaseous species	Sept
In-vessel Corium			
U-Fe-O-Zr	3	Introduction + documentation	Dec.
Fe-B-C	4	Introduction + documentation	Sept.
Pu-Zr-O	5	Introduction + documentation	Sept.
U-Pu-B	6	Introduction + documentation	Sept.
Fission Products			
Ce-O	7	Introduction + documentation	Sept.
Nd-U-O	8	Introduction of binaries + documentation	Sept.
GdO_2	9	Introduction of C1 phase	Dec.
UO_2-La_2O_3	10	Biblio + description + introduction + documentation	Dec.
Pd-Rh-Ru-Te-Mo	11	Comparison of available descriptions (UOIT,CEA)	Sept.
Mo-Te, Te-O, Ru-O	12	Introduction + documentation	Sept.
Cs-Te	13	Introduction + documentation	Sept.
Cs-I	14	Introduction + documentation	Sept.
Cs-O	15	Introduction + documentation	Sept.

TAF-ID DATABASE ➡ 2013 PROGRAMME OF WORK (2)

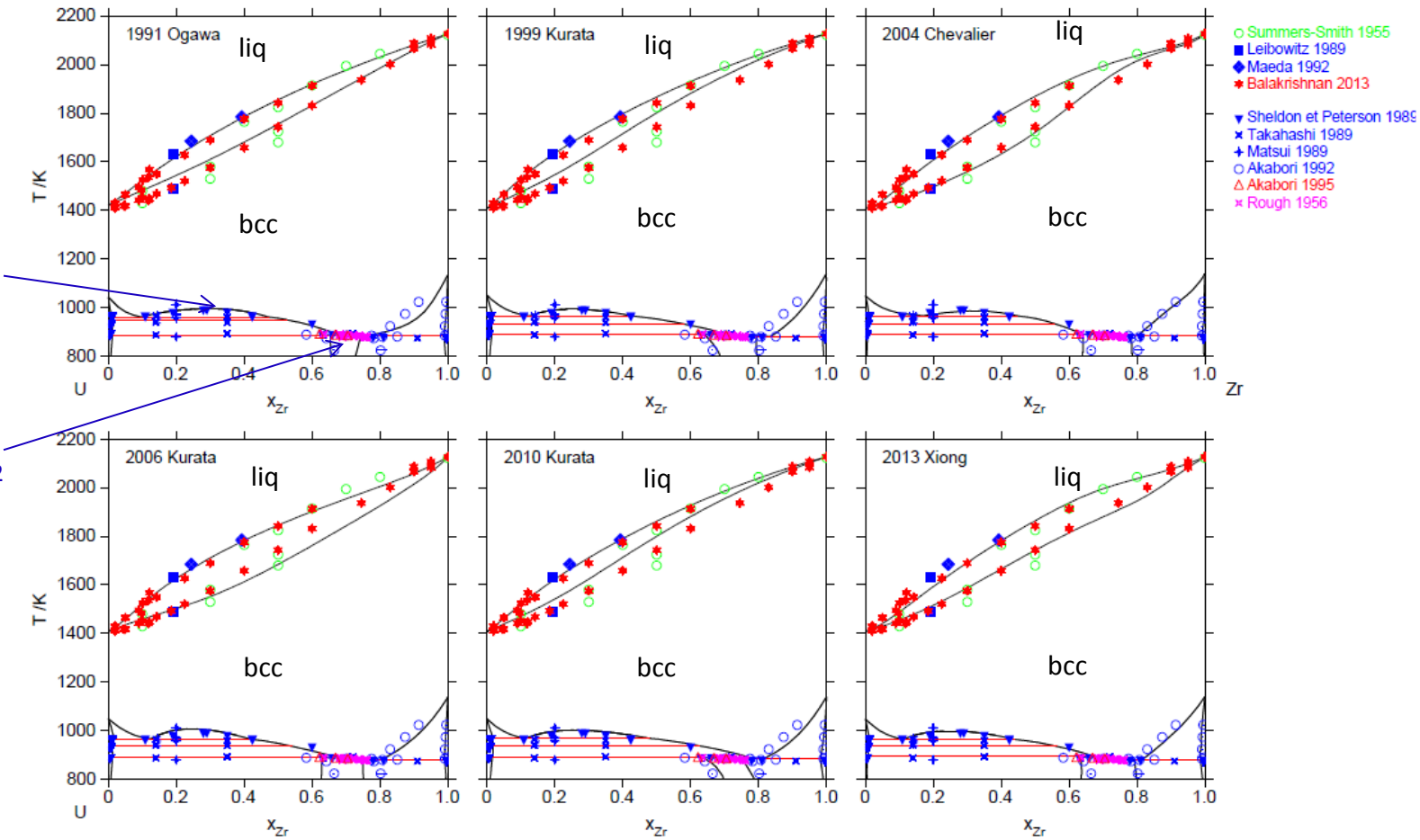
Application	Task	Work	Delivery
Corium Ex-vessel			
UO_2-SiO_2	16	Biblio + description + introduction + documentation	Dec.
UO_2-CaO	17	Biblio + description + introduction + documentation	Dec.
UO_2-Al_2O_3	18	Biblio + description + introduction + documentation	Dec.
UO_2-MgO	19	Biblio + description + introduction + documentation	Dec.
ZrO_2-SiO_2	20	Biblio + introduction + documentation	Dec.
ZrO_2-CaO	21	Biblio + introduction + documentation	Dec.
ZrO_2-Al_2O_3	22	Biblio + introduction + documentation	Dec.
ZrO_2-MgO	23	Biblio + introduction + documentation	Dec.

COMPARISON OF EXISTING DESCRIPTIONS
EXAMPLE: U-ZR SYSTEM (1)

- U-Zr is a key system to describe metallic fuels (U-Pu-Zr system) and chemical interaction between UO_2/Zr during severe accident (U-Zr-O system)
- 6 different assessments: Ogawa 1991, Kurata 1999, Chevalier 2004, Kurata 2006, Kurata 2010, Xiong 2013

Miscibility Gap in bcc

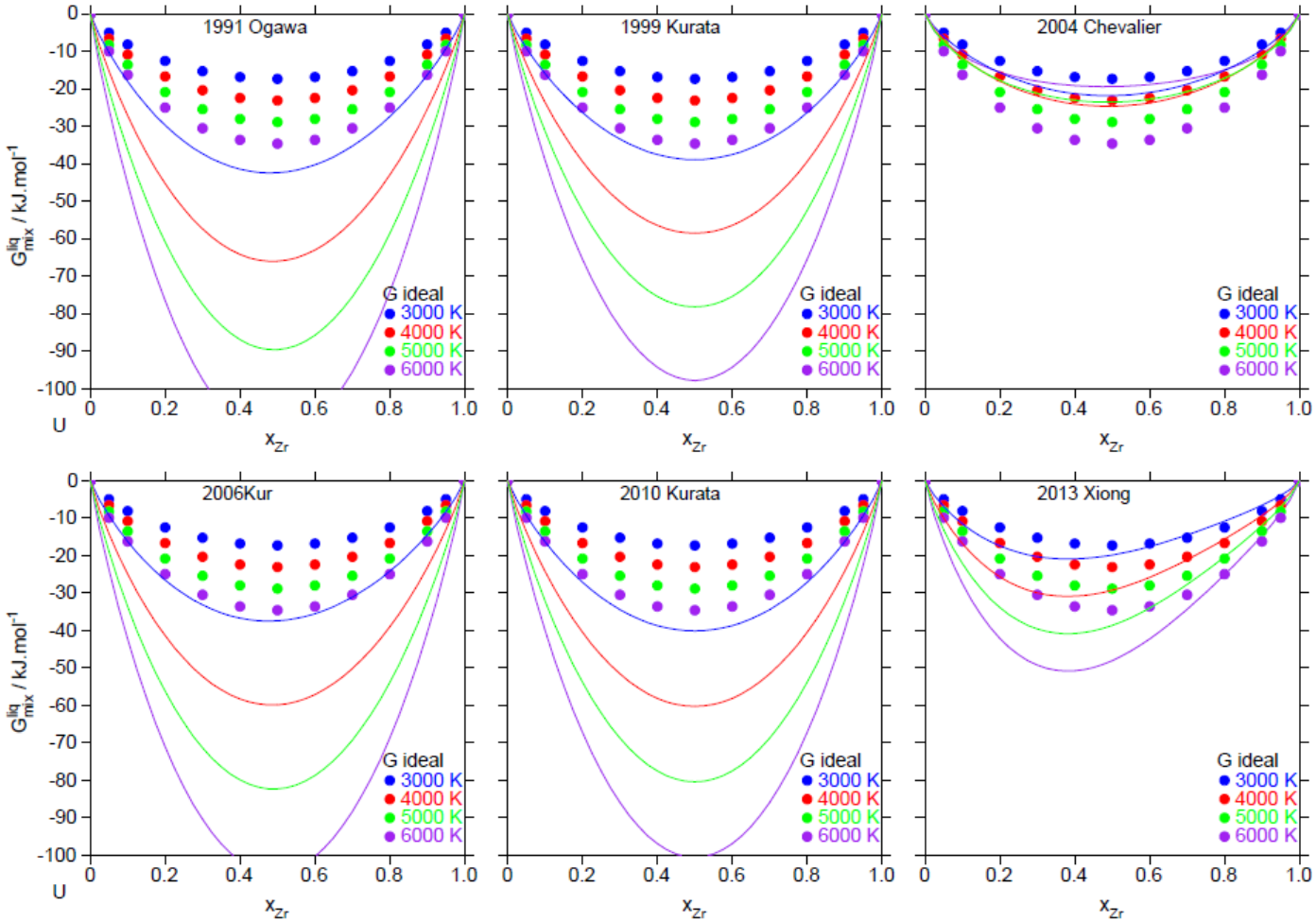
Delta-UZr_2



- ➡ The phase equilibria are not well established in the region close to delta-UZr_2 phase
- ➡ According to the liq/bcc phase boundaries, an ideal mixing is expected in the liquid.

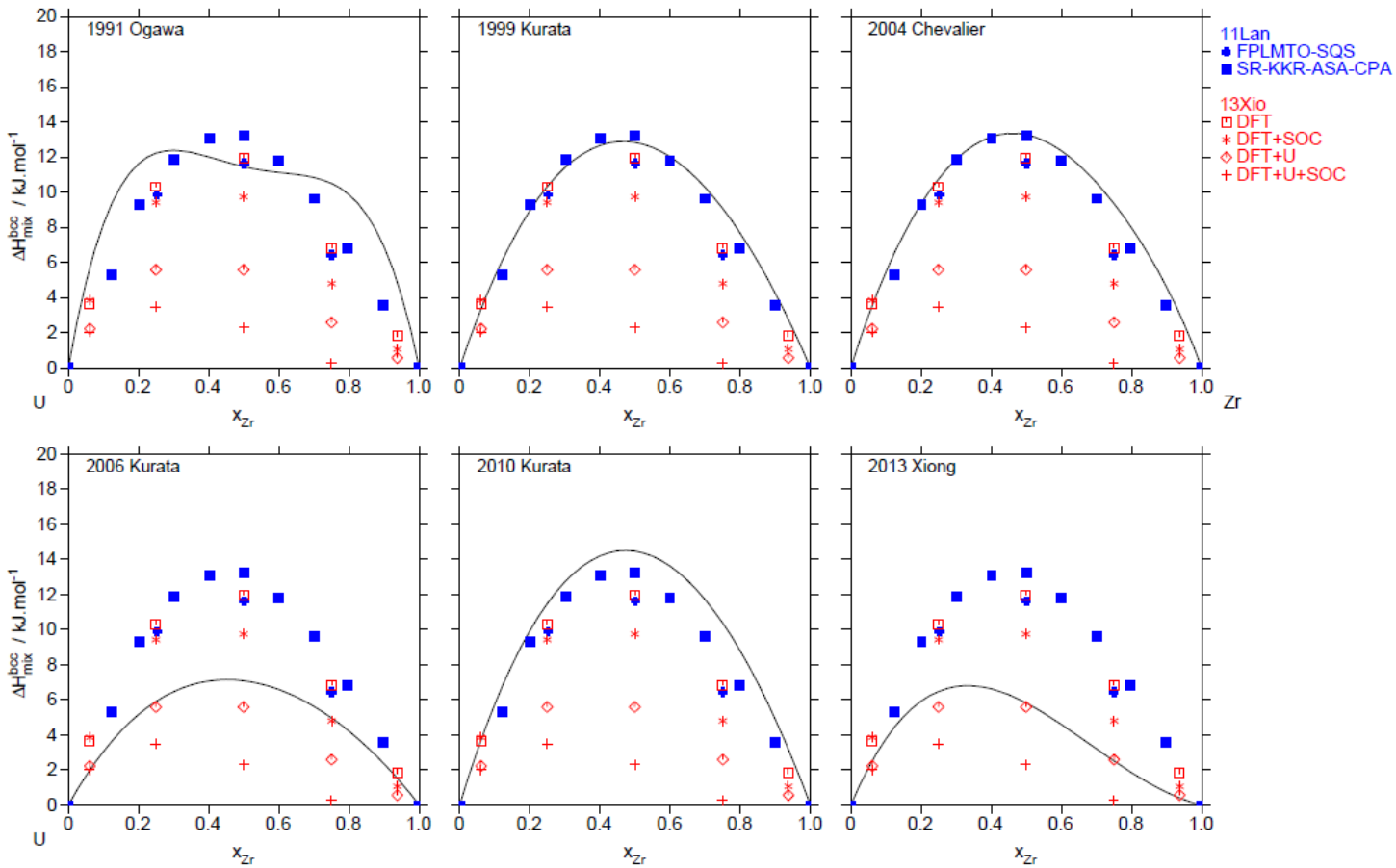
COMPARISON OF EXISTING DESCRIPTIONS
EXAMPLE: U-ZR SYSTEM (2)

Mixing in the liquid phase at high temperature



➡ Different extrapolations at high T whereas an ideal behaviour is expected

Mixing enthalpy in the bcc phase



➡ Two different datasets for mixing enthalpy in bcc solid solution from ab-initio

DOCUMENTATION (N. DUPIN)

TAF-ID : Thermodynamics of Advanced Fuels - International Database

Home | Introduction | Models | Phases | Systems

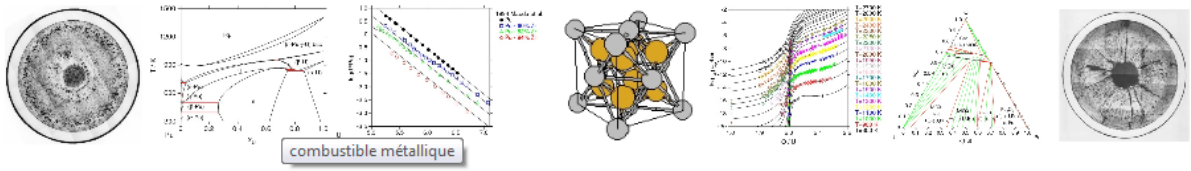
Introduction

Thermodynamic models

Phases described
- usual name
- database name
- prototype
- StrukturBericht
- table

Assessed systems
- elements
- binary systems
- ternary systems
- periodic table

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.



Participants:	Canada	AECL, RMCC, UOIT	D. Barber, M. Piro, E.C. Corcoran, M. Kaye
	France	CEA	C. Guéneau (Chair), J.C. Dumas, S. Gossé
	Japon	JAEA, CRIEPI	M. Kurata, T. Ogata
	Netherlands	NRG	R. Hania, G.-J. de Haas
	Rep. of Korea	KAERI	B.-O. Lee, J.-H. Kim
	USA	DOE	T. Besmann (Vice-Ch.), P. Turchi, R. Kennedy, M. Stan

Consultant N. Dupin, Calcul Thermodynamique, France

Coordinator: OECD-NEA (S. Massara)

Remarks and comments on this documentaion 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 other person. The database and the documentation cannot be modified. *This documentation uses javascript. You must allow it in order to get it work properly.*

Current version : 2013 September 16th

DOCUMENTATION (N. DUPIN)

TAF-ID : Thermodynamics of Advanced Fuels - International Database

Home | Introduction | Models | Phases | Systems

Phases described by
- usual name: ε-Pu...
- database name: BCC_A2...
- prototype: W...
- StrukturBericht: *A2*...

Table

BETA_RHOMBO_B	B	*hR*105		(B)
BETA_SIALON	β-Si_3N_4	*hP*14		$(Si^{4+})_3$ $(N^{3-})_3$ (N^{3-})
BF_CASI	BCr	*oC*8	*Bf*	(Ca) (Si)
BH_MC_SHP	WC	*hP*2	B_h	(Mo, Re, W) (C)
MoC, WC				
C1_MO2	CaF_2	*cF*12	*C*1	$(Al^{3+}, Am^{3+}, Am^{4+}, Ca^{2+}, Np^{3+}, Np^{4+}, Pu^{3+}, Pu^{4+}, Th^{4+}, U^{3+}, U^{4+}, U^{5+}, Zr^{2+}, Zr^{4+})$ $(O^{2-}, \varnothing)_2$ $(O^{2-}, \varnothing)$
UO_2, PuO_2, ZrO_2, AmO_2, NpO_2				
C2_MTE2	FeS_2	*cP*12	*C*2	(Rh,Ru,∅) $(Te)_2$
Pyrite				
C4_RUTILE	TiO_2	*tP*6	*C*4	(Ru^{4+}, Ti^{4+}) $(O^{2-})_2$
C4_TI2N	TiO_2	*tP*6	*C*4	$(Ti)_2$ (C, N)
Described independently of TiO_2				
C6_B81	NiAs	*hP*4	$B8_1$	(Pd,Rh) (Pd,Rh,∅) $(Te)_2$
Identical model for CdI_2				
C6_B81_2	CdI_2	*hP*3	*C*6	(Pd,Rh
Identical model for NiAs				
C11B_AGM2	$MoSi_2$	*tI*6	$C11_b$	(Ag) (
C11B_MOSI2	$MoSi_2$	*tI*6	$C11_b$	(Mo, R
C12_CASI2	$CaSi_2$	*hR*18	*C*12	(Ca) (
C14_LAVES	$MgZn_2$	*hP*12	*C*14	(Cr, Fe
C15_LAVES	$MgCu_2$	*cF*8	*C*15	(Al, Cr
C16_THETA	$CuAl_2$	*tI*12	*C*16	(Ta, Z
C23_CA2SI	Co_2Si	*oP*12	*C*23	$(Ca)_2$

TAF-ID : Thermodynamics of Advanced Fuels - International Database

Home | Introduction | Models | Phases | Systems

Usual name

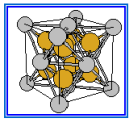
ZrO_2-γ, PuO_2, UO_2, AmO_2, NpO_2, ThO_2

Name in the database

C1_MO2

Crystallography

Prototype CaF_2 ***StrukturBericht*** *C*1 ***Pearson*** *cF*12 ***Space Group*** *Fm-3m*



Characteristics of the different sites for the prototype

Occupation	Multiplicity	Wickoff	Symmetry
F	8	c	-43m
Ca	4	a	m-3m

Thermodynamic model

$(Al^{3+}, Am^{3+}, Am^{4+}, Ca^{2+}, Np^{3+}, Np^{4+}, Pu^{3+}, Pu^{4+}, Th^{4+}, U^{3+}, U^{4+}, U^{5+}, Zr^{2+}, Zr^{4+})$ $(O^{2-}, \varnothing)_2$ $(O^{2-}, \varnothing)$

Assessed systems where the phase is stable

Am-O Ce-O Np-O O-Pu O-Th O-U O-Zr Am-O-Pu C-O-Pu C-O-U Nd-O-U O-Pu-U O-Pu-Zr

DOCUMENTATION (N. DUPIN)

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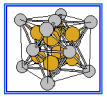

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Assessed systems where the phase is stable

Am-O

TAF-ID : Thermodynamics of Advanced Fuels - International Database

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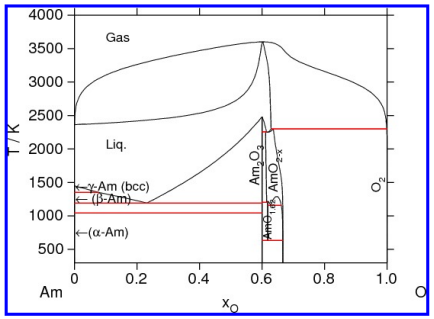
Binary system Am-O

Am-O-Pu Am O Binary systems Ternary systems Periodic table



Source of the description

P. Gotcu-Freis, P.Y. Colle, C. Guéneau, N. Dupin, B. Sundman, E.J.M Konings, *J. Nucl. Mater.*, **414**, 408-421 (2011)

Comments

This system is not very well known experimentally.

Comparison with experiments

Bibliography

Divers files

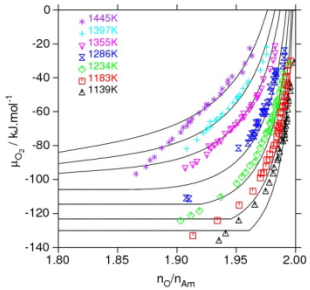
TAF-ID : Thermodynamics of Advanced Fuels - International Database

Home | Introduction | Models | Phases | Systems

Binary system Am-O

Am-O-Pu Am O Binary systems Ternary systems Periodic table

Comparison with experiments



Main page for the system Am-O **Bibliography** Divers files

Other links Am-O-Pu Am O Binary systems Ternary systems Periodic table

CONCLUSION

- Version 1 of TAF-ID database is already available for the participants of the TAF-ID project
- A public version restricted to only published work will be available for all the member countries of OECD in dec. 2013
- The TAF-ID database will be available in Thermo-Calc and FACTSAGE formats
  - Problems of conversion to be solved
- A technical meeting will be held in Chicago on 17-18 Oct. to exchange and hopefully agree on descriptions of many different systems
- **New participants are welcome !**