FUELBASE, TAF-ID DATABASES AND OC SOFTWARE: ADVANCED COMPUTATIONAL TOOLS TO PERFORM THERMODYNAMIC CALCULATIONS ON NUCLEAR FUEL MATERIALS

C. Guéneau[1], S. Gossé[1], A. Quaini[1], N. Dupin[2], B. Sundman[3], M. Kurata[4], T. Besmann[5], P. Turchi[6] , J.-C. Dumas[7], E. Corcoran[8], M. Piro[9], T. Ogata[10] , R. Hania[11] , B.O. Lee[12], R. Kennedy[13], S. Massara[14]

[1] CEA, DEN, DPC, SCCME , 91191 Gif-sur-Yvette Cedex, France
[2] Calcul Thermodynamique, 63670 Orcet, France
[3] CEA, INSTN, 91191 Gif-sur-Yvette Cedex, France
[4] JAEA, Tokai-mura, Ibaraki-ken, 319-1195 Japan
[5] ORNL, Oak Ridge, TN 37831, USA
[6] LLNL, Livermore, USA
[7] CEA, DEN, DEC, SESC, 13108 Saint-Paul lez Durance, France
[8] RMC , Station Forces Kingston, Canada
[9]CNL, Chalk River, Ontario K0J 1J0, Canada
[10] CRIEPI, Tokyo 201-8511, Japan
[11] NRG, NL-1755 LE Petten, Netherlands
[12] KAERI, Taejon 305600, South Korea
[13] INL, Idaho Falls, ID 83415, USA
[14] OECD, NEA, Nuclear Science Section, 92130 Issy-les-Moulineaux, France

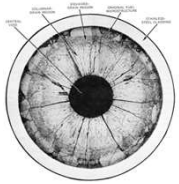
CONTEXT AND AIM OF THE TAF-ID PROJECT

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 facilitate the **exchange** on the models, review of experimental data, softwares, assessments ….

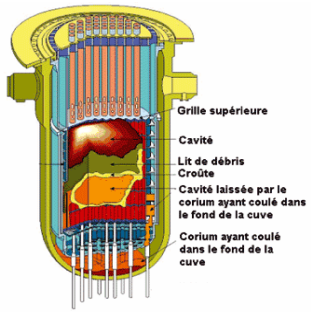
CONTEXT ➡ Fuel materials for Generation 2,3 & 4 reactors

- **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



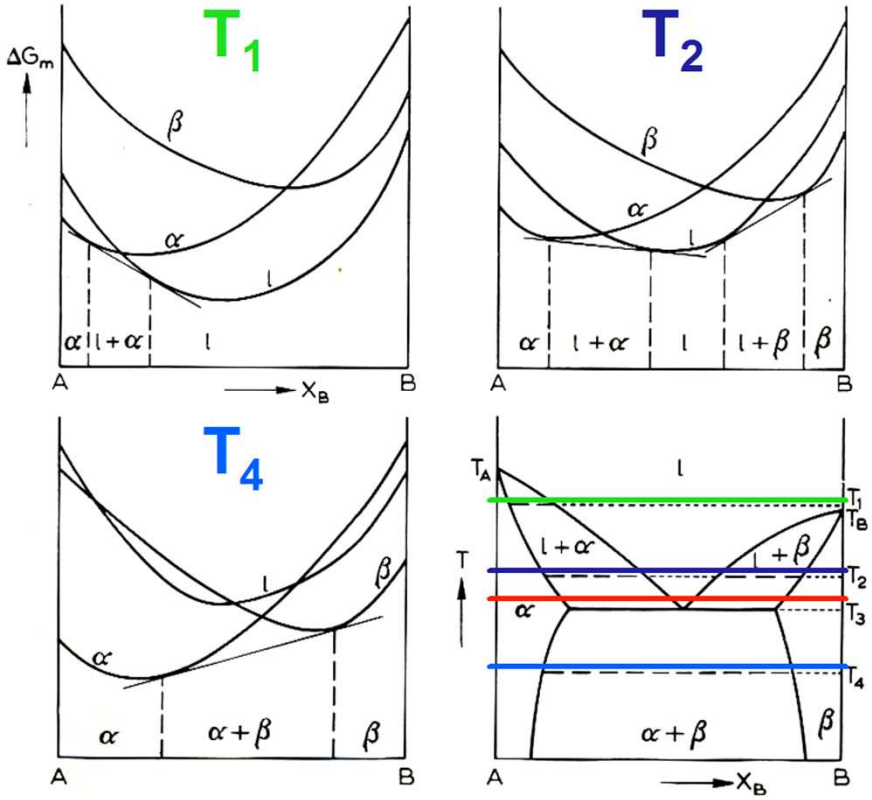
APPLICATIONS

- **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**



CALPHAD METHOD

The thermodynamic equilibrium is calculated by minimizing the Gibbs energy of the system

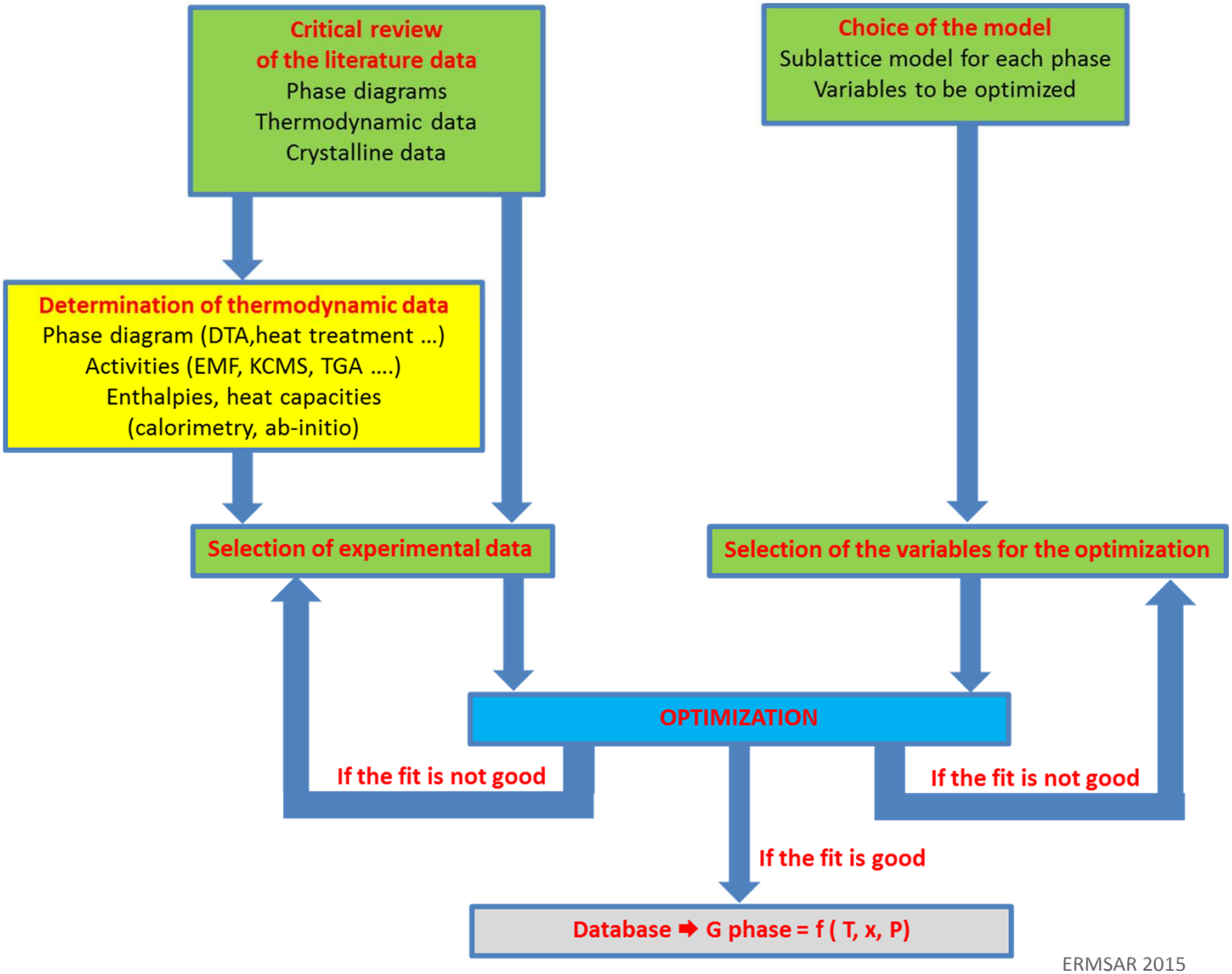


$$G = \sum_{\alpha} m^{\alpha} . G_m^{\alpha}$$

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

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

CALPHAD METHOD



FUELBASE DATABASE
FOR ADVANCED NUCLEAR FUEL MATERIALS (2005-2012)

- **FUELBASE was a collaboration between CEA, Calcul Thermo, ITU, NRG and ORNL and served as the model for the expanded TAF-ID project**

Am	C	Cr	Mo	N	Nb	Np	O	Pu	Re	Ru	Si	Ta	Ti	U	V	W	Zr
Am						Kurata 10	Gotcu 10	Dupin 07						Kurata 10			
C		Andersson 87	Andersson 88	SGTE	Lee 01		SGTE	CEA 10	Dupin 07		Grobner 96	Frisk 96	Dumitrescu 98	Dupin 09	Huang 91	Jonsson 93	Guillermet 95
Cr			Frisk 88	Frisk 91			Taylor 90	Dupin 06			Coughanowr 94		Saunders 98	Dupin 05	Lee 92		Zeng 98
Mo				Frisk 91			Sundman 07	Dupin 05	Dupin 08		Liu 05		Chung 99	Dupin 06			Jerlerud 03
N							SGTE	Sundman 05			Hillert 92		Jonsson 96	Chevalier 00	Ohtani 91		Ma 04
Nb								Berche 09			Shao 04			Liu 08			Guillermet 91
Np							Benes 09	Dupin 08						Kurata 10			
O								Gueneau 08			Hallsted 93		Sundman 05	Gueneau			Liang 01
Pu									Berche 08	Dupin 08	Dupin 05	Berche 09	Dupin 06	Kurata 99	Berche 09	Berche 08	Kurata 99
Re											Dupin			Berche 08		Dupin	
Ru														Dupin 08			
Si												Vahmas 89	Seifert 00	Berche 09		Vahmas 89	Guéneau 94
Ta														Berche 09			
Ti														Rado 06	Saunders 98		Kumar 94
U															Berche 09	Sundman 07	Chevalier 04

NRG

cea

OAK RIDGE National Laboratory

JRC - Institute for Transuranium Elements

F-BRIDGE

ACTINET
A NETWORK FOR ACTINIDE SCIENCES

ACTINET i3
Integrated Infrastructure Initiative for Actinide Science

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**

		Programme Review Group	
Country	**Signatory**	**Representative**	**Alternate Member**
Canada	CNL, RMCC, UOIT	M. Piro	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 maintenance and documentation
- Coordinated by S. Massara from OECD/NEA➡ http://www.oecd-nea.org/science/taf-id

DATABASES DEVELOPED BY THE PARTNERS

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

Thermochemical models may be inconsistent between contributors and need to be reconciled

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)

Models for MOX solid solution with Lanthanides

ORNL: determines oxygen potential versus burnup

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

➡ Phase diagrams can be calculated

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

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

The aims of our project are:

- **Merge the different databases,**
- **Agree on thermodynamic descriptions** for binary and ternary sub-systems
- **Propose experiments** to improve the descriptions

TAF-ID DATABASE (VERSION 5 – JANUARY 2015)

ELEMENTS ➡ 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-Te-Th-Ti-U-V-W-Zr

BINARY SYSTEMS

Ag-I Ag-O Ag-Ti Ag-Zr
Al-Ca Al-Cr Al-Fe Al-Mg Al-O Al-Si Al-U Al-Zr
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 B-Zr
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 Ca-U* Ca-Zr*
Ce-Cr Ce-Fe Ce-O*
Cr-Cs* Cr-Fe Cr-H Cr-I Cr-La* Cr-Mo Cr-N Cr-Nd* 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-Nd Fe-Ni Fe-Np Fe-O Fe-Pu Fe-Si Fe-U Fe-Zr
Gd-O Gd-U*
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 Mg-U* Mg-Zr*
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 Ni-U
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-Tc* Pd-Te* Pd-Zr
Pu-Re* Pu-Ru* Pu-Si* Pu-Ti* Pu-U Pu-W* Pu-Zr
Re-Si Re-U* Re-W
Rh-Ru* Rh-Tc* 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*: 72 assessed within our project
In blue: 116 coming from the literature

TERNARY SYSTEMS

Al-Ca-O Al-Cr-O Al-Fe-O Al-Mg-O
Al-O-Si Al-O-U Al-O-Zr
Am-O-Pu
B-C-Fe B-C-U B-C-Zr B-Fe-Zr B-Pu-U
C-Mo-Re C-Mo-Si C-Mo-Ti C-Mo-U C-N-Ti C-N-U
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 Ca-O-U Ca-O-Zr
Cr-Fe-O Cr-Fe-Zr
Cs-Mo-O Cs-O-U Cs-O-Zr
Fe-O-Si Fe-O-U Fe-O-Zr Fe-U-Zr
Gd-O-U
La-O-U
Mg-O-Si Mg-O-U Mg-O-Zr
Mo-O-U Mo-Pd-Rh Mo-Pd-Ru Mo-Rh-Ru
Nb-O-U
Nd-O-U
Ni-O-Si
O-Pu-U O-Pu-Zr O-Si-U O-Si-Zr O-U-Zr
Pd-Rh-Ru
Pu-U-Zr

CRYSTALLOGRAPHIC DATA

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	$(Al^{3+}, Am^{3+}, Am^{4+}, Ca^{2+}, Np^{3+}, Np^{4+}, Pu^{3+}, Pu^{4+}, Th^{4+}, U^{3+}, U^{4+}, U^{5+}, Zr^{2+}, Zr^{4+})$ $(O^{2-}, ∅)_2$ $(O^{2-}, ∅)$
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	TlO_2	*tP*6	*C*4	$(Tl)_2$ (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

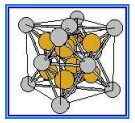
ZrO_2-Y, PuO_2, UO_2, AmO_2, NpO_2, ThO_2

Name in the database

C1_MO2

Crystallography

Prototype CaF_2 ***StrukturBericht*** *C1* ***Pearson*** *cF12* ***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

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

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

A single sublattice model for all the phases with the same crystalline structure

Cr-Fe-Zr

The source of the description is clearly mentioned.

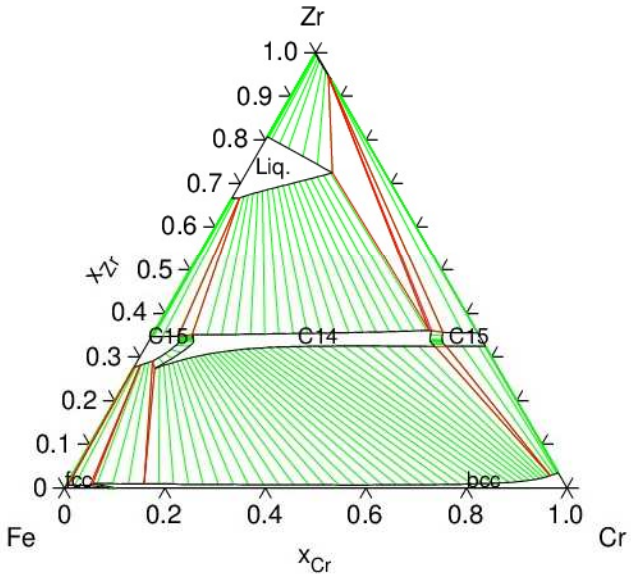
TAF-ID : Thermodynamics of Advanced Fuels - International Database

Home | Introduction | Models | Phases | Systems | TDB

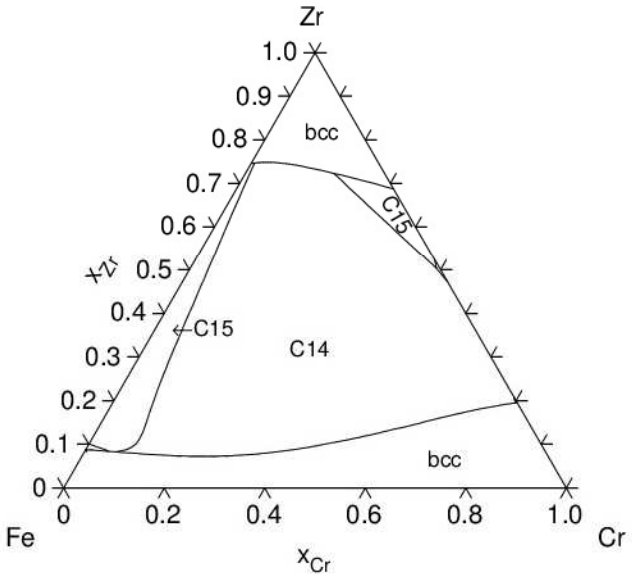
Ternary system Cr-Fe-Zr

Cr-Fe Cr-Zr Fe-Zr Cr Fe Zr Binary systems Ternary systems Periodic table

Isothermal section at 1500 K



Projection of the monovariant lines of the liquidus



Source of the description

N. Dupin, TAF-ID, 2014 october

Comments

The description recently published by Yang *et al.* was not used because of different descriptions for the Fe-Zr and Cr-Zr subsystems. The early description by Barberis *et al.* was using a Cr-Zr description with an overestimated enthalpy of formation for the Laves phases.

Cr-Fe-Zr

TAF-ID : Thermodynamics of Advanced Fuels - International Database

Home | Introduction | Models | Phases | Systems | TDB

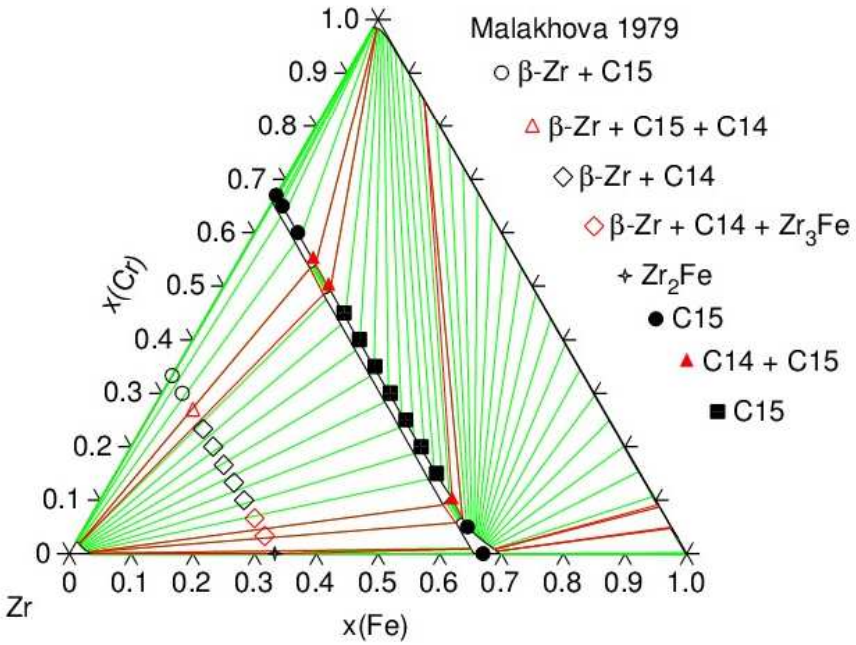
Ternary system Cr-Fe-Zr

Cr-Fe Cr-Zr Fe-Zr Cr Fe Zr Binary systems Ternary systems Periodic table

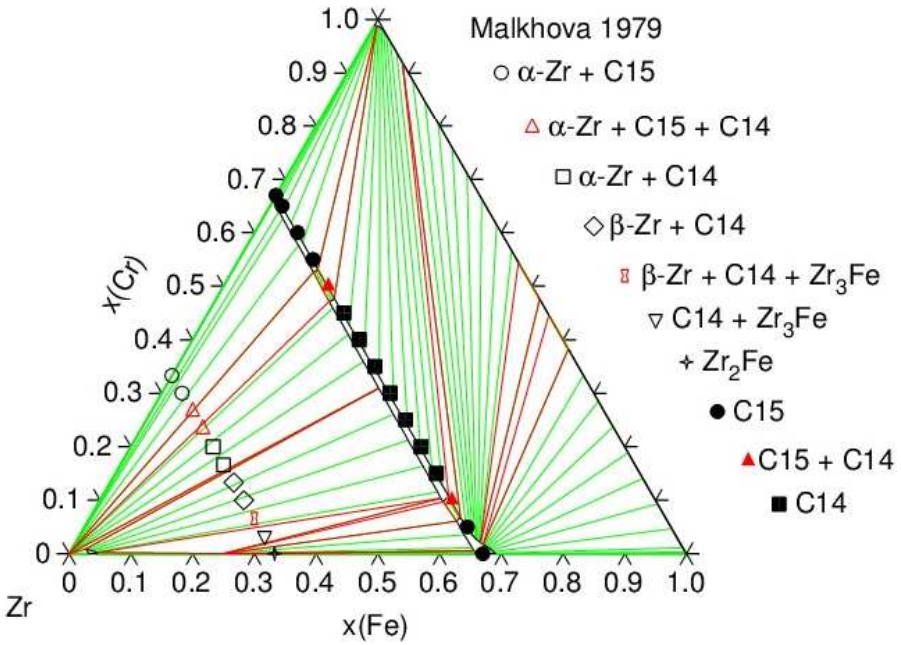
Comparison with experiments

Phase diagram

T = 1148 K



T = 1073 K



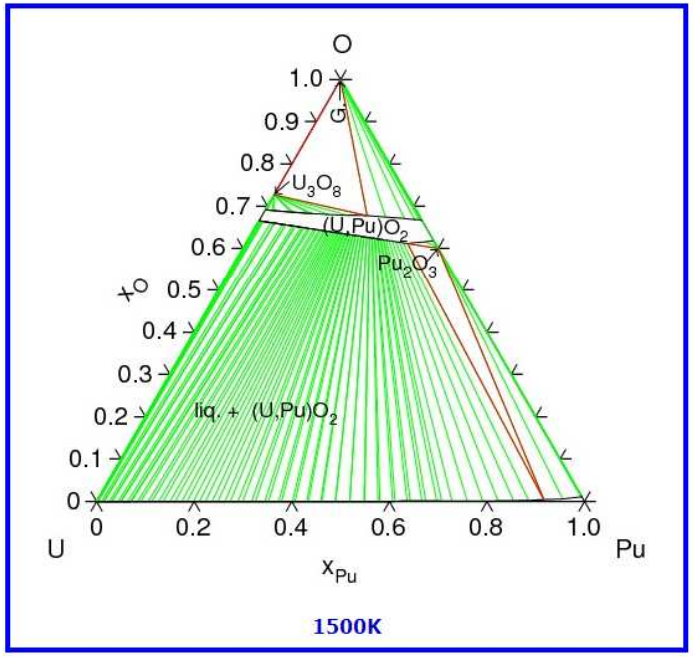
O-PU-U

TAF-ID : Thermodynamics of Advanced Fuels - International Database

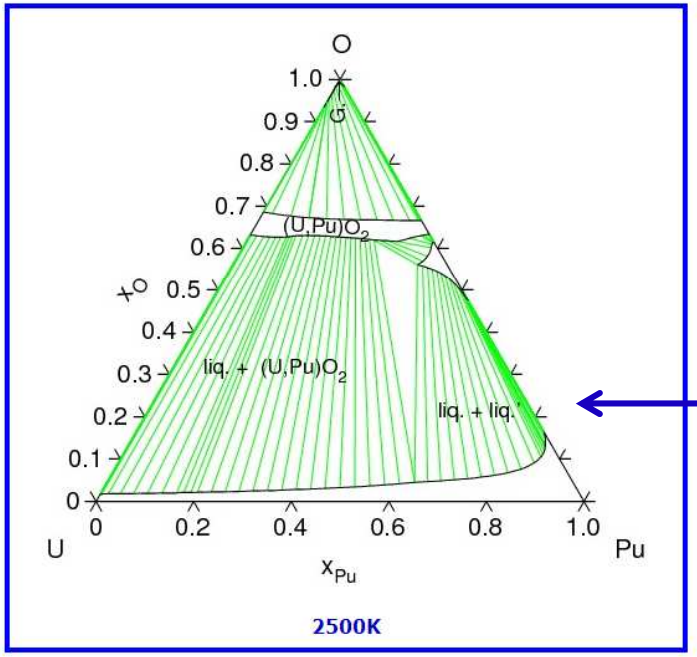
Home | Introduction | Models | Phases | Systems

Ternary system O-Pu-U

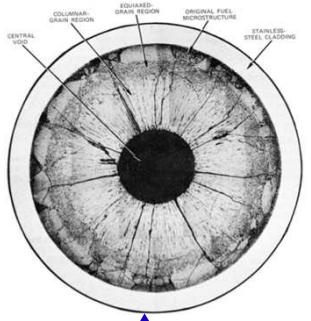
O-Pu O-U Pu-U O Pu U Binary systems Ternary systems Periodic table



1500K



2500K



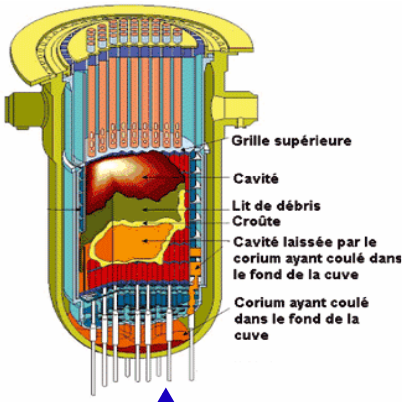
MOX fuel in SFR

-Interaction with steel clad
- Fission product chemistry

Source of the description

C. Guéneau, N. Dupin, B. Sundman, C. Martial, J.C. Dumas, S. Gossé, S. Chatain, F. De Bruycker, D. Manara, R. Konings, Thermodynamic modelling of advanced oxide and carbide nuclear fuels: Description of U-Pu-O-C systems, *J. Nuclear Materials*, **419** (2011) 145-167

O-U-ZR



TAF-ID : Thermodynamics of Advanced Fuels - International Database

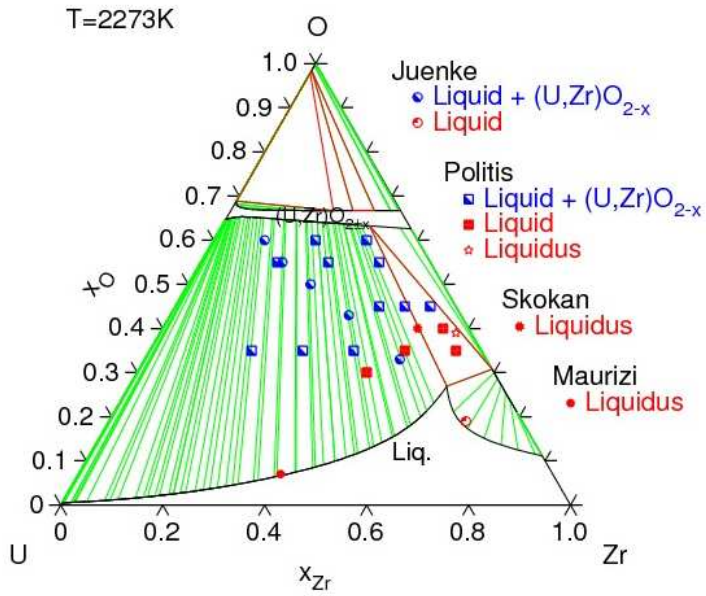
Home | Introduction | Models | Phases | Systems

Ternary system O-U-Zr

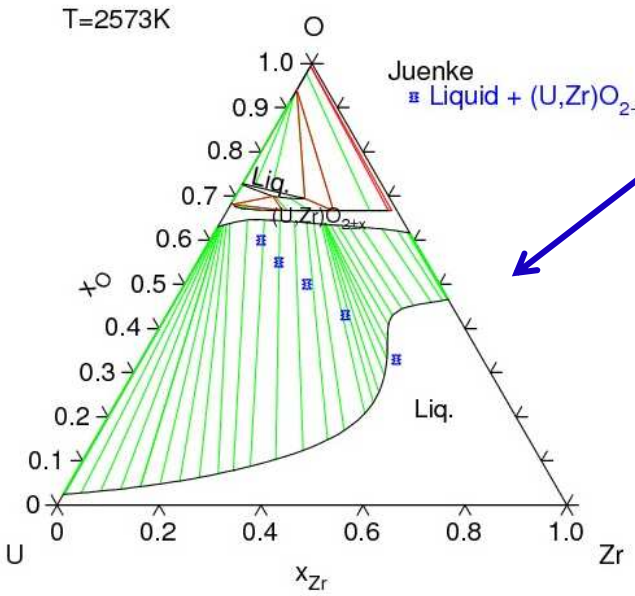
O-U O-Zr U-Zr O U Zr Binary systems Ternary system

Comparison with experiments

Isothermal section at 2273K

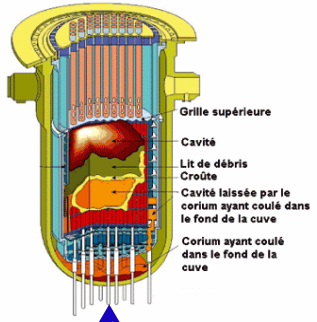


Isothermal section at 2573K



Corium (U,Zr,O) forming from UO_2/Zr interaction

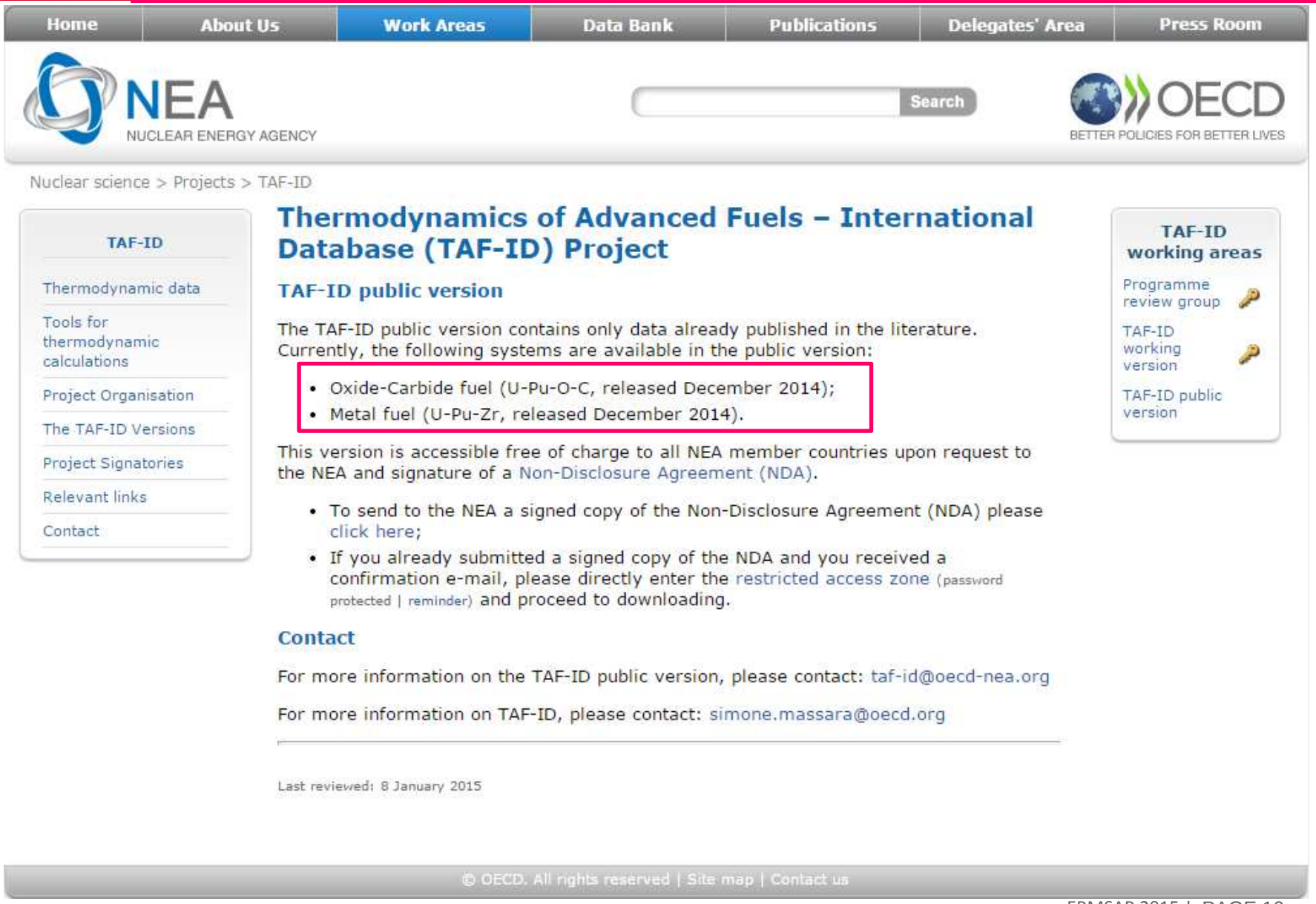
UO2-CONCRETE AND ZRO2-CONCRETE INTERACTION



Corium (U,Zr,O,Fe) forming from UO_2/Zr/Fe interaction

$UO_2 + ZrO_2$ + concrete

PUBLIC VERSION



Home | About Us | Work Areas | Data Bank | Publications | Delegates' Area | Press Room

NEA NUCLEAR ENERGY AGENCY

Search

OECD BETTER POLICIES FOR BETTER LIVES

Nuclear science > Projects > TAF-ID

TAF-ID

- Thermodynamic data
- Tools for thermodynamic calculations
- Project Organisation
- The TAF-ID Versions
- Project Signatories
- Relevant links
- Contact

Thermodynamics of Advanced Fuels – International Database (TAF-ID) Project

TAF-ID public version

The TAF-ID public version contains only data already published in the literature. Currently, the following systems are available in the public version:

- Oxide-Carbide fuel (U-Pu-O-C, released December 2014);
- Metal fuel (U-Pu-Zr, released December 2014).

This version is accessible free of charge to all NEA member countries upon request to the NEA and signature of a Non-Disclosure Agreement (NDA).

- To send to the NEA a signed copy of the Non-Disclosure Agreement (NDA) please click here;
- If you already submitted a signed copy of the NDA and you received a confirmation e-mail, please directly enter the restricted access zone (password protected | reminder) and proceed to downloading.

Contact

For more information on the TAF-ID public version, please contact: taf-id@oecd-nea.org

For more information on TAF-ID, please contact: simone.massara@oecd.org

Last reviewed: 8 January 2015

TAF-ID working areas

- Programme review group
- TAF-ID working version
- TAF-ID public version

© OECD. All rights reserved | Site map | Contact us

OpenCalphad SOFTWARE (BO SUNDMAN)

http://www.opencalphad.com/

- OC is an **open source** software to perform thermodynamic calculations using the CALPHAD method.
- OC uses as input data Databases containing mathematical models of the Gibbs energy as function of temperature, composition and pressure. **OC is compatible with databases in Thermo-Calc format (TDB)**.
- A **grid minimizer** is used to provide an initial global minima as a start point for the equilibrium calculation. This solves the problems related to miscibility gaps.
- A **software interface module** is available to compute a thermodynamic equilibrium for given conditions (P,T,N,x_i) to provide input data to other softwares (diffusion, phase field, thermohydraulics, thermal …).
- Calculations as function of temperature (or composition) can be performed.
- Binary phase diagrams can be calculated.
- A project is in progress to develop a **new assessment module** to develop the Gibbs Energy database.

SUMMARY

- The Working Version 5 of TAF-ID database is available for the participants of the TAF-ID project
- A public version restricted to only published work is available for all the member countries of OECD/NEA since december 2014
  - ➡ https://www.oecd-nea.org/science/taf-id/taf-id-public/
- The TAF-ID database will be available in Thermo-Calc and FACTSAGE formats
  - A software to convert the databases from both codes is being developed
- The TAF-ID project may be extended for 3 years
- OC is an open source software for CALPHAD calculations, compatible with Thermo-Calc format (TDB)
- Both OC and TAF-ID are operational computational tools to perform thermodynamic calculations on corium systems for severe accident applications