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The OpenCalphad thermodynamic software interface

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(Article begins on next page)

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Corresponding Author: Prof. Bo Sundman, PhD

Corresponding Author's Institution: -

First Author: Bo Sundman, PhD

Order of Authors: Bo Sundman, PhD; Ursula R Kattner, PhD; Christophe
Sigli, PhD; Matthias Stratmann, PhD; Romain Le Tellier, PhD; Mauro
Palumbo, PhD; Suzana G Fries, PhD

Abstract: Thermodynamic data are needed for all kinds of simulations of materials processes. Thermodynamics determines the set of stable phases and also provides chemical potentials, compositions and driving forces for nucleation of new phases and phase transformations. Software to simulate materials properties needs accurate and consistent thermodynamic data to predict metastable states that occur during phase transformations. Due to long calculation times thermodynamic data are frequently pre-calculated into ``lookup tables'' to speed up calculations. This creates additional uncertainties as data must be interpolated or extrapolated and conditions may differ from those assumed for creating the lookup table. Speed and accuracy requires that thermodynamic software is fully parallelized and the OpenCalphad (OC) software is the first thermodynamic software supporting this feature.

This paper gives a brief introduction to computational thermodynamics and introduces the basic features of the OC software and presents four different application examples to demonstrate its versatility.

This paper is a collaboration between several scientist to present the possibilities of a free thermodynamic software in different kinds of applications in materials science. This software provides the possibility to calculate equilibria in parallel and we believe this is a new feature not yet available in similar software and this feature is important for improving the future use of thermodynamics in simulations. We provide a brief description of the software and several examples of its use.

Response to reviewer, reviewer text indicated by ->

-> Reviewer #1: In this paper, the authors describe recent developments of the

-> OpenCalphad open-source thermodynamic software package and its
-> interfacing with 'client' programs. Several examples are given
-> to illustrate the possibilities for concrete applications.

-> The subject is interesting, and in my opinion the presented
-> material deserves publication. However, I believe that substantial
-> modifications to the manuscript are necessary before publication.

-> The problem for me is: what is the target audience, and what
-> is the purpose of the paper? Some parts read almost like a
-> user manual for the program, and the appendices 2 and 3 belong
-> rather to the documentation part of a software website than to
-> a scientific article (appendix 2 is a list of functions -
-> what is its use in an article? If I can click on each item
-> and get additional instructions, such as is the case in
-> well-designed web sites, then it's useful, but in paper form
-> I do not see what information this list should add). Other parts
-> are intended to explain to a general audience of materials
-> scientists the usefulness of the tools on the example of
-> concrete problems.

The authors are pleased with the careful review and we acknowledge that the paper has been too detailed in some parts and too brief in others.

The audience we are aiming for are all materials scientists who develop or use software using thermodynamic data. We have tried to expand the explanations of the basic functions and algorithm and we have removed the details of the current software, such as Appendices 2 and 3.

-> For me, the latter aspect of the paper should be reinforced.
-> But this requires adding quite some information - in many
-> places, the presented material is partial or sketchy. A few
-> concrete examples and recommendations are:

-> 1) After Eq. (1): what do you mean by 'surface of reference'?
-> This is a non-standard term (at least for me)

The explanation of the Gibbs energy function for the models are now extended although it is difficult to make this short.

-> 2) Beginning of Section 2.3: The first paragraph is an
-> enumeration of other papers that I should read to understand
-> what is said here. I would expect a little more information
-> to make the paper self-contained. For example, what is the
-> principle of Hillert's algorithm and why is it better than
-> others? How is the stability of a phase prescribed?

The algorithm is explained in more details including the technique to select the set of stable phases.

Comparison with other (commercial) software is not easy as they have not published enough details of their algorithms. One of the authors

has long experience working with the Thermo-Calc (TC) software and the OC software is a new implementation of the same algorithm. A very rough attempt to compare the calculation speed between OC and TC is to calculate a phase diagram for a 5 component steel, this takes OC 13 CPU seconds to calculate and the same diagram with TC takes 12 CPU seconds on the same computer. But a phase diagram calculation involves much more than just equilibrium calculations, there are step control, occasional global minimizations etc. The authors are convinced the calculation speed and results using OC are comparable with commercial thermodynamic software but we have no "hard facts" to put in a paper.

-> 3) End of Sec. 2.4: there is a standard methodology to demonstrate the scalability of an algorithm, that is, the same calculation is performed with different number of processors, and the execution time is compared. Here, only one statement is made (in addition with the 'wishi-washi' word 'almost'). We need some hard numbers here to make the case for scalability.

We have added a table with a detailed comparison of sequential/parallel calculation. Also in section 4.2 there is a verifiable comparison using sequential/parallel calculations.

-> 4) Section 3, beginning of second paragraph: I do not believe that it is useful to announce things that are 'under development'; by the time the article is published, hopefully things will have moved forward. Stated otherwise, things that are not finished should not be published. Same for the next paragraph.

OCASI is fully operational but the documentation is rudimentary and incomplete. There will also be a revision based on the experiences of the use.

-> 5) Section 3.1: it is very unusual to refer to a Ph.D. thesis unless it is already publicly available.

The sentence was reworded and the in-text reference to the thesis was removed.

-> 6) Section 4: 'Some of the examples presented here may require the use of proprietary software'. Since the examples were all calculated, I do not understand the 'may'. Which of the examples? Which software?

The proprietary software using OCASI is the one used for the simulation of the aluminium alloy in section 4.2. This is now indicated in the text.

-> 7) Section 4.2, second paragraph (page 14): the simulation setup is not at all described. What does 'a two-dimensional cylindrical geometry' mean? Are all the potential readers supposed to know what is back-diffusion, the Scheil model, and the secondary dendrite arm spacing? A more detailed description is needed here.

This section has now been extended with additional references.

-> 8) End of Section 4.2: again, problems with the documentation of

- > scalability. The ratio between 3.5 days and 3.5 hours is 24;
- > this cannot be explained by the use of 12 processors. How much
- > is the execution time of the SAME software on a single processor ?

A precise figure is now given for the effect of parallelization. The previous value was comparing the old sequential version of the software using a different equilibrium algorithm with the revised using OCASI, i.e. included many other changes than parallelization.

- > In summary, the article should be enriched and made more self-consistent
- > to be useful for a wider audience; appendices 2 and 3 could be deleted.

We have tried to make the article more readable and interesting and removed Appendices 2 and 3.

No graphical abstract

The OpenCalphad thermodynamic software interface

Bo Sundman^a, Ursula R Kattner^b, Christophe Sigli^c, Matthias Stratmann^d,
Romain Le Tellier^e, Mauro Palumbo^f, Suzana G Fries^d

^a*INSTN, CEA Saclay, France*

^b*National Institute of Standards and Technology, Gaithersburg, MD, USA*

^c*Constellium Technology Center, CS 10027, F-38341 Voreppe Cedex, France*

^d*Ruhr University Bochum, Germany*

^e*CEA, DEN, DTN/SMTA/LPMA Cadarache, F-13108 St Paul-lez-Durance, France*

^f*SISSA, Trieste, Italy*

Abstract

Thermodynamic data are needed for all kinds of simulations of materials processes. Thermodynamics determines the set of stable phases and also provides chemical potentials, compositions and driving forces for nucleation of new phases and phase transformations. Software to simulate materials properties needs accurate and consistent thermodynamic data to predict metastable states that occur during phase transformations. Due to long calculation times thermodynamic data are frequently pre-calculated into “lookup tables” to speed up calculations. This creates additional uncertainties as data must be interpolated or extrapolated and conditions may differ from those assumed for creating the lookup table. Speed and accuracy requires that thermodynamic software is fully parallelized and the OpenCalphad (OC) software is the first thermodynamic software supporting this feature.

This paper gives a brief introduction to computational thermodynam-

Email address: bo.sundman@gmail.com (Bo Sundman)

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9 ics and introduces the basic features of the OC software and presents four
10 different application examples to demonstrate its versatility.

11 *Keywords:* Computational Thermodynamics, CALPHAD, Phase
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13 Transformations, Simulations, Parallel Computing, Free Software,
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17 18 **1. Introduction**

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21 The background for the development of a free thermodynamic software
22 for multicomponent calculations using the CALPHAD method (CALculation
23 of PHase Diagrams) is described by Sundman et al. [1]. A very limited first
24 version of the OpenCalphad (OC) software was released in 2013. In February
25 2015, a second version, capable also of multicomponent single equilibrium
26 and some phase diagram calculations, was released. During 2015, extensive
27 new facilities were added and the software became much more stable and is
28 available at [2]. Version 3 includes parallel equilibrium calculations using the
29 OpenMP [3] library, a rudimentary assessment module and an Application
30 Software Interface, OCASI, callable from C++ as well as Fortran and other
31 programming languages. OC is written using the new Fortran standard,
32 including 2008 additions.
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44 There is extensive literature describing the use of thermodynamic data
45 for materials and process development. Although somewhat outdated, an
46 excellent review can be found in the book by Saunders and Miodownik [4].
47 Especially the development of phase field techniques [5, 6, 7] and other tech-
48 niques, such as fluid dynamics [8] and finite element methods [9] for simulat-
49 ing micro-structure evolution has increased the demand for fast and reliable
50 calculations of chemical potentials, driving forces, mobilities etc. Within the
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9 framework of Integrated Computational Materials Engineering (ICME) [10]
10 the necessary tools are coupled to accomplish the desired simulations and
11 the OC software has the intention of becoming one of these tools.
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15 The OC software has been extensively tested on Windows and Linux sys-
16 tems and, although there are occasional problems, the overall performance is
17 comparable with that of commercial thermodynamics software for materials
18 science, such as FactSage [11], Pandat [12] or Thermo-Calc [13]. In addi-
19 tion, the OC software can perform many equilibrium calculations in parallel,
20 including calls to OCASI. The OCASI interface is fully functional and has
21 been used in some applications presented here to demonstrate how it can be
22 integrated in simulations using consistent thermodynamic data. Examples
23 and full documentation of the source code, as well as macro files showing
24 the interactive use of the software, are available on the OC web site [2].
25 A development version with several new features is available at the github
26 repository [14].
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39 **2. Computational Thermodynamics**

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42 Computational thermodynamics consists of software and databases for
43 calculating phase diagrams and all other kinds of thermodynamic data. The
44 databases are a collection of parameterized model descriptions of the phases
45 as function of constitution and state variables. The parameterized model
46 descriptions are obtained from the thermodynamic assessment (or optimiza-
47 tion) of individual systems. The basic techniques are described in the book
48 by Lukas et al. [15]. An important advantage from using an assessed ther-
49 modynamic database is that all data calculated with such a database are
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9 consistent, including phase solubilities, melting temperatures, heat capaci-
10 ties, chemical potentials etc.
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12 13 14 *2.1. State variables*

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16 A thermodynamic system is described by a set of state variables such
17 as T, P, V for temperature, pressure, volume. The amounts of the compo-
18 nents can be given in moles, N , mole fractions, x , or mass fractions, w . All
19 these variables determine functions of the state, such as internal energy, U ,
20 Helmholtz energy, A , or the Gibbs energy, G .
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24 Many properties derived from the state functions can be calculated, or
25 even used to control the system like chemical potentials, μ , enthalpies, H ,
26 etc. All state variables available in OC are listed in the Appendix.
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32 *2.2. Thermodynamic models*

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34 In CALPHAD-type thermodynamic databases the Gibbs energy of each
35 phase is described by a model reflecting its structure. Several different kinds
36 of models are described by Lukas et al. [15]. Many different kinds of models,
37 for example ideal gases, regular solutions, interstitial models and chemical
38 ordering are included in the Compound Energy Formalism (CEF)[16]. This
39 formalism describes the Gibbs energy per mole formula unit, G_M , of the
40 phase α with several sublattices as:
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$$48 \quad G_M^\alpha(T, P, y_{is}) = {}^{\text{srf}}G_M^\alpha - T {}^{\text{cfg}}S_M^\alpha + {}^E G_M^\alpha + {}^{\text{phys}}G_M^\alpha \quad (1)$$

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50 where T, P are temperature and pressure and y_{is}^α is the fraction of constituent
51 i on sublattice s and
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$$54 \quad {}^{\text{srf}}G_M^\alpha = \sum_I \Pi_I(y_{is}^\alpha) {}^\circ G_I^\alpha(T, P) \quad (2)$$

$${}^{\text{cfg}}S_M^\alpha = RT \sum_s a_s^\alpha \sum_i y_{is}^\alpha \ln(y_{is}^\alpha) \quad (3)$$

$${}^E G_M^\alpha = \sum_J \Pi(y_{js}^\alpha) L_J^\alpha(T, P, y_{js}^\alpha) \quad (4)$$

$$(5)$$

${}^{\text{srf}}G_M^\alpha$ is the so-called surface of reference where I is a constituent array specifying one constituent i in each sublattice, s and ${}^\circ G_I^\alpha$ is the Gibbs energy of formation of the compound I with the α structure from the standard states of the elements. ${}^{\text{cfg}}S_M^\alpha$ is the configurational entropy, assuming random mixing on each sublattice, a_s^α is the number of sites on sublattice s and R the gas constant. ${}^E G_M^\alpha$ contain modeling parameters that describe the composition dependence due to interactions between constituents specified by J . ${}^{\text{phys}}G_M^\alpha$ can be used for particular physical contributions such as from ferro-magnetism. The magnetic contribution to the Gibbs energy is usually modeled with a composition dependent Curie or Néel temperature and Bohr magneton number [17].

For liquids with strong short range ordering (SRO), the partially ionic 2-sublattice liquid model [18] is also implemented. Since the source code of OC is available, skilled users can implement their own models.

The mole fraction of a component j in a phase, x_j^α , can be calculated from the constituent fractions using

$$x_j = \frac{\sum_s a_s \sum_i b_{ji} y_{is}}{\sum_k \sum_s a_s \sum_i b_{ki} y_{is}} \quad (6)$$

where b_{ji} is the stoichiometric factor of element j in constituent i .

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9 *2.3. Equilibrium calculations*

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11 The model of the Gibbs energy of a phase forms a surface in a hyperspace
12 as a function of T, P and its constitution. A stable state, i.e. an equilibrium,
13 for a given set of conditions, is described by the minimum or maximum of
14 the appropriate thermodynamic function. From classical thermodynamics
15 we know that a system at constant T, P and amounts of all elements is at
16 equilibrium at a minimum in the Gibbs energy of the system. The equilibrium
17 Gibbs energy is given by:
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$$24 \quad G_m = \sum_i x_i \mu_i \quad (7)$$

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26 where G_m is Gibbs energy per mole of component and μ_i the chemical po-
27 tential of element i . If the system contains several phases we have for each
28 stable phase
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$$34 \quad G_m^\alpha = \sum_i x_i^\alpha \mu_i \quad (8)$$

$$35 \quad x_i = \sum_\varphi \aleph^\varphi x_i^\varphi \quad (9)$$

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37 where \aleph^φ is the number of formula units of phase φ .
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43 The equilibrium algorithm used in OC was proposed by Hillert [19] and
44 implemented by Jansson [20] in the POLY module of the Thermo-Calc soft-
45 ware and in the PMLFKT software by Lukas et al. [21]. The implementation
46 of this algorithm in OC is presented in [22]. It uses the chemical potentials
47 and the amount of the stable phases as variables to find the equilibrium by
48 iteratively varying the constituent fractions in all phases. If the amount of
49 a stable phase becomes less than zero at an iteration it is removed from the
50 stable phase set. If the Gibbs energy of an unstable phase, at its current
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composition, becomes lower than the Gibbs energy calculated for this composition using the current values of the chemical potentials, this phase is added to the stable phase set. Such changes in the set of stable phases must be treated with great care as described in[22].

The algorithm minimizes the Gibbs energy for a set of phases, φ :

$$G = \sum_{\varphi} N^{\varphi} G_M^{\varphi}(T, P, y_{is}^{\varphi}) \quad (10)$$

Different types of conditions and constraints can be used by adding the constraints multiplied with Lagrange multipliers. For known T, P and overall composition, the Lagrange function can be written:

$$L = G + \sum_i f_i \mu_i + \sum_{\varphi} \eta_s^{\varphi} g_s^{\varphi} \quad (11)$$

$$f_i = \sum_{\varphi} N^{\varphi} x_i^{\varphi} - \tilde{N}_i \quad (12)$$

$$g_s^{\varphi} = 1 - \sum_j y_{js}^{\varphi} \quad (13)$$

where \tilde{N}_i is the prescribed amount of element i and μ_i and η_s^{φ} are Lagrange multipliers. The Lagrange function will have the same extrema as the Gibbs energy when the constraints are fulfilled, i.e. zero. Additional terms with multipliers can be included for other conditions, for example if the volume is known or if the phase has charged constituents.

At equilibrium all partial derivatives of L should be zero and in particular that with respect to the amount of the phases:

$$\frac{\partial L}{\partial N^{\varphi}} = G_M^{\varphi} - \sum_i x_i \mu_i = 0 \quad (14)$$

and comparing with eq. 8 we find that the Lagrange multiplier μ_i is the chemical potential of element i . Hillert[19] proposed that the equilibrium

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9 calculation should be made in two steps which allows for both a change in
10 the set of stable phases as well as the constitutions of the phases to obtain the
11 minimum. Sundman et al.[22] give a detailed description of the implementa-
12 tion of Hillert’s algorithm in OC and also show how to calculate additional
13 properties such as heat capacities and slopes of liquidus surfaces from the
14 results of an equilibrium calculation. OC has an additional grid minimizer
15 to ensure that the calculated equilibrium is global and not local and to also
16 to detect miscibility gaps in phases.
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25 *2.4. Parallel calculations*

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27 The assessment (optimization) of model parameters usually includes a
28 large number of experimental data, each representing a value determined at
29 equilibrium. The assessment procedure calculates these experimental equi-
30 libria many times varying the selected model parameters in order to find the
31 best fit. Therefore, the ability to calculate multiple equilibria in parallel is
32 not only useful for simulations but also for the assessment (optimization) of
33 model parameters. The data structure in OC, described in [1], is specifically
34 designed to allow calculation of multiple equilibria in parallel.
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43 The OC source code is distributed with a macro file for calculating 400
44 separate equilibria of a 6 component steel. When tested on a 4 core Intel(R)
45 i7-4810MQ @ 2.80GHz and 8 GB 64 bit memory using 8 threads, the time
46 for an equilibrium calculation, either sequentially or with OpenMP and OC
47 compiled with or without -O2 optimization, is shown in Table 1. The gain
48 in clock speed using 8 threads is almost 4 i.e. there is very little overhead.
49 Looping the same test for very long time did not show any loss of memory
50 due to leaks and on a Linux system OC has been tested with the Valgrind
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Table 1: Times to calculate an equilibrium for a 6 component steel either sequentially or with 8 thread/4 kernels, with or without -O2 compiler option using the GNU Fortran 5.1 version. The times are mean values of 10 calculations of 400 different equilibria.

Time				
version	seq.	seq. -O2	8 threads	8 threads -O2
CPU seconds	1.135310^{-2}	5.33910^{-3}	2.09210^{-2}	9.22010^{-3}
clock cycles	11.321	5.347	2.988	1.213

software [23] system which indicate negligible memory loss during calculations.

It is not possible to make accurate comparisons with commercial software because these softwares do not provide detailed information of the time to calculate an equilibrium. But running the same calculation with a similar commercial software on the same hardware gave the impression that there is no significant difference in time for sequential calculations. There is currently no commercial software which can calculate equilibria in parallel.

2.5. Additional data depending on T, P and phase composition

Thermodynamic data are important but not sufficient for the simulation of phase transformations. For example, to consider the diffusion of elements, their mobilities together with the thermodynamic factor need to be described to provide the diffusion coefficient. In addition, there are also interface energies, interface mobilities, elastic constants and other phase-based properties that may be of interest [24].

In the OC software, such property descriptions can be stored together with the thermodynamic descriptions for each phase as functions of T, P and

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9 constitution of the phase. The values of these properties are calculated to-
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14 together with the thermodynamic properties during the equilibrium calculation
and can be accessed by the application software.

15 As an example, the natural logarithm of the mobility of Fe in the face
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17 centered cubic (FCC) phase, $M_{\text{Fe}}^{\text{FCC}}$, is denoted in OC as $MQ\&Fe(\text{FCC})$, and
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19 in the Fe-Ni system it can be described by a linear function:

$$20$$

$$21 M_{\text{Fe}}^{\text{FCC}} = MQ\&Fe(\text{FCC}) = x_{\text{Fe}}^{\text{FCC}} MQ\&Fe(\text{FCC}, \text{Fe}) + x_{\text{Ni}}^{\text{FCC}} MQ\&Fe(\text{FCC}, \text{Ni}) \quad (15)$$

$$22$$

23 where $x_{\text{Fe}}^{\text{FCC}}$ and $x_{\text{Ni}}^{\text{FCC}}$ are the mole fractions of Fe and Ni in FCC, re-
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25 spectively, $MQ\&Fe(\text{FCC}, \text{Fe})$ is the mobility of Fe in pure FCC Fe and
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27 $MQ\&Fe(\text{FCC}, \text{Ni})$ is the mobility of a single Fe atom in pure FCC Ni, both
28
29 of which can be expressed as:

$$30$$

$$31 MQ\&Fe(\text{FCC}, \text{Ni}) = \frac{Q_{\text{Fe}}}{RT} \ln(\nu_{\text{Fe}}) \quad (16)$$

$$32$$

33 where Q_{Fe} is the activation energy and ν_{Fe} the pre-exponential factor, both
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35 of which can depend on T and P .

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38 As mobilities can vary several orders of magnitude, it is a better approx-
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40 imation to describe the composition dependence by a linear relation of its
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42 logarithm. If sufficient experimental or theoretical data are available, non-
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44 linear terms can be added to eq. 15. Together with the thermodynamic
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46 factor, the mobility can be transformed to a diffusion coefficient needed for
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48 the simulation of micro-structure evolution in diffusion controlled processes.
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51 **3. The OCASI application software interface**

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54 Software interfaces for coupling thermodynamic calculations with appli-
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56 cation software following a proposed standard [25] have been implemented
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9 in ChemApp [26] and Thermo-Calc TQ [27]. The idea behind this proposed
10 standard is to overcome the differences between the different thermodynamic
11 software packages which can make the implementation into application soft-
12 ware difficult. The methodology with many separate subroutines and func-
13 tions performing specific tasks makes the implementation straightforward
14 and is also used for the OCASI.
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20 A preliminary version of the OCASI software and documentation is avail-
21 able at the websites mentioned above. As the source code is open and free
22 any user can directly access the whole source code and make modifications.
23 However, any such access should be done with caution as the data structure
24 and source code may change at a later update whereas the OCASI interface
25 should remain stable.
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32 33 *3.1. Interfacing with Java* 34

35 Based on Fortran and C++ compatibility, it is also possible to interface
36 OC functionalities in the Java language thanks to the Java Native Interface
37 (JNI) [28]. Such an interface has been developed for the PROCOR soft-
38 ware [29], implementing a Cahn-Hilliard based multicomponent multiphase
39 diffusion model [30]. This is briefly described in section 4.4. In addition to
40 the Cahn-Hilliard model requirements, equilibrium calculation functionali-
41 ties have been interfaced for the coupling with other physical models. Some
42 physical parameters needed by these models have to be evaluated from the
43 phase compositions obtained from the OC-based equilibrium calculations.
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3.2. Reading the database

The thermodynamic data are normally read from a database. The OC software supports the TDB format [31] proposed originally by SGTE [32] and used by several other software, most notably by Thermo-Calc and DICTRA[13]. The application software allows selection of elements for which data should be extracted from the database and also allows suspending phases that are not of interest for the application.

The application software must initiate the OC memory structure by a special subroutine call. This returns a pointer to an equilibrium record in the dynamic memory structure which contains information about the constitution and calculated results for all the phases. After this initialization other subroutines, such as reading the database, can be used. The models of the phases and their thermodynamic model parameters read from the database are stored in the static part of the OC memory structure since they are independent of the external conditions that will be applied for a calculation. The external conditions, the constitution of the phases, as well as calculated results are in the dynamic data memory. A schematic representation of both static and dynamic memory is shown in Fig. 1.

3.3. Setting conditions

The conditions are set one by one by a call to a subroutine. These represent, for example, the temperature, T , pressure, P , and amount of the components, N_i , in the simplest case. The number of conditions needed to calculate the equilibrium is given by Gibbs phase rule

$$f = n + 2 - p \tag{17}$$

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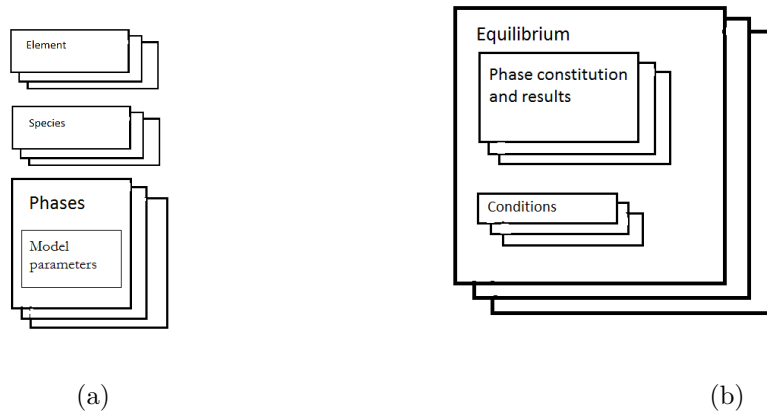


Figure 1: The static (a) and dynamic (b) parts of the OC memory. Each equilibrium record in (b) is independent and can be calculated in parallel.

where f is the degrees of freedom, n the number of components and p is the number of stable phases. Normally the number of stable phases is the result of the equilibrium calculation but OC allows that one or more phases can be prescribed to be stable. If there are no prescribed stable phases, $f = n + 2$, i.e. a system with a single component must have 3 conditions, normally T , P and N where N is the size of the system. For a binary system 4 conditions must be set and so on. If there is a major component the size of the system can be given as N and the amounts of other components as mole or mass fractions, x_i or w_i . Chemical potentials or activities of the components can be used instead of amounts as conditions as well as many other state variables.

3.4. Creating additional equilibria

It is possible to create additional equilibrium records to represent various local equilibria in a simulation. These may be calculated sequentially or in parallel as they are independent. The software provides a pointer to each equilibrium record with its own dynamic data structure and such a pointer

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9 must be supplied in calls to most subroutines to indicate which equilibrium
10 should be used.
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12 13 14 *3.5. Calculations*

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16 When the degrees of freedom according to eq. 17 are zero the equilib-
17 rium is calculated by a subroutine call. The calculation procedure in OC
18 is described in detail in [22]. In the call it is possible to specify whether
19 the global minimizer should be used or the calculation should start from the
20 last calculated set of stable phases and constitutions. As already stated each
21 equilibrium has its own set of conditions and can be calculated independently.
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28 29 *3.6. Extracting results*

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31 The normal way to obtain calculated results from OCASI is to call a
32 subroutine using the state variable symbols as listed in the Appendix. The
33 values of properties that are modeled separately, like the Curie temperature,
34 mobilities, etc. can be obtained in the same way.
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38
39 The pointer to the equilibrium data structure that was created when the
40 database was read is used to extract the results of an equilibrium calculation
41 for each phase. For example, with the equilibrium record pointer (called
42 “ceq” below) the user can access the array of records for the phases (called
43 “phase_varres”) and, after an equilibrium calculation, extract the value of
44 the Gibbs energy per mole formula unit of a phase with index lokcs as:
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49

```
50  
51 gfu = ceq%phase_varres(lokcs)%gval(1,1)  
52
```

53
54 where the % sign is used in the 2008 Fortran standard to indicate items in a
55 record. The lokcs variable is the index of the phase and provides an easy link
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9 between the phase and all its data. If the calculated mobility of component
10 1 of the same phase is stored in the location “imob”, its value is obtained by

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12
13
14 `mob(1) = ceq%phase_varres(lokcs)%gval(1,imob)`

15
16 Great care must be taken that the relevant data is extracted and the appli-
17 cation program must not change any data in the OC data structure.
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22 4. Examples of application software

23
24 Several simple examples of how to use the OC software interface are
25 provided with the source code. In one of the examples presented below
26 (section 4.2), the OCASI library is used inside proprietary software.
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31 4.1. *The beginnings of a reactor module*

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33 A process can sometimes be simulated by dividing it into sections, each of
34 which represent local equilibrium, and a transport of matter occurs between
35 these by diffusion or other means. The equilibrium calculation can be done
36 by OC in a function “localequil” with the following arguments:
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```
41 integer function localequil(todo,nsel,selel,tp,nmz,outspec,outval)
42 !
43 ! This routine calculates an equilibrium and return values
44 ! todo   in character variable      with "commands" to be executed
45 ! nsel   in integer                number of components
46 ! selel  in character*2 array       element names
47 ! tp     in double precision array(2) with values of T and P
48 ! nmz    in double precision array(nsel) with molar content for elements
49 ! outspec in character variable     information which data to extract
50 ! outval out double precision array(*) extracted values
51 ! if there is no error the function value returned is zero
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54 The todo argument passes a number of instructions to localequil, nsel
55 gives the number of elements specified in argument selel, tp provides the
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9 values of T and P and nmz the element amounts. The argument outspec
10 specifies which values will be returned in outval. A call of this function to
11 calculate a local equilibrium for the system Si-O-C at 1800 K and 1 bar with
12
13
14
15 a specified amount in moles of the elements can be

```
16  
17 nsel=3  
18 selel(1)='C '; selel(2)='O '; selel(3)='Si'  
19 tp(1)=1.80D2; tp(2)=1.0D5;  
20 nmz(si)=1.0D0; nmz(2)=2.0D0; nmz(3)=1.8D0  
21 todo='verbose open cosi.tdb '  
22 outspec='n(gas,*) mu(*) h h(gas) '  
23 ierr=localequil(todo,nsel,selel,tp,nmz,outspec,outval)  
24
```

25
26 The todo argument specifies VERBOSE to generate extra output, then to
27 OPEN the database file COSI.TDB and read the data for the system with the
28 elements in the argument selel and calculate the equilibrium at the specified
29 values of T , P and amounts of the elements. After a successful calculation the
30 amounts of all elements in the gas, n(gas,*), their chemical potentials, mu(*),
31 the total enthalpy, h, and the enthalpy of the gas, h(gas), will be provided
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39 in the array outval. The state variables that can be used are listed in the
40 Appendix. If an error occurs the function will return a non-zero number.

41
42 The localequil function will remember whether it has already read a
43 database and if no database is specified in a subsequent call the routine will
44 use the static data that it has already stored. In the application software the
45 amounts of elements in the gas and the other phases may be redistributed
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65 between different local equilibria to simulate a process. The range of options
in the localequil function can easily be extended. One of the intentions of
this function is to provide an application programmer simple access to the
OCASI interface.

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9 *4.2. Simulation of the homogenization of an alloy AA7449*

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11 The OpenCalphad Software has been recently coupled to the Constellium
12 thermodynamic package, ProPhase, using OCASI in order to use the faster
13 equilibrium routine included in OC as well as the new parallelization recently
14 implemented. One application of this software is to simulate the solidification
15 path and the segregations within dendrites formed after solidification.
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21 The simplest model to simulate solidification path (liquid concentration
22 evolution), solid fraction versus temperature curves and microsegregation
23 during casting is the Scheil model[33]. The assumptions are threefold: equi-
24 librium at the liquid/solid interface, no diffusion in the solid phases and
25 infinite diffusion in the liquid phase. The coupling of a Scheil solidification
26 routine with a phase diagram subroutine is straightforward and proceeds as
27 follows. The temperature is first set equal to the liquidus temperature corre-
28 sponding to the alloy composition. It is then decreased by a small decrement
29 and the equilibrium is calculated. The alloy composition is then set equal to
30 the liquid composition, the temperature decreased again by a small decre-
31 ment and the equilibrium calculated again. This process is iterated until no
32 liquid is found at equilibrium.
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43 For fast diffusing atoms in the aluminum solid solution, like magnesium,
44 silicon, copper, zinc and lithium, a more accurate treatment consists in eval-
45 uating, at each iterative step “s”, the amount of back-diffusion, i.e., solute
46 redistribution, occurring in solid solution[34]. This is done by using a finite
47 difference scheme, by assuming local equilibrium in each volume element of
48 secondary dendrite arms, by calculating the solute concentration in solid so-
49 lution in each volume element (i.e. applying the phase diagram subroutine
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for the current temperature and the local alloy concentration) and by solving Fick’s laws. For each time step, the finite difference routine performs many independent equilibrium calculations, one for each volume element, and is very well suited for parallelization.

The simulation of a laboratory heat treatment at 472°C of a multicomponent aerospace 7449 alloy (an Al-Mg-Zn-Cu-Fe-Si alloy see Table 2) after casting has been used to measure the gain in speed provided by OpenCalphad when parallelization is activated. The cast was performed in laboratory using the Aluminum Association TP-1 grain refiner test crucible. The corresponding cast structure, as shown in Fig. 2, was simulated using the finite-difference routine described above. For that particular case, a cylindrical geometry was chosen with a diameter of 61 μm corresponding to spacing between secondary dendrite arms. Care must be taken to mesh the dendritic cylinder with sufficiently small enough radius increments especially towards the end of solidification where the concentration gradients may be very steep.

Table 2: Composition range of AA7449 according to Aluminium Association

	Si	Fe	Cu	Mn	Mg	Zn	Ti+Zr	rest
min	0	0	1.4	0	1.8	7.5	0	Al
max	0.12	0.15	2.1	0.2	2.7	8.7	0.25	Al

The evolution of composition gradients has been studied for a heat treatment consisting in a linear heating up of 40°/h up to 472°C followed by a holding of 10h at 472°C. The corresponding predicted and measured incipient melting temperatures and integrated enthalpies for the DSC first melting peak are compared in Fig. 3. Overall, a good agreement is obtained indi-

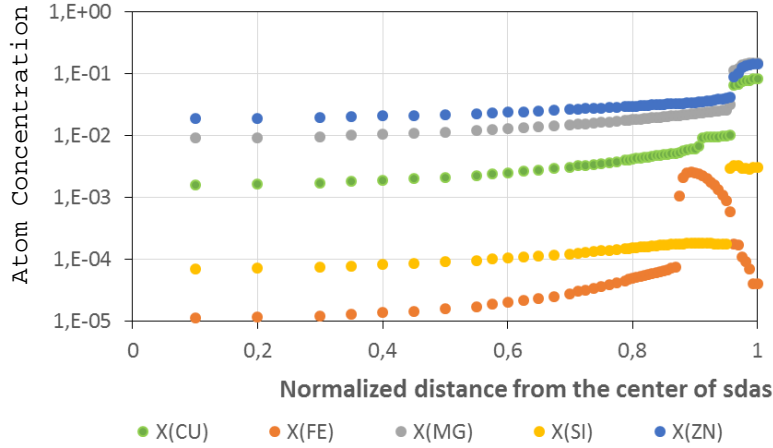


Figure 2: Simulated concentration gradients after solidification of aerospace alloy AA7449

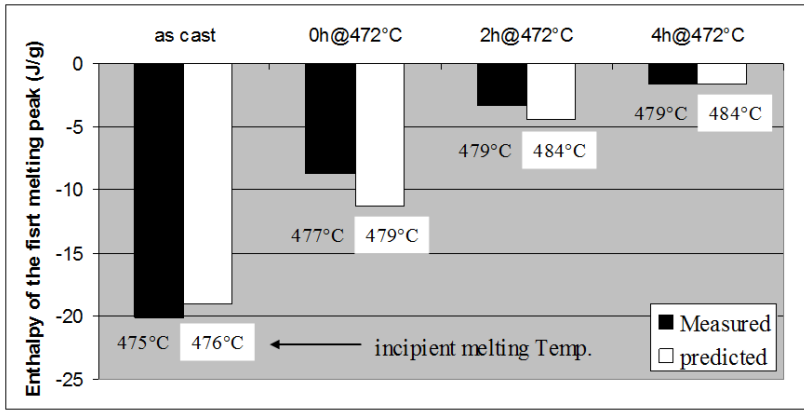


Figure 3: Simulated and predicted incipient melting temperatures and enthalpies of the first melting peak during calorimetric heat up at a rate of 20 °C/min.

cating that the proposed physical description of the phenomena occurring during solidification and homogenizing is acceptable.

This calculation was accelerated by a factor of 2.9 (from 239s to 82s) when

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9 using 4 cores instead of 1 (processor was Intel(R) i7-2760QM @2.4GHz).

12 4.3. Phase-field models and OCASI

14 Many modern thermodynamically consistent phase-field models [35, 36,
15 37, 38] rely on thermodynamic and kinetic properties that can be obtained
16 or approximated from CALPHAD calculations. OC as an open source tool
17 is perfectly suited for this purpose. The speed of the OCASI/C++ interface
18 with additional parallelization allows for an efficient way to compute separate
19 phase properties in the diffuse interfaces with equilibrium calculations or
20 with separate phase compositions as input parameters. Coupled with the
21 phase-field software framework, OpenPhase (OP), has been proven to be an
22 efficient tool for mesoscale micro-structure simulations. As extensive phase
23 data can be shared via this interface too, a user friendly simulation setup
24 can be arranged.

25 The equations governing phase transformations and diffusion in the phase-
26 field model with finite interface dissipation [38], require values of the molar
27 Gibbs energies of the phases, G_m^α , their derivatives with respect to the phase
28 constitution, $\partial G_m^\alpha / \partial y_{is}^\alpha$ and the atomic mobilities M_i^α . These can be calcu-
29 lated for a local equilibrium by the OCASI software by providing the values of
30 the temperature T , pressure P and overall composition x_i .

31 Parallelization of the code does not only speed up the simulations and
32 thus increasing the accuracy of the results, it also allows for a quick simula-
33 tion setup of multicomponent, multiphase simulations, as the OP software is
34 independent of the thermodynamic modeling details of the phases.

35 As an example we simulate the diffusion process between two austenitic
36 steel samples with different alloy composition. The two samples, containing

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9 iron, manganese, silicon and carbon, are chosen to represent the uphill diffu-
10 sion experiment by Darken[39]. Apart from the chosen thermodynamic and
11 kinetic databases [40] under the Open Database License, only the tempera-
12 ture, the time step and the grid spacing had to be specified as in Tab. 3
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18 Table 3: Simulation parameters

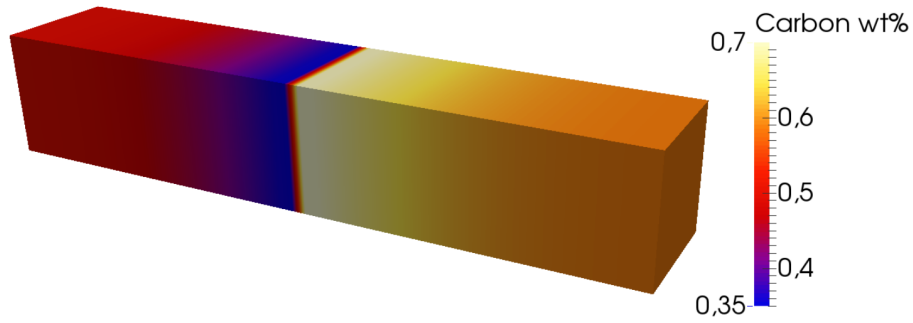
19	Box size	50x10x10
20	Grid spacing	0.001 m
21	Time step	1000 s
22	Temperature T	1323 K
23		
24	Thermodynamic database	MatCalc mc_fe_v2.057.tdb [40]
25	Mobility database	MatCalc mc_fe_v2.008.ddb [40]
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36 The results of the quaternary simulation for the duration of 10 days at
37 constant 1323 K are shown in Fig. 4 and are compared to the results of a
38 DICTRA sharp-interface simulation as well as measurements from [39]. The
39 results obtained with only openly available tools and data show excellent
40 agreement with the results from the commercial DICTRA software[13].
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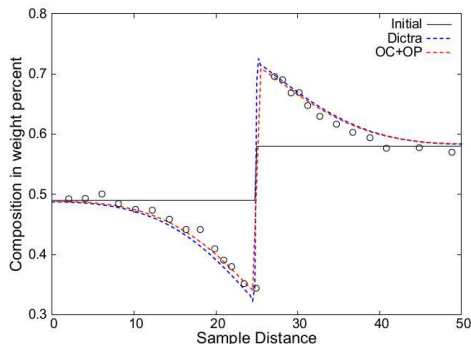
45 Beyond that, the coupling of OC and phase-field software like OP has
46 been proven to be an efficient, accurate and easy to use tool for mesoscopic
47 micro-structure simulations.
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51 52 *4.4. Mesoscopic modeling of a liquid miscibility gap*

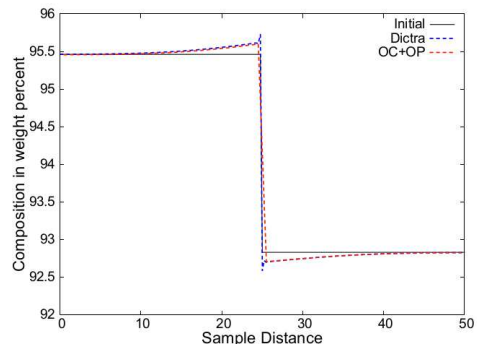
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54 For the study of severe accidents in light water nuclear reactors (LWR),
55 the OpenCalphad software has been used in the development of a Cahn-
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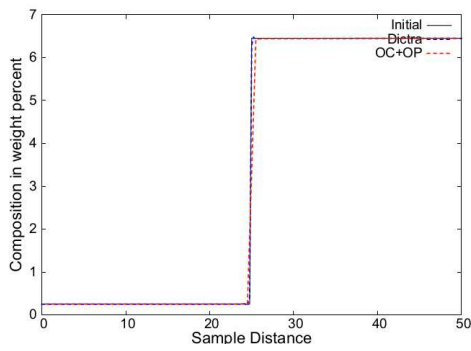
(a) Carbon composition over the whole simulation sample



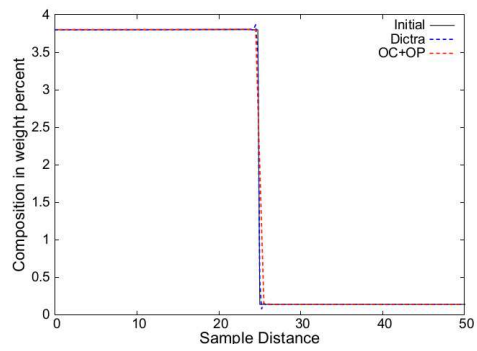
(b) Carbon composition



(c) Iron composition



(d) Manganese composition



(e) Silicon composition

Figure 4: Simulated composition profiles after 10 days of tempering at 1050 °C, with experimental measurements by [39] as circles.

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9 Hilliard based diffusion model for treating the liquid-liquid miscibility gap
10 associated with the corium (oxidic and metallic liquid materials present dur-
11 ing a reactor core meltdown) in a reactor vessel lower head.
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15 In-vessel corium is a complex thermodynamic system because of the
16 phases that are present. In addition to partially oxidized cladding and
17 fuel materials (UO_2 , ZrO_2 and Zr), the corium contains stainless steel el-
18 ements (Fe , Ni , Cr) from the reactor's internal structures. In particular, the
19 U-O-Zr-Fe system exhibits a liquid-liquid miscibility gap responsible for a
20 liquid-phases stratification of tremendous importance on the corium propa-
21 gation [41].
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28 This stratification phenomenon results from the combination of multi-
29 component multiphase species diffusion and gravitational material move-
30 ment due to buoyancy effects initiated by Rayleigh-Taylor instabilities. The
31 natural convection in a corium pool can be simulated by Computational
32 Fluid Dynamics (CFD) and Cahn-Hilliard based mesoscopic modeling of the
33 species diffusion appears to be a promising way to account for the strati-
34 fication phenomenon in detailed simulations. This R&D effort regarding a
35 detailed modeling of the thermohydraulic/thermochemical behavior of in-
36 vessel corium has been undertaken at CEA Cadarache in support of integral
37 models development in the context of the PROCOR software platform for
38 sensitivity/uncertainty studies regarding corium propagation.
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49 A Cahn-Hilliard based diffusion model was first developed for the U-O
50 binary case [30] and is under development for U-O-Zr and U-O-Zr-Fe. Un-
51 der the assumption of constant molar volume, the model for a n-component
52 system is based on Cahn-Hilliard evolution equations for molar fractions of
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n-1 components with diffusion potentials, $\tilde{\mu}_i$, and is expressed as:

$$\tilde{\mu}_i = a \left(\frac{\partial \tilde{g}}{\partial x_i} \right)_{x_j \neq i} - \sum_j \kappa_{i,j} \nabla^2 x_j \quad (18)$$

where a and $\kappa_{i,j}$ are parameters of the model related to the interface thickness (a “numerical” parameter in this mesoscopic modeling) and interface tension while \tilde{g} is the free energy density (in Jm^{-3}) of the homogeneous system.

In the binary U-O case, the $\left(\frac{d\tilde{g}}{dx_O} \right)$ term was calculated applying the chain rule to the liquid Gibbs energy G_m^{liq} and its derivatives $\partial G_m^{liq} / \partial y_i^{liq}$ obtained from the OCASI interface [30].

For the ternary and quaternary systems, the additional hypothesis that local equilibrium of the redox chemical reactions is instantaneously reached is made. Therefore, $\left(\frac{\partial \tilde{g}}{\partial x_i} \right)_{x_j \neq i}$ can be related to the chemical potentials obtained from “local” equilibrium calculations performed using the OCASI interface where the phase separation associated with the miscibility gap is not taken into account (*i.e.* the grid minimizer is turned off, *cf.* Section 2.3).

In order to illustrate this modeling, a 1D calculation for the U-O binary case is presented here; the domain length is $L = 10.0$ cm and the temperature and pressure conditions are $T = 3200$ K and $p = 1$ bar. Fig. 5 schematically depicts the initial configuration and the steady-state of the system. The interface thickness has been set to 1 cm.

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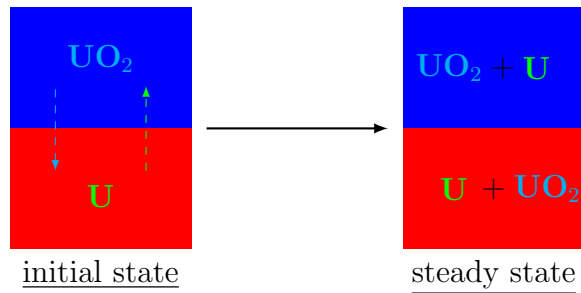


Figure 5: U-O system initial configuration and steady state

The initial configuration corresponds to an oxidic phase (uranium dioxide UO_2) above a metallic phase (in this case metallic uranium U). The diffusion process leads to a steady state configuration with two suboxidized phases. Fig. 6 shows the profile of oxygen molar fraction x_O in the initial state and at steady state where it is verified that, far from the interface, the oxygen molar fraction is equal to the values given by an equilibrium calculation.

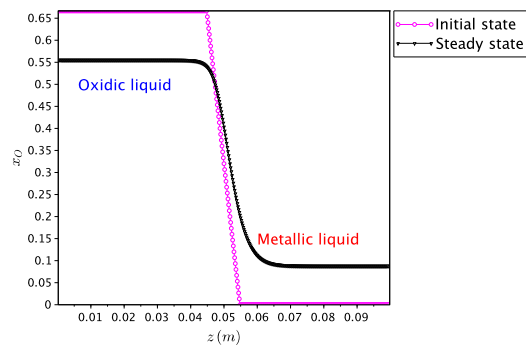


Figure 6: Profile of molar oxygen fraction x_O versus position z

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5. Summary

Thermodynamic data needed in simulations, such as melting temperatures, solubilities, heat capacities etc., are often collected from different sources. In many cases little or no efforts are made to verify that these data are consistent. Data from calculations using thermodynamic databases have the advantage of providing consistent data but the fact that millions of equilibrium calculations that are needed in simulations takes considerable time poses a severe problem.

The possibility to calculate the local equilibrium for each point in space in parallel is a major improvement. Still, it may not be possible to completely avoid the use of interpolation methods in very large simulations with several thousand grid-points. In this case, the availability of parallel processing offers the the opportunity to update the results in real time during the simulation avoiding the need for pre-calculated look-up tables and thus allowing more accurate thermodynamic input for the simulation. The flexibility of software has been demonstrated with selected examples from very different application fields.

6. Acknowledgement

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Appendix

Available state variables and their representation in OC.

All of them can be used to extract calculated values and many of them also to set conditions. For example N is the total amount in moles, $N(O)$ is the total amount of oxygen in moles, $N(\text{GAS},O)$ is the amount of moles of O in the gas phase, $NM(\text{GAS},O)$ or $X(\text{GAS},O)$ is the mole fraction of O in the gas. The total number of moles of components in gas is $NP(\text{GAS})$.

The heat capacity is not in this list but can be calculated as a “dot derivative, H.T” , as explained in [22].

Symbol	Index		Normalizing suffix	Meaning
	1	2		
Intensive properties				
T	-	-	-	Temperature
P	-	-	-	Pressure
MU	component	-/phase	-	Chemical potential
AC	component	-/phase	-	Activity
LNAC	component	-/phase	-	LN(activity)
Extensive and normalized properties				
U	-/phase#set	-	-	Internal energy for system
UM	-/phase#set	-	M	Internal energy per mole atoms
UW	-/phase#set	-	W	Internal energy per mass
UV	-/phase#set	-	V	Internal energy per m ³
UF	phase#set	-	F	Internal energy/mole formula unit
Sz	-/phase#set	-	-/M/W/V/F	Entropy
Vz	-/phase#set	-	-/M/W/V/F	Volume
H _z	-/phase#set	-	-/M/W/V/F	Enthalpy
A _z	-/phase#set	-	-/M/W/V/F	Helmholtz energy
G _z	-/phase#set	-	-/M/W/V/F	Gibbs energy
NP _z	phase#set	-	-/M/W/V/F	Moles of phase
BP _z	phase#set	-	-/M/W/V/F	Mass of phase
Q _z	phase#set	-	-/M/W/V/F	Stability of phase
DG _z	phase#set	-	-/M/W/V/F	Driving force of phase
N _z	-/phase#set/comp	-/comp	-/M/W/V/F	Moles of component
X	phase#set/comp	-/comp	-%	Mole fraction/per cent
B _z	-/phase#set/comp	-/comp	-/M/W/V/F	Mass of component
W	phase#set/comp	-/comp	-%	Mass fraction/per cent
Y	phase#set	const#subl	-	Constituent fraction
Some model parameter identifiers				
TC	phase#set	-	-	Curie temperature
BMAG	phase#set	-	-	Aver. Bohr magneton number
MQ&X	phase#set	constituent	-	Mobility of X
THET	phase#set	-	-	Debye temperature