18-month Post-Doctoral Fellowship Proposal:

Diffuse interface modeling of multicomponent multiphase mass transfer in buoyancy-driven flows and application to in-vessel corium pool behavior

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1. Context

This post-doctoral research work is related to the study of severe accidents in light water reactors (LWR) for the improvement of their prevention and/or mitigation. Within this framework, a key element is the phenomenology associated with a corium pool (oxidic and metallic liquid materials coming from a reactor core meltdown) that can be formed after the loss of primary coolant and the induced core degradation. For instance, for an "in-vessel retention" safety approach where it is the second barrier (*i.e.* the vessel) that is intended to contain the corium, the heat flux from the corium pool to the vessel wall determines the chances of success of such a strategy by the reflooding of the reactor pit ("External Reactor Vessel Cooling" ERVC) [1, 2].

The behavior of a corium pool results from the combination of two main types of phenomena. Firstly, the associated thermochemistry defines the segregation of the pool in different phases (both liquid and solid, oxide and metal), secondly, thermalhydraulics determines through natural convection (that may be laminar or turbulent depending on the size of the pool) the heat flux at the pool interface. In-vessel corium is a complex thermodynamic system in terms of the phases present as, in general, in addition to partially oxidized cladding and fuel materials (UO_2, ZrO_2 and Zr), the corium contains stainless steel elements (Fe, Ni, Cr) from the reactor internal structures. In particular, the miscibility gap in the in-vessel corium thermodynamic system leads, at thermodynamic equilibrium, to a gravity-induced segregation into two liquid phases (one "metallic" and one "oxidic"). One important feature of the stratification phenomenon is that the metallic phase can be heavier or lighter than the oxidic one depending, in particular, on the system overall composition. This post-doctoral work will be focused on this feature of corium thermochemistry; more details on other thermochemistry-related issues of in-vessel corium can be found in [3]. The stratified configuration of the pool associated with this thermochemical interaction has a large impact on the heat flux at the pool boundary with the vessel wall: indeed, the metallic layer on top of the pool can lead to a so-called "focusing effect" of the heat flux on its lateral surface that is a main threat for the vessel wall integrity. For more details on this effect and its modeling in severe accident scenario codes, the interested reader is refered to [4] and the references therein.

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Within this framework, this research work is related to the numerical simulation of the thermal-hydraulic behavior of the multiphase (oxide-metal) in-vessel corium pool with a special interest on the stratification kinetics. This work will be based on a "Computational Fluid Dynamics" (CFD) approach of thermal-hydraulics. Indeed, as shown by recent research activities [5, 6, 7], with the increase in computing resources and the maturation of turbulence models, CFD simulations of natural convection appear as valuable for assessing specific in-vessel corium thermalhydraulic phenomena and obtaining quantitative results that could then be used to improve integral code models.

The objective of this work is to enhance the scope of such CFD simulations by taking into account the stratification phenomenon related to both the species diffusion and thermochemical interaction between the liquid phases and the gravitational material movement due to buoyancy effects. In order to do so, the diffuse interface model proposed in C. Cardon Ph.D. research work (see [8, 9]) will be considered. This Cahn-Hilliard model is presented in Section 2 while the coupling with fluid dynamics equations, the main research avenue of this post-doctoral proposal, is discussed in Section 3.

2. Isothermal Cahn-Hilliard model

Under uniform and constant temperature T, this model proposed by C. Cardon (see [8, 9]) describes the multiphase diffusion for multicomponent systems related to in-vessel corium using a diffuse interface formalism based on a Gibbs free energy functional (the natural thermodynamic potential for materials whose density change with pressure is neglected, see [10]). It can be classified as a phase-field type model and the reader is refered to [11] for a general review on this modeling approach. In this model, the molar volume (denoted V_m) is assumed to be constant.

For a $n-$component system, the dependent variables $\underline{\phi} = \{\phi_i\}_i$ of this model are $(n-1)$ component molar fractions $\{x_i\}_{i\in\mathcal{E}}$; for the quaternary system U-O-Zr-Fe, in practice, $\mathcal{E} = \{U, Zr, Fe\}$ has been used. For a closed system, these quantities obey conservation laws in such a way that, considering a diffuse interface modeling, an adequate choice of kinetic equation is the Cahn-Hilliard (C-H) equation. In this framework, the fourth-order set of partial differential equations to be solved consists in: $\forall i \in \mathcal{E}$, $\forall \vec{r} \in V$

$$\frac{\partial \phi_i}{\partial t} = \nabla \cdot \left(\sum_{j\in\mathcal{E}} \mathcal{M}_{i,j} \left[\underline{\phi}\right] \nabla \tilde{\mu}_j \left[\underline{\phi}\right] \right) \tag{1}$$

$$\tilde{\mu}_i \left[\underline{\phi}\right] = \frac{a}{V_m} \frac{\partial \tilde{g}^{liq}}{\partial \phi_i} \left[\underline{\phi}\right] - \sum_{j\in\mathcal{E}} \kappa_{i,j} \Delta \phi_j \tag{2}$$

supplemented by homogeneous Neumann conditions for ϕ_i and $\tilde{\mu}_i$ on ∂V.

The kinetic coefficients $\mathcal{M}_{i,j}\left[\underline{\phi}\right]$ depend on the order parameters (so called "degenerate mobilities") under the form: $\forall i, j \in \mathcal{E}$,

$$\mathcal{M}_{i,j}\left[\underline{\phi}\right] = V_m \sum_{k=\{U,Zr,Fe,O\}} \left[\delta_{j,k} - \phi_j\right] \left[\delta_{i,k} - \phi_i\right] x_k M_k \tag{3}$$

where the atomic mobilities M_k are obtained, in practice, from data on self-diffusion D_k coefficients through the Einstein relation $D_k = RTM_k$. Such a form is consistent with the linear flux-force theory postulated by L. Onsager (see [12]) and ensures that $\underline{\phi} = \{\phi_i\}_{i\in\mathcal{E}}$ will remain in $[0,1]^{n-1}$ and that $1 - \sum_{i\in\mathcal{E}} \phi_i \in [0,1]$ at all time provided that the initial state complies with these constraints. *A fortiori*, consistency with the sub-system limiting cases is guaranteed.

The diffusion potentials $\tilde{\mu}_i\left[\underline{\phi}\right]$ are obtained as functional derivatives of the following Gibbs free energy functional:

$$\mathcal{F}\left[\underline{\phi}\right] = \int_V \frac{1}{2} \sum_{i\in\mathcal{E}} \sum_{j\in\mathcal{E}} \kappa_{i,j} \nabla\phi_i \cdot \nabla\phi_j + a\tilde{g}^{liq}\left[\underline{\phi}\right] \, dV \tag{4}$$

where:

- the homogeneous free energy density $\tilde{g}^{liq}\left[\underline{\phi}\right]$ (in J/m^3) is obtained, under a local equilibrium hypothesis of the redox reactions, from the CALPHAD data $\mathcal{G}_m^{liq}$, the Gibbs energy per mole of components for the liquid phase (in J/mol), a function of the species molar fractions $\{y_i\}_{i\in\mathcal{S}}$. More precisely,

$$\tilde{g}^{liq}\left[\underline{\phi}\right] = \frac{1}{V_m}\mathcal{G}_m^{liq}\left(\left\{y_i^{loc,eq}\right\}_{i\in\mathcal{S}}\left[\underline{\phi}\right]\right) \tag{5}$$

where

$$\left\{y_i^{loc,eq}\right\}_{i\in\mathcal{S}}\left[\underline{\phi}\right] = \underset{\{y_i\}_{i\in\mathcal{S}}}{\arg\min}\left(\mathcal{G}_m^{liq}\right) \text{ subject to } \sum_{i\in\mathcal{S}} y_i = 1 \text{ and } x_i\left(\{y_i\}_{i\in\mathcal{S}}\right) = \phi_i \tag{6}$$

The last constraint in the optimization problem Eq. 6 simply states the conservation of the element inventory as imposed by $\left[\underline{\phi}\right]$;

- a is an upscaling parameter;
- $\kappa_{i,j}$ is the energy coefficient associated with the interfacial gradient term $\nabla\phi_i \cdot \nabla\phi_j$.

In C. Cardon work, a "mock-up" of this model has been developed and tested in 1D slab geometries based on some of the PROCOR platform [13] functionalities, in particular, its interface with the CALPHAD numerical tool Open-Calphad (see [14, 15]). While some questions have been left opened regarding the choice of the energy coefficient matrix $\kappa_{i,j}$, this work has shown encouraging results toward the simulation of in-vessel stratified corium pools that motivate this post-doctoral proposal.

3. Coupling with fluid dynamics equations under the Boussinesq approximation

As a first step in this research project, an advective term will be added to the previous Cahn-Hilliard model *i.e.* Eq. 1 will be modified as follows, $\forall i \in \mathcal{E}$, $\forall \vec{r} \in V$

$$\frac{\partial\phi_i}{\partial t} + \vec{v}\cdot\nabla\phi_i = \nabla\cdot\left(\sum_{j\in\mathcal{E}}\mathcal{M}_{i,j}\left[\underline{\phi}\right]\nabla\tilde{\mu}_j\left[\underline{\phi}\right]\right) \tag{7}$$

where $\vec{v}$ is the liquid velocity. This Cahn-Hilliard model will then be coupled with Navier-Stokes (N-S) hydrodynamics under the Boussinesq approximation (see the review in [16]) in such a way that Eqs. 7 and 2 will be supplemented by the following mass and momentum conservation equations: $\forall \vec{r} \in V$,

$$\nabla\cdot\vec{v} = 0 \tag{8}$$

$$\rho^{\text{ref}}\frac{\partial\vec{v}}{\partial t} + \vec{v}\cdot\nabla\vec{v} = -\nabla p + \nabla\cdot\eta\left[\underline{\phi}\right]\left(\nabla\vec{v} + (\nabla\vec{v})^T\right) + F_\sigma\left[\underline{\phi}\right] + \rho\left[\underline{\phi}\right]\vec{g} \tag{9}$$

with appropriate boundary conditions and where

- p is the pressure;
- $\rho\left[\underline{\phi}\right]$ (resp. $\eta\left[\underline{\phi}\right]$) is the liquid density (resp. the dynamic viscosity) depending on the local composition through $\left[\underline{\phi}\right]$;
- ρ^{ref} is a reference density;
- $F_\sigma\left[\underline{\phi}\right]$ is a force accounting for surface tension (see [17] for more detail).

The development of this modeling will be done in the existing NEPTUNE_CFD platform [18] that offers adequate numerical capabilities and software architecure to do so. In addition, NEPTUNE_CFD has already been used and partially validated for the simulation of natural convection in homogeneous corium or metallic pools (see [6]). In this context, the Cahn-Hilliard model of Eqs. 7 and 2 will be spatially discretized and implemented using the available NEPTUNE_CFD framework for finite volume schemes on unstructured grids. Regarding the time scheme, a semi-implicit scheme that decouples C-H and N-S equations may be used a starting point (see [16, 19]) and the C-H may be solved, as in [9] with a fully implicit scheme involving Newton-Raphson iterations.

In this context, the modeling and numerical aspects related to the different closure terms in the system of equation will have to be analyzed carefully in the verification and validation process of the model. In particular, the pending questions in [9] regarding the terms of the energy coefficient matrix $\kappa_{i,j}$ will be addressed.

When the isothermal coupled model is matured enough, this post-doctoral research work will focus on the extension to the anisothermal case in order to obtain a thermal-hydraulic modeling of in-vessel stratified corium pools. In order to do so, this post-doctoral research work will benefit from the outcome of a Ph.D. thesis untitled "Thermohydraulics/thermodynamical coupling methodology for the modeling of in-vessel corium" started in November 2016 by Vaishnvi Tiwari at CEA, LPMA. Indeed, in the framework of this Ph.D. work, the diffusive Cahn-Hilliard model proposed by C. Cardon will be extended to treat anisothermal configurations. Indeed, if the phase-field formalism can be applied (based on an entropy functional instead of an energy functional, see [20, 21]), the thermodynamic consistency in the coupling between the C-H equations and the thermal balance (written in enthalpy or internal energy in this case) is to be treated carefully.

Finally, in the framework of this post-doctoral research, the validity of the Boussinesq approximation for in-vessel stratified corium pool modeling could be evaluated by comparison with a variable density model (see, for instance, [22] for such a comparison on simple Rayleigh-Taylor instability configurations).

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