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### ► To cite this version:

Arthur Tourneix, Romain Le Tellier, Pascal Piluso, Cyrille de Saint Jean. Towards the improvement of liquid Gibbs energy assessment by the assimilation of surface tension data. Calphad 2024 - 51th International Conference on Computer Coupling of Phase Diagrams and Thermochemistry, May 2024, Mannheim, Germany. cea-04720675

**HAL Id: cea-04720675**

**<https://cea.hal.science/cea-04720675v1>**

Submitted on 3 Oct 2024

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# Towards the improvement of liquid Gibbs energy assessment by the assimilation of surface tension data

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**Key words:** Liquid-gas interfaces, Bayesian method, Liquid Gibbs energy, CALPHAD assessment

In the event of a severe nuclear accident such as the TMI2 or Fukushima disasters, the incapacity to cool off the reactor core has led to its meltdown (temperatures exceeding 2700K). The resulting liquid mixture, known as corium, is mainly composed of molten UO<sub>2</sub> fuel pellets, partially oxidized Zircaloy clads and other structural material (primarily, made of steel grades). The behaviour of this internally heated (because of the decay heat) liquid is made complex because of the multicomponent nature of the mixture. For instance, in a large composition range, it displays a miscibility gap with an oxide phase and a metallic phase that can be lighter or heavier depending on the conditions (e.g. Zirconium oxidation degree). To ensure the safety of reactors and prevent environmental contamination, it is essential to predict its propagation during the accident through numerical simulations (e.g. multiphase thermohydraulics). In this context, Calphad modelling of the underlying multicomponent systems is crucial in providing thermochemical data to provide closures for both thermochemical and thermalhydraulic models. However, due to the high temperatures involved, thermodynamic knowledge of the corium liquid phase is limited because of the lack of experimental data, such as calorimetry measurements.

Consequently, thermodynamic quantities (e.g. enthalpies, chemical potentials) evaluated from a Calphad Gibbs energy model outside of global equilibrium conditions are subject to large (albeit unquantified) uncertainties and the predictive capability of corium numerical models is reduced. This trend was clearly highlighted in recent work [1] where liquid-liquid interfacial energies for U-O and U-O-Zr-Fe systems were evaluated using a Butler model approach [2] and a Calphad Gibbs energy model of the liquid phase. The comparison between calculations considering, for the liquid phase, either a non-ideal associate model (NUCLEA database [3]) or a ionic liquid model (TAF-ID database [4]), has shown that the important differences in extrapolation of the chemical potentials out of global equilibrium conditions have a large impact on the calculated interfacial energies (up to a factor of six between the two databases).

In our current research, we aim to capitalize on the remarkable sensitivity of liquid interfacial energies to enhance the liquid model itself. More precisely, confronting the results of a Butler model with surface

tension measurements should provide valuable feedback on the parameters of the Gibbs energy model of the liquid phase, given its sensitivity.

This work presents advancements made to establish a proof of concept to integrate liquid-gas surface tension data into the assimilation process of Calphad database. The chosen system of reference is the binary copper magnesium used at the School for Advanced Thermodynamic Assessments (SATA). It is a well-known system with multiple experimental data including activities, mixing and formation enthalpies or equilibrium measurements. This system is also simpler with no miscibility gap in the liquid phase. In an effort to take into account uncertainties, this work is done in a rigorous Bayesian framework to assimilate those data [5].

## References

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## Biographical Note

Arthur Tourneix is a second-year Ph.D. student attached to the Grenoble-Alpes University and doing its thesis at

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