M. Eboli et al.

Implementation of the Chemical PbLi/Water Reaction in the SIMMER Code

Implementation of the Chemical PbLi/Water Reaction in the SIMMER Code

Marica Eboli[a,*], Nicola Forgione[a], Alessandro Del Nevo[b]

[a]*DICI – University of Pisa, Largo Lucio Lazzarino 2, 56122 Pisa, Italy*
[b]*ENEA FSN-ING-PAN, CR Brasimone, 40032 Camugnano (BO), Italy*

The availability of a qualified system code for the deterministic safety analysis of the in-box LOCA postulated accident is of primary importance. Considering the renewed interest for the WCLL breeding blanket, such code shall be multi-fluid multi-phase, shall manage the thermodynamic interaction among the fluids, and shall include the exothermic chemical reaction between lithium-lead and water, generating oxides and hydrogen. The paper presents the implementation of the chemical correlations in SIMMER-III code, the verification of the code model in simple geometries and the first validation activity based on BLAST Test N°5 experimental data. The validation activity will be based on a validation matrix, which will be based on the new experimental infrastructure LIFUS5/Mod3 installed at ENEA CR Brasimone.

Keywords: water, lithium-lead, SIMMER, chemical reaction, WCLL breeding blanket, safety

1. Introduction

The Water Cooled Lithium Lead blanket concept has been recently reconsidered as candidate breeding blanket for DEMO reactor [1]. Therefore, the safety issues connected with the postulated interaction between lithium-lead and water is a potential hazard that shall be considered in the analyses.

Past studies and experiments were carried out in US and EU to investigate and to characterize the lithium-lead alloy water interaction and to evaluate the safety of WCLL breeding blanket. Nevertheless, the numerical activities focused only on the chemical phenomena occurring during the interaction [2]-[4], neglecting the thermodynamic processes. R&D activities reported in Refs. [5] [6] and the experimental campaign performed in BLAST facility [7] evidenced that the interaction can be divided into a short-term process dominated by mixing and pressurization, and a long-term process dominated by the chemical aspects.

In view of these considerations, a numerical tool able to deal with water and liquid metals, to predict the evaluation of pressure and temperature trends during a postulated accidents, and to evaluate the hydrogen production due to the exothermic chemical reaction is of primary importance for performing deterministic safety analysis.

The aim of the paper is to describe the implementation of the chemical correlations between lithium-lead and water in SIMMER-III code. To address this objective, Section 2 briefly describes SIMMER code, then the implementation of the correlations are illustrates in Section 3, while Section 4 provides the verification and validation activities against BLAST Test No.5 experimental data. Finally, conclusions ad perspectives are illustrated in the final Section.

2. SIMMER code

SIMMER is a computer code developed to simulate core disruptive accidents in a liquid metal fast reactor [8]. SIMMER-III is the two-dimensional version of the code, multi-velocity field (up to eight, for distinguish liquid fuel, liquid coolant, droplets, liquid steel, gas, and so on), multiphase, multicomponent, Eulerian, fluid-dynamic code coupled with a neutronic kinetics model. The conceptual overall framework of the code is shown in Figure 1.
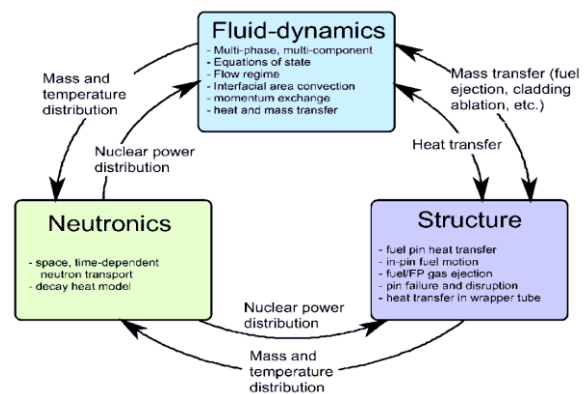

Fig. 1. Overall framework of SIMMER-III code

Components present in SIMMER can consist as either density and/or energy components. The mass and energy conservation equations are solved for each density and energy components in order to model complex flow situations during Fuel Coolant Interaction, which can describe also the phenomena occurring during the interaction between heavy liquid metals and water. A comprehensive and systematic program of code assessment for FCI was performed by Morita et. al [9] to validate the code under these key accident phenomena relevant for CDA. Recently, a code assessment campaign for the application of FCI to the interaction
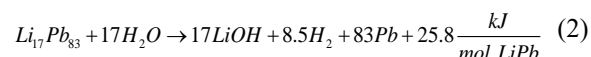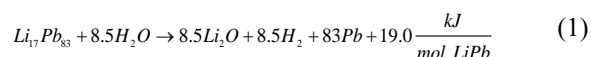
---

between HLM and water was performed by ENEA/UNIPI [10], [11].

Then, equation of state model is required to close and complete the fluid-dynamic conservation equations. The EOS are crucial from the viewpoints of numerical stability and computing efficiency. Especially the accuracy of the EOS plays an important role in FCI simulations, since the properties of the materials vary significantly.

3. Implementation of the chemical correlations

SIMMER-III code Ver.3F [12] is the reference code for the activity. In principle, it has the capability to simulate the chemical reaction between sodium and water.

The chemical reactions between PbLi and water implemented in the code and found in the literature [13] are:

$$Li_{17}Pb_{83} + 8.5H_2O \rightarrow 8.5Li_2O + 8.5H_2 + 83Pb + 19.0\frac{kJ}{mol\ LiPb} \quad (1)$$

$$Li_{17}Pb_{83} + 17H_2O \rightarrow 17LiOH + 8.5H_2 + 83Pb + 25.8\frac{kJ}{mol\ LiPb} \quad (2)$$

The predominant reaction is chosen by an user-dependent factor (FMOL) that varies from 0.5 (Eq. 2) to 1.0 (Eq. 1). Assigning an intermediate value that represents the conversion ratio of water into hydrogen, the model simulates both reactions. At each time step, the model checks what is the reactant in greater amount and takes the moles needed for the reaction. Then, it calculates the amount of the hydrogen production on the basis of the needed moles, the binary contact area between the two reactants and the rate of the reaction. As final step, the model calculates the residual reactant moles and checks if the reaction continues or not.

The implemented model assigns for each component a different material. Therefore, new thermodynamic properties for lithium-lead, lithium oxide, lithium hydroxide and hydrogen were added, even though in literature some information are missed. Available CEA data for thermodynamic properties and correlations are implemented [14].

4. Verification and Validation

In order to obtain reliable prediction of the thermo-hydraulic main parameters and the evolution of the transient during a postulated accident, numerical codes adapted to deterministic safety analysis must be validated. Therefore, the implemented chemical correlations underwent a phase of verification and first validation, which will continue in the forthcoming years.

4.1 Verification against simple geometry

According with IAEA glossary the verification step consists in the process of determining whether a computational model correctly implements the intended conceptual model or mathematical model [15].

Verification of the implemented correlations for the chemical reaction was performed, considering a simple 2D geometry. The model consists of three axial mesh cells and one radial mesh cell, as depicted in Figure 2. The lower cell is filled with water, covered by a certain amount of initial hydrogen. The upper cell is filled with lithium lead, separated by a virtual wall which opens at t = 0 s.

Two different calculations were performed, changing the predominant reaction, in one case considering the Eq. 1 and in the other case the Eq. 2.
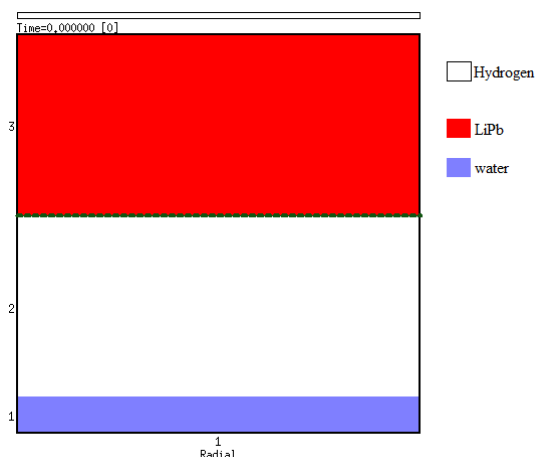

Fig. 2. Geometric model for the verification of the chemical correlations

The simulations were performed with the implemented SIMMER-III code. Nevertheless, using the standard post-processor, the hydrogen mass could not be visualized. In order to calculate the total mass of the components, and in particular the hydrogen mass, the BFCAL tool was needed [16]. This tool was developed to post-process the base-file of SIMMER computations. Indeed, it allows to obtain some additional parameters (such as the total mass, total energy, average temperature, location of the center of mass, etc…) in specified macro-regions. By using this tool, the total mass of the components at the start and at the end of the simulations can be calculated using the data results of the SIMMER calculations. The initial data of the simulations are reported in Table 1.

Table 1. Initial data of the simulations.

t = 0 s	PbLi	Water	H2
p (bar)	1	1	1
T (K)	623.15	343.15	343.15
m (kg)	150.80	3.068	0.0011
MW (g/mol)	173.1558	18	2
n (mol)	870.88	170.05	0.55

The rationale for the verification was:
- To calculate the theoretical stoichiometric hydrogen mass, starting from the initial data reported in Table 1. In Eq. 1, the ratio between moles of water and hydrogen is 1:1, while in Eq. 2, the ratio is 2:1. Once

calculated the moles of hydrogen produced in the reaction, the mass in kilograms was evaluated.
- To compare the theoretical value (summed to the initial hydrogen) with the hydrogen mass calculated by the implemented SIMMER-III.

In both calculations, the error was evaluated less than 0.6%. The theoretical (th) and calculated (SIII) results considering Eq. 1 and Eq. 2 are reported in Table 2.

Table 2. Comparison between theoretical and calculated hydrogen mass values considering Eq.1 or Eq.2.

t = 15 s	H2 th (1)	H2 SIII (1)	H2 th (2)	H2 SIII (2)
m (kg)	0.3412	0.3401	0.1712	0.1703

4.2 Validation on BLAST Test No. 5 experimental data

A preliminary validation against BLAST Test No. 5 was carried out considering the input of Ref. [17] and applying the implemented chemical correlations.

BLAST Test No.5 was chosen for performing the validation of the code considering the following rationale:
- quality of the documentation and data available,
- regular execution of the test and well defined simple and symmetric computational domain.

The SIMMER-III nodalization models the injector, the reaction vessel, the expansion tube, and the expansion vessel, as illustrated in Figure 3. The geometrical domain is composed by 16 radial and 73 axial mesh cells. The reference cells in which the pressure trends are considered correspond to the reference cells of the Ref. [17], i.e. cell [13,39] for the reaction vessel, and cell [13,73] for the expansion vessel. In this way, a better comparison of the results is provided.
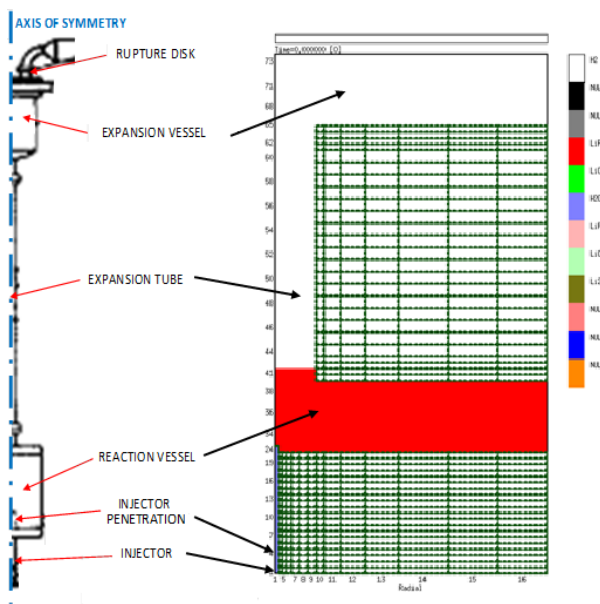
Fig. 3. BLAST facility nodalization by SIMMER-III code with the implemented chemical correlations

Hereafter, the code results of pressure trends of two simulations are presented. The simulations, carried out using the same nodalization, and set up with the same boundary conditions, were performed 1) not considering the lithium-lead water chemical reaction, as the current capabilities of the SIMMER-III code, and 2) using the implemented correlations.

Figure 4 shows the results where the chemical reaction is neglected. The code results, notwithstanding qualitatively similar with the experimental trends, appear largely underestimated. This can be explained to the capabilities of the code to perform thermo-hydraulic interaction between heavy liquid metals and water, but the largely underestimation is due to the underprediction of the energy in the system (i.e. the exothermic chemical reaction is neglected). The first pressure peak, predicted by the code in the reaction vessel, is 10 bar lower than experimental value. The predicted pressure in the system is stabilized at about 20 bar less than in the experiment.
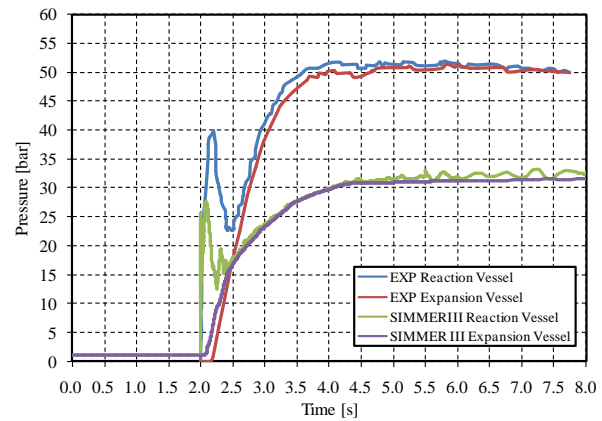
Fig. 4. Experimental and calculated pressure trends without chemical reaction

The experimental and calculated pressure time trends considering the implemented chemical correlations are shown in Figure 5. Results show that the simulation of the first peak is qualitatively and quantitatively well predicted. The pressure peak value in the reaction vessel reaches 40 bar, in line with the experiment. The code simulation predicts a pressure rise, which is slightly anticipated in time.

Considering the uncertainty in boundary conditions of experimental test, in particular of the mass flow rate and the total mass of injected water, it is challenging to demonstrate whether the differences between code simulation and experimental data are connected with the injection or the dynamic interaction between the fluids. It is expected that the chemical reaction should play a secondary role, besides the faster pressure increase will imply a faster hydrogen production rate. This will be addressed in the next experimental campaign on LIFUS5/Mod3, designed to code validation purposes, which will be provided qualified and reliable data, in particular concerning the injected mass of water, the hydrogen production, and pressure and temperature trends.

Nevertheless, SIMMER-III with the implemented chemical model simulates the pressure trends of the test from qualitative and quantitative point of view. Some numerical instability problems and failures of the code runs occurred during this preliminary validation of the code. These might be due to the rough properties implementation of the PbLi and its chemical products, which has to be improved.
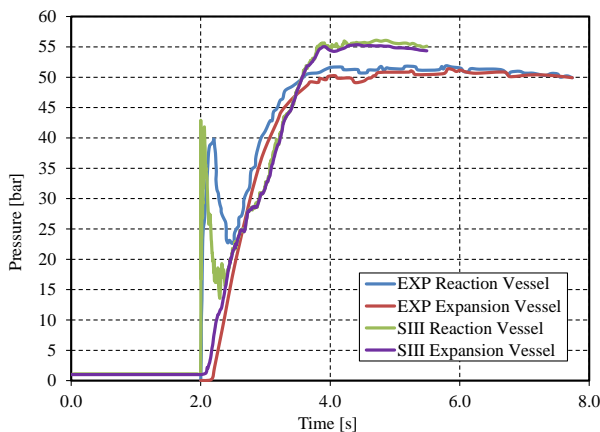

Fig. 5. Experimental and calculated pressure trends with implemented chemical correlations

5. Conclusions

The review of the literature and of numerical activities carried out in the past to characterize the lithium lead-water interaction, bring to the following conclusions:

• numerical tools developed in the past were focused on the chemical reaction neglecting the thermo-dynamic processes occurring during the interaction and affecting the short term period of the transient,

• SIMMER-III has the capability to deal with liquid metals and water interaction. Therefore, the implementation of the chemical correlations was of primary importance in view of deterministic safety analysis of WCLL breeding blanket.

Technical advancements are summarized as follows:

• the implementation of chemical correlations in SIMMER-III code to simulate the exothermic reaction between water and lithium lead, and the hydrogen production is concluded,

• the verification of the models in simple geometry and the first validation against BLAST Test No.5 experimental data were successfully completed,

• effect of uncertainties on implemented properties of lithium lead and its chemical products due to reactions will be investigated,

• the experimental data available in literature are few and not satisfactory to code validation due to uncertainties in I&B conditions. Therefore a new experimental campaign on LIFUS5/Mod3 is foreseen, providing reliable and qualified data suitable to code validation.

• once the validation of the code will be completed, SIMMER-III will be applied to DSA of water tube rupture accident scenario in WCLL blanket configuration, providing information on the design and the safety issues of this component.

Nomenclature

BLAST	BLAnkety Safety Test
BFCAL	Base File CALculation
CDA	Core Disruptive Accident
DEMO	DEMOnstration power plant
DSA	Deterministic Safety Analysis
EOS	Equation Of State
EU	European Commission
FCI	Fuel Coolant Interaction
HLM	Heavy Liquid Metal
LIFUS	LIthium FUSion
PbLi	Lithium-lead eutectic
R&D	Research and Development
SIMMER	S_n Implicit Multifield Multicomponent Eulerian Recriticality
US	United States of America
WCLL	Water Cooled Lithium Lead

Acknowledgments

References

[1] European Fusion Development Agreement, Fusion Electricity: A roadmap to the realization of fusion energy, EFDA 2012.

[2] M.L. Corradini, J.P. Herzog, Modelling of lithium lead/water interactions in a fusion reactor design, University of Wisconsin UWFDM-559, August 1984.

[3] J.P. Herzog, M.L. Corradini, Modelling of lithium lead / water interactions in a fusion reactor design: Part II modeling improvements, University of Wisconsin UWFDM-649, September 1985.

[4] P.O. Biney et al., A mass transport model for hydrogen generation during lithium-lead/water interactions, Proc. IEEE Meeting on Fusion Energy, Knoxville, October 1989.

[5] H. Kottowski, et al., Studies with respect to the estimation of liquid metal blanket safety, Fusion Eng. Des., 14 (1991), pp. 445-458

[6] H. Kottowski, G Kuhlbörsch, Li17Pb83 and Li7Pb2 water interaction, Programme Progress Report on Fusion Technology and Safety, CEC, JRC - Ispra, Italy, pp. 82-85, 1986.

[7] C. Savatteri, A. Gemelli, Lithium-lead/water interaction. Large break experiments, Fusion Eng. Des., 17 (1991), pp. 343-349.

[8] S. Kondo, et al., Status and achievement of assessment programme for SIMMER III a multiphase multicomponent code for LMFBR safety analysis, NURETH 8, 1997.

[9] K. Morita, et al., SIMMER-III applications to fuel-coolant interactions, Nuclear Eng. Des., 189 (1999), pp. 337-357.

[10] A. Ciampichetti, et. al., Experimental and computational investigation of LBE-water interaction n LIFUS5 facility, Nuclear Eng. Des., 220 (2003), pp. 240-254

[11] A. Ciampichetti, et. al., LBE-water interaction in LIFUS5 facility under different operating conditions, Journal of Nuclear Materials, Vol. 415 Issue 3 (2011), pp. 449-459.

[12] AA.VV., SIMMER-III (Version 3.F) Input Manual, O-arai Engineering Center, Japan Nuclear Cycle Development Institute, May 2012.

[13] K.A. McCarthy, Safety issues related to liquid metals, APEX meeting, Albuquerque, July 1998.

[14] C. Blanchard, Modelling of the Lithium-lead/water interaction, improvement of the kinetics of the pressure evolution, CEA H0-200-5010-3090.

[15] IAEA, IAEA SAFETY GLOSSARY – Terminology Used in Nuclear Safety and Radiation Protection – 2007 Edition, Vienna 2007

[16] AA.VV., *bfcal (Ver.4.5) manual*, May 2012.

[17] M. Eboli, et. al., Simulation study of pressure trends in the case of loss of coolant accident in Water Cooled Lithium Lead blanket module, Fusion Eng. Des., (2015).