Length of the extended abstract minimum 4 pages

Analysis of Multicomponent Diffusion Impact on Hydrogen Issues in Water Cooled Nuclear Reactors

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In water cooled nuclear reactors, hydrogen generation is an undesirable phenomenon, whose dramatic consequences have been touched in 2011 with the Fukushima accident occurred in Japan. After this disaster, improving safety strategies became really important in the scientific and industrial communities. Passive Autocathalytic Recombination (PAR), Fuel Cladding and the development of Computer Safety Models could be effective solutions to this problem, although they need a very reliable experimental database to be defined.

Such experimental investigations are particularly hazardous due to the very high hydrogen flammability, and accurate results are difficult to achieve. This is the reason why the role of Computational Fluid Dynamics is so important in this field, and significant efforts have been aimed at improving the predictive capabilities of the numerical models. These latter have to be very accurate to give sufficiently reliable predictions for design and safety purposes. Difficulties in meeting these requirements are represented by the need of analysing huge computational domains, for simulated time of the order of days or weeks, or the presence of many phenomena belonging to different space and time scales, e.g. condensation, diffusion, combustion and dispersion. Given this complexity, the best approach is to singularly calibrate the models with separate analysis instead of going for multi-physical models. The nuclear authorities agreed with this approach, and supported various experimental programs [1] focused on light gas stratification inside enclosures. The experimental program THAI carried out in 2009 is one of them.

The dispersion problem, so simplified, is regulated by turbulent mixing and molecular diffusion. The diffusion contribution is central in the present work, and its numerical modelling was refined to improve light gases concentration predictions. Indeed, the Fick's law is commonly applied in this kind of investigations, although it is acceptable only in case of dilute mixtures: the extension of this law to multispecies contexts requires a completely different treatment for the diluent species to satisfy the mass conservation constraint. However, the mixture cannot be view as dilute in presence of stratified layers of light gases and the above approach is meant to fail in such circumstances. More generally, this problem occurs when diffusion is modelled by assuming a diagonal diffusivity matrix.

The outlined limitation could be overcome with the use of detailed multicomponent diffusion models accounting for off-diagonal contributions, e.g. the Ramshaw model and the Stefan Maxwell equations. The name Ramshaw identifies a group of approximate models based on the projection of a diagonal diffusion approximation, so that the resulting model is able to satisfy the mass constraint. The Stefan Maxwell equations derive from the kinetic theory of gases and they then represent the most accurate diffusion treatment under the hypothesis of continuum. In the context of light gases dispersion, these equations have been simplified to neglect pressure and thermal diffusion contributions. Herein, attention is paid on cross diffusion, which is driven by differences in species concentrations.

The choice of the cross diffusion driving force is particularly critical in multispecies contexts since mass fraction gradients and molar fraction gradients are not interchangeable. The diagonal molar fraction formulation (Hireschfelder-Curtiss law) is better than the diagonal mass fraction one (Fick's law) as shown by Giovangigli [2], who developed the expansion series of the diffusivity matrix. Two Ramshaw models can be also formulated on mass fraction or molar fraction basis, according with the choice of the diffusion driving force.

The multicomponent diffusion analysis was carried out by following the structured approach depicted in Figure 1. After having investigated the mathematical properties of the above models, they were implemented in the OpenFoam 4.0 environment starting from a baseline solver, representing an implementation of the Fick's law with Lewis number different than one. While the implementation of the other models consisted in modifying the definition of the diffusion fluxes, the Stefan Maxwell model was more demanding as it required the regularisation of the diffusion matrix and an LU decomposition to be performed in each numerical cell.

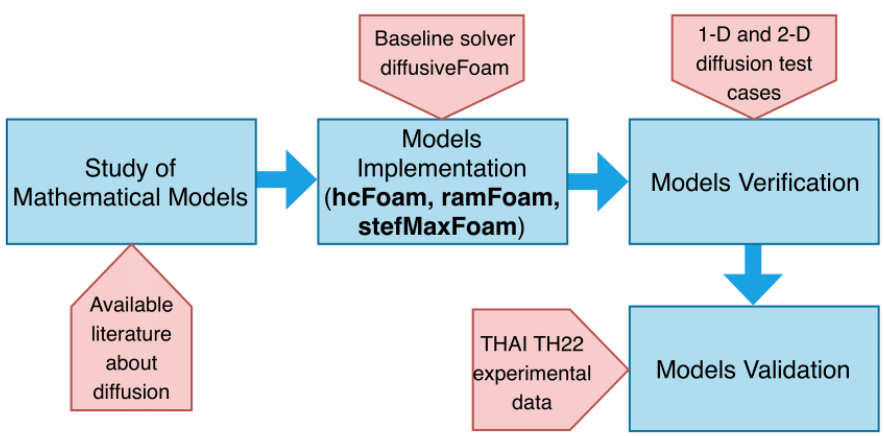

Figure 1 Project methodology.

Four verification test cases followed the implementation of the models to check that they effectively worked as expected. The first test involved a 1-D thin layer separating two regions at specified hydrogen and helium concentrations. The steady state solutions were obtained starting from a uniform species distribution, and they were then compared to the analytical helium distributions. As shown in Figure 2, the agreement was excellent with all the diffusion models.

Another 1-D test case was carried out to test the implementation of the Stefan Maxwell equations in OpenFoam 4.0 and highlight the inability of diagonal models to reproduce counter gradient diffusion that can arise during transients. The Duncan and Torr experiment was numerically implemented [3] as a one-dimensional problem, and the *uphill* diffusion of nitrogen occurring under these conditions was reproduced. The resulting counter gradient evolution represented in Figure 3 could not be captured by diagonal models as it is caused by multispecies interactions.

Table 1. Semi-discrete form of the diffusion fluxes accordingly with the diffusion model implemented in OpenFoam 4.0.

Model implemented	Temporal Discretisation
Fick's law	$\boldsymbol{J_i}^{n+1} = -(\rho D_i)^n \nabla Y_i^{n+1}$
Hireschfelder-Curtiss law	$\boldsymbol{J_i}^{n+1} = -\left(\rho D_i Y_i \frac{\nabla X_i}{X_i}\right)^n$
Ramshaw model (mass fraction basis)	$\boldsymbol{J_i}^{n+1} = -(\rho D_i)^n \nabla Y_i^{n+1} + Y_i^{n+1} \sum_{j=1}^{N} (\rho D_j)^n \nabla Y_j^n$
Ramshaw model (molar fraction basis)	$\boldsymbol{J_i}^{n+1} = -(\rho D_i)^n \left(\frac{\nabla X_i}{X_i}\right)^n + (\rho Y_i)^n \sum_{j=1}^{N} \left(\frac{Y_j D_j}{X_j}\right)^n \nabla X_j^n$
Stefan Maxwell equations	$\boldsymbol{J_i}^{n+1} = \left(\tilde{\Delta}^{-1}\right)^n (\rho Y_i)^n \nabla X_i^n$

Laminar mixing in a 2-D axisymmetric pipe was also analysed for verification purposes by implementing the conditions reported in [4]. The results were employed to compare the multicomponent diffusion approaches in terms of diffusion fluxes along the interface separating the equimolar mixtures of argon and hydrogen and argon and methane. While the Fick's law resulted completely unreliable, the other models (Hireschfelder-Curtiss law and Ramshaw models) tended to approach the Stefan Maxwell solution.

The last 2-D test case was developed to test the diffusion models in presence of a turbulent unsteady jet inside an enclosure [5]. Unsteady simulations with Stefan Maxwell diffusion required a very small time step size to satisfy the stability criterion, i.e. the Neumann number had to be lower than 0.25. These very severe time step limitations were avoided later on by applying the Ramshaw model at the first iteration of the PIMPLE algorithm, to work with a first guess of the solution when the Stefan Maxwell diffusion fluxes need to be explicitly calculated (see Table 1) in the successive outer iterations. This approach enabled to reduce the time step size by a factor of 40.

The experiment THAI TH22 performed in the THAI facility was chosen for the validation of the models. The experiment consisted in three main phases: during the first one, a stable natural convection flow field established inside the vessel represented in Figure 11, driven by a temperature gap of 80 degrees between the upper and the lower walls. The flow field was characterised by two counter rotating loops developing inside the vessel (Figure 4). This flow field was numerically reproduced starting from 2-D axisymmetric simulations (Figure 4), and then passing to 3-D simulations of the domain represented in Figure 5, given the significant 3-D effects created by the presence of the injector. In the frame of this analysis, two turbulence models modifications [6-7] were implemented and tested to account for anisotropic effects generated by gravity. The numerical model was able to capture the structure of the flow field, as shown in Figure 6 and 7 which represent the experimental PIV map of the velocity field and the numerical velocity field near the loops interface, respectively.

The natural convection flow field resulted from steady state simulations was considered accurate enough to move forward with the second phase of the experiment. This latter was characterised by the injection of helium in air for 216 s inside the THAI vessel and the generation of a stable light gas stratification at the top of the vessel. The Fick's law, the Ramshaw model and the Stefan Maxwell model were activated during this phase and the concentrations achieved at different measurement points were compared to the experimental profiles.

Good light gases predictions were obtained at most measurement points with inaccuracies that do not exceed the 5 %. Such inaccuracies can be primarily attributed to the geometrical details that are missing in the present model. Numerical simulations revealed also the presence of oscillations in time which can be related to the interaction of the helium jet with the upper natural convection loop. This interaction reflects into the periodical deceleration of the upper natural convection loop by the buoyant forces. This fluctuating behavior seems to not affect the experimental profiles since the oscillations were probably damped by the helium sampling line. The best agreement with the experimental profiles was achieved at the highest locations, where the helium concentrations were relatively high. At these points, the solution was also more sensitive to the choice of the diffusion model: the Stefan Maxwell model implemented in OpenFoam improved the comparison with the experimental data, as shown in Figure 8, 9 and 10. Indeed, the dilute mixture approximation does not hold in this region, where nitrogen cannot be view as the diluent and a detailed approach is thus able to better reproduce diffusion in the stratified layer of light gases.

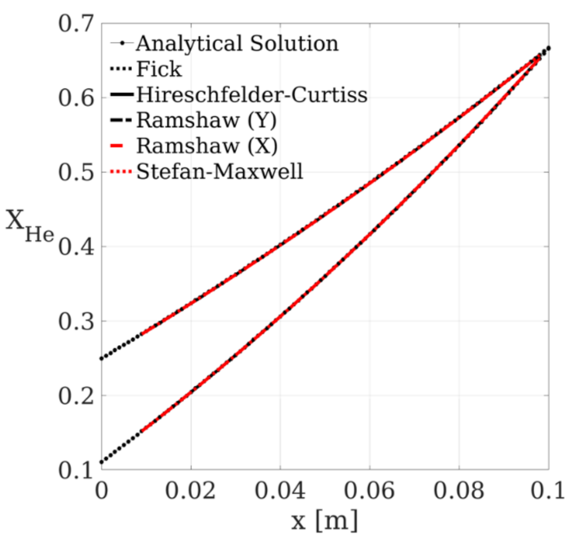

Figure 2 Comparison between the analytical and numerical solutions (first verification test case).

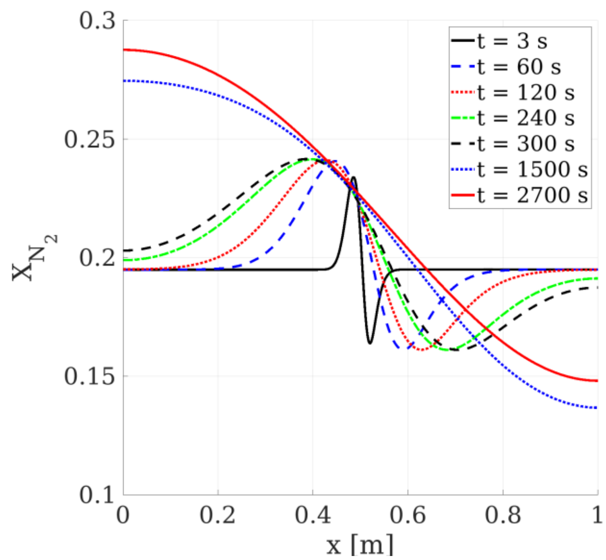

Figure 3 Counter gradient evolution of the nitrogen concentration obtained with the Stefan Maxwell model implemented in OpenFoam 4.0.

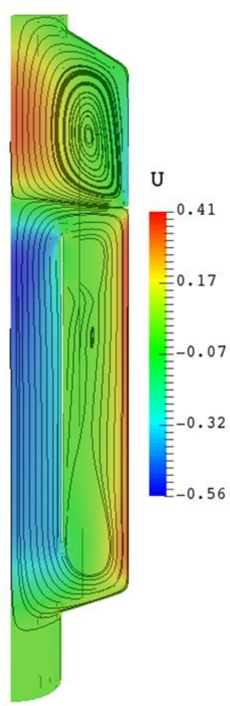

Figure 4. Velocity field establishing during Phase 1 inside the THAI vessel resulting from 2-D axisymmetric simulations

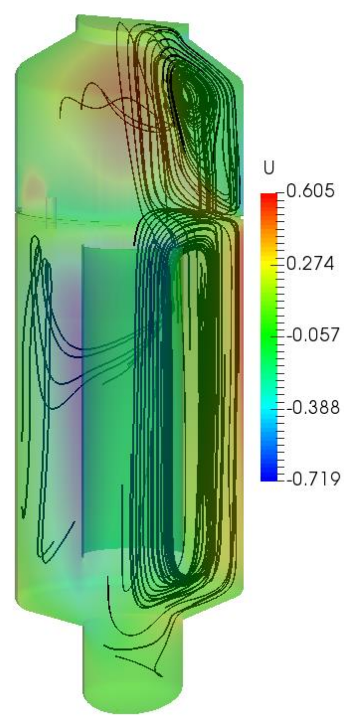

Figure 5. Preconditioned velocity field obtained from the 3-D simulation.

The third phase of the experiment is characterised by the erosion of the stable stratification achieved in the previous phase by means of the natural convection flow field and molecular diffusion. Therefore, the choice of the diffusion model is even more important in this phase to achieve very accurate results.

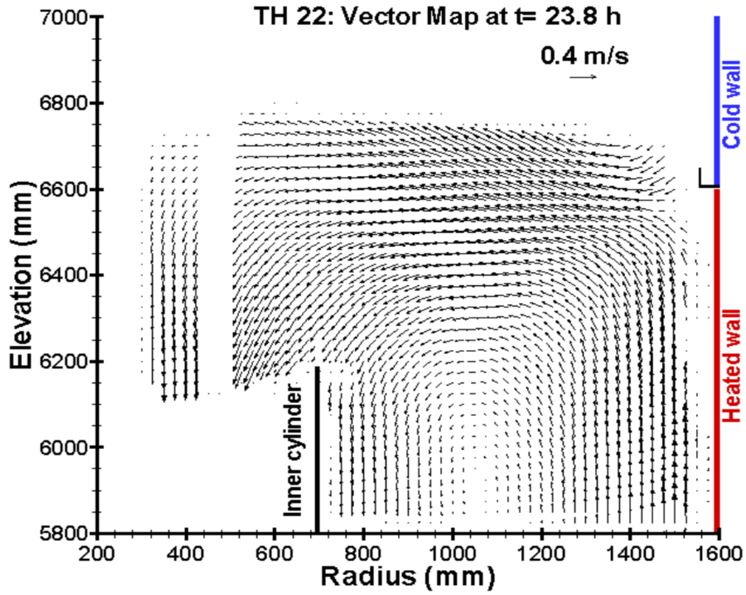

Figure 6. PIV map of the mean velocity field.

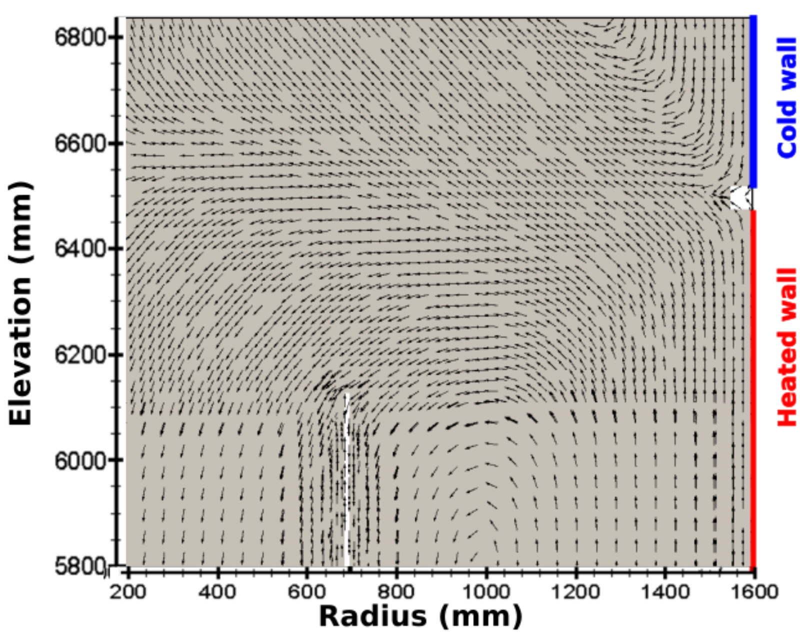

Figure 7. Velocity field obtained in 3-D simulations near the loops interface.

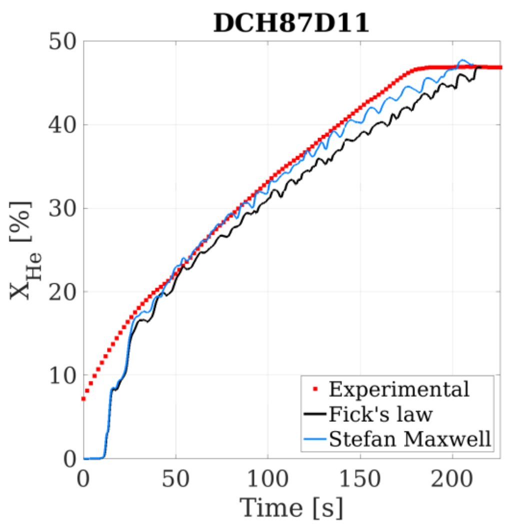

Figure 8. Helium concentration achieved over time inside the THAI vessel at the elevation of 8.7 m.

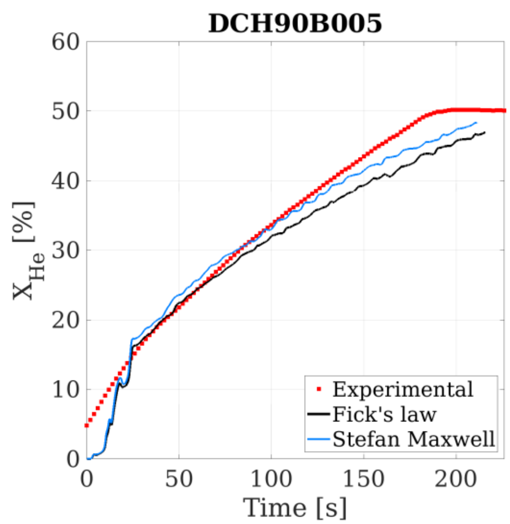

Figure 9. Helium concentration achieved over time inside the THAI vessel at the elevation of 9.0 m.

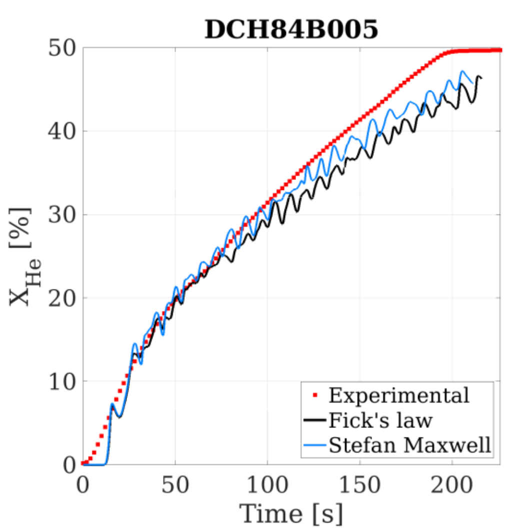

Figure 10. Helium concentration achieved over time inside the THAI vessel at the elevation of 8.4 m.

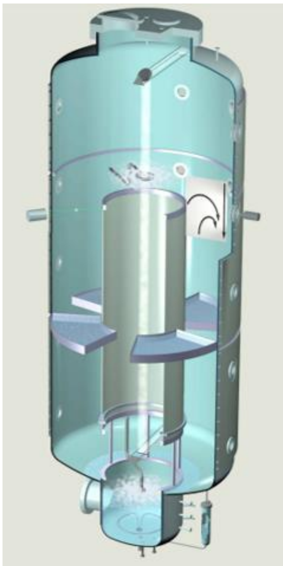

Figure 11. THAI vessel employed for the experimental campaign.

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