

# INTEGRATED CFD SIMULATION OF PASSIVE AUTOCATALYTIC RECOMBINER IN AN ENCLOSURE FILLED WITH HYDROGEN WITH TWO APPROACHES

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## ABSTRACT

In water power cooled reactors, significant quantities of hydrogen could be produced following a severe accident (loss-of-coolant-accident along with non availability of emergency core cooling system) from the reaction between steam and zirconium at high fuel clad temperature. In order to prevent the containment and other safety relevant components from incurring serious damage caused by a detonation of the hydrogen/air-mixture generated during a severe accident in water cooled power reactors, passive autocatalytic recombiners (PAR) are used for hydrogen removal in an increasing number of French, German and Russian plants. Numerical models were developed from the experimental data for codes like COCOSYS or ASTEC in order to optimise the passive autocatalytic recombiners location and to assess the efficiency of passive autocatalytic recombiners implementation in different scenarios. However, these models are usually simple (black-box type) and based on the manufacturer's correlation to calculate the hydrogen depletion rate. Recently, uses of enhanced CFD models have shown significant improvements towards modeling such phenomenon in complex geometry. The work presents CFD analysis of interaction of a representative nuclear power plant containment atmosphere with passive autocatalytic recombiners simulated using the commercial Computational Fluid Dynamics code for PAR Interaction Studies (PARIS benchmarks) exercise. A two-dimensional geometrical model of the simulation domain was used. The containment was represented by an adiabatic rectangular box with two passive autocatalytic recombiners located at intermediate elevations near opposite walls. In the first approach PAR volume has been modelled as fluid region and separate energy equation was solved through user coding for heat transfer to solid plate from reaction and from solid plates to fluid media. In the second approach PAR volume has been modelled as a porous media and pressure drop, heat generation and hydrogen conversion rate has been incorporated. The flow in the simulation domain was modelled as single-phase. The results of the simulations are presented and compared against reported simulation results.

## INTRODUCTION

During a severe accident in water cooled power reactor nuclear power plant, large amounts of hydrogen would presumably be generated due to metal oxidation during core degradation and released into the containment. The integrity of the containment could be threatened due to hydrogen combustion. Suitable mitigation means have been incorporated all over world to control the hydrogen concentration. Some methods for mitigation are: (a) deliberate ignition of hydrogen-air mixture; (b) inerting of hydrogen-air mixture by  $N_2/CO_2$  or by promoting mixing between the compartments by appropriate means; (c) catalytic recombination of hydrogen with oxygen from air; and (d) or a combination of these methods. Among these, the one based on oxidation of hydrogen in the presence of catalyst such as platinum/ palladium or the alloys of platinum/palladium, has proved to be most attractive [1]. The passive autocatalytic recombiner are passive devices, can remove hydrogen even in the presence of steam, at temperatures as low as ambient and at hydrogen concentration as low as 0.05 %v/v; and during the process of recombination, which is exothermic, it also promotes homogenisation of gas mixture due to buoyancy-induced gaseous flow.

However, a detailed analysis is required to optimize the passive autocatalytic recombiner locations and numbers in different compartment of containment. As a part of accident analysis various numerical model are used for assessment of passive autocatalytic recombiner. Modelling of passive autocatalytic recombiner requires modelling of process inside PAR (i.e. heat generation, buoyancy induced flow and hydrogen removal) and interaction of PAR with containment atmosphere (i.e. mixing). Recently use of enhanced Computational Fluid Dynamics (CFD) model has shown significant improvements towards modelling such phenomena. But integration of detailed catalytic recombiner modelling with hydrogen distribution CFD code is complicated. Since the mesh size requirement to fully resolve the recombiner plates is a very fine mesh size inside the recombiner channels (of the order of 0.1 mm) whereas for containment mesh size requirement is of the order of 100 mm. Another difficulty in modelling the recombiner in CFD code is the large number of recombiner placed in a nuclear containment, thus

geometry creation is itself a big task. Thus a simplified approach is required based on empirical correlation to model recombiner with containment CFD model. Experiments empirical correlations have been developed to compute the hydrogen removal rate of a particular make recombiner by various manufacturers to study the aspects like efficiency, start up condition, hydrogen conversion rate, effect of operating pressure, temperature and other inlet conditions. These correlations describe the hydrogen consumption rate for a reference PAR type as a function of gas composition, temperature and pressure. The hydrogen consumption rates proposed by the different manufacturers are given by the following empirical correlations [2].

$$\text{AECL} \quad m_{H_2} = (0.15196C_{H_2} + 0.0126C_{H_2}^2) \left(\frac{298}{T}\right)^{1.10974} p^{0.57769} \quad (1)$$

$$\text{AREVA} \quad m_{H_2} = \eta \cdot \min(X_{H_2}, 2X_{O_2}, 8)(Ap + B) \tanh(X_{H_2} - 0.5) \quad (2)$$

$$\text{NIS} \quad m_{H_2} = 1.134C_{H_2}^{1.307} \frac{p}{RT} \quad (3)$$

Where  $m_{H_2}$  is hydrogen consumption rates,  $X_i$  is concentration of  $i^{\text{th}}$  species in vol. %,  $C_i$  is volume fraction of  $i^{\text{th}}$  species,  $p$  is the absolute pressure of the mixture, in N/mm<sup>2</sup>,  $T$  is the absolute temperature of the mixture in K,  $A$  and  $B$  are factors that depends on the recombiner model (FR90/1-150, 320, 780, 960, 1500),  $\eta$  is the efficiency of recombiner under different conditions. As a simplified approach to integrate the recombiner model with containment CFD calculation, these correlations can be used by means of volumetric sink of hydrogen, oxygen and source of energy and water vapour in the CFD codes. Kudriakov et al. [3] have developed CFD code TONUS where they have modeled the recombiner with space averaged boundary condition at recombiner inlet and outlet. At the outlet of recombiner hydrogen mass fraction was calculated based on mass balance where hydrogen removal rate was calculated by manufacturer's correlation. Apart from this simplified model based on manufacturer's correlation are used in GASFLOW code developed by FZK [4]. A one-D detailed model was implemented in ASTEC [5] and COCOSYS [6] codes. A simplified recombiner model has also built in GOTHIC [7] CFD code. In GOTHIC the built-in PAR model burns a user-specified fraction of the hydrogen flowing through the PAR based on manufacturer's correlation. Flow through PAR was calculated by the code depending upon buoyancy driving force.

In the present work a PAR model is developed based on manufacture's correlation to integrate with commercial CFD codes used for hydrogen distribution modelling. The model has been used to simulate PAR Interaction Studies (PARIS-1) benchmark problem. PARIS-1 benchmark exercise has been organized by participants in the severe accident research network of excellence (SARNET) which is a part of the 6<sup>th</sup> EU framework program [8], in order to investigate the general adapted models and approaches for modelling PAR and focus on the impact of PAR elevation on the phenomenon of stratification. Some of the details of benchmark exercise and numerical results obtained by CFX are available in [9].

## DESCRIPTION OF THE PROBLEM

In the frame of SARNET WP12-2 (containment atmosphere mixing) PAR interaction studies were performed in order to generally investigate the adopted models and approaches. The first benchmark (PARIS-1) focused on the impact of PAR elevation on the phenomenon of stratification. As part of this study, a PAR of the AREVA FR90/1-150 design was considered in a 2D rectangular domain. Height of PAR ( $h$ ) was 1 m, and width ( $w$ ) was 0.2 m. PAR entry and exit section widths are also equal to 0.2 m. Each PAR has 15 autocatalytic plates with dimensions of 0.15 m x 0.15 m x 0.0001 m (height x depth x width), and an inter-space of 0.01 m. The containment is represented by square box, with side length of 5 m. There are two PARs located in the containment as depicted in Fig. 1. No heat or mass transfer (steam condensation) occurs on the walls. The starting point for the 3000 s calculation was a homogeneous mixture at 3.36 bar, 120 °C, containing 0.18 % hydrogen mass fraction and steam at saturation conditions. The main purpose of the first benchmark simulation was to observe the containment atmosphere mixing phenomena during a postulated severe-accident scenario.

## MATHEMATICAL MODEL

The 2D transient Navier-Stokes equation along with the energy and species transport was solved using commercial CFD software CFD-ACE+ [10]. To model the mixture behaviour of hydrogen, air and steam, a continuum approach was used, where only one velocity field was defined using the average density of gas mixture.

The independent behaviour of hydrogen, oxygen, nitrogen and steam was considered using species transport equation. The computational domain was divided in 100X100 grids. Two approaches have been used for modelling catalytic recombiner.

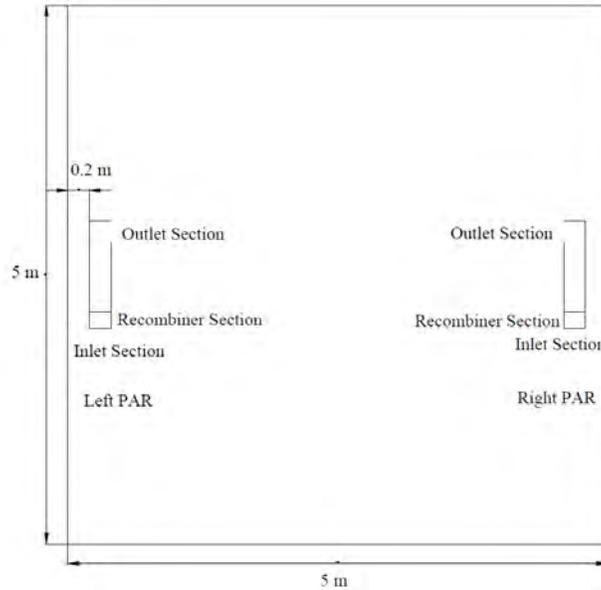


Fig. 1: PARIS-1 model geometry with 2 PARs

In first approach the recombiner section has been modelled as fluid medium. The pressure loss  $\Delta p/L$  across the catalyst plates was modelled as a sink term in momentum equation through user coding with the following equation

$$\Delta p/L = C_1 V + C_2 \rho V^2 \quad (4)$$

Where  $V$  is velocity of the mixture in m/s, and  $\rho$  is density of the mixture in  $\text{kg/m}^3$  and value of  $C_1$  and  $C_2$  was taken as constant as 3.154 and 1.739 respectively. A separate analysis was performed with actual recombiner geometry with plates and pressure drop was calculated for different velocities. The hydrogen conversion was modelled by means of the empirical AREVA correlation Eq. (2). Where the two parameters A and B have the values:  $A = 0.48 \times 10^{-8}$ ,  $B = 0.58 \times 10^{-3}$ . The high energy output from the hydrogen oxygen reaction results in a considerable heat transmission from surfaces by convection and radiation to environment. It was assumed that the entire heat generated because of recombination is taken by solid plates and then transferred to fluid in the recombiner section by natural convection. The following lumped equation for solid plate temperature equation was solved simultaneously in every time step.

$$m_s C_{p_s} \frac{dT_s}{dt} = m_{H_2} Q - hA(T_s - T_f) \quad (5)$$

Where  $m_s$  is mass of the catalytic plates in kg,  $C_{p_s}$  is specific heat of the plates in  $\text{J/kg/K}$ ,  $T_s$  is homogeneous temperature of the catalytic plates in K,  $t$  is time in s,  $Q$  is heat release during the catalytic reaction in  $\text{J/kg}$  of hydrogen burnt,  $h$  is heat transfer coefficient in  $\text{W/m}^2/\text{K}$ ,  $A$  is exchange area of the plates in  $\text{m}^2$ ,  $T_f$  is average fluid temperature at recombiner section in K. First the hydrogen conversion rate was evaluated based on the manufacturer's correlation then it was used to obtain the solid plate temperature with Eq. (5). The last term of Eq. (5) represents the heat convected by fluid mixture through the recombiner section and has been used as source term for energy equation solved for recombiner section. The heat transfer coefficient was calculated based on the empirical correlation for natural convection over vertical multiple plates. Based on the hydrogen conversion rate corresponding mass sink term for hydrogen and oxygen and mass source term for steam was incorporated through user coding.

In second approach the recombiner section has been modelled as porous media. The entire heat generated because of recombination is applied as a source term at porous region with porosity 0.9925. The pressure drop and heat transfer through porous medium was calculated by code itself. The corresponding sink term for hydrogen was

calculated by means of the empirical AREVA correlation. Based on the hydrogen conversion rate corresponding mass sink term for hydrogen and oxygen and mass source term for steam was incorporated through user coding.. All other modelling parameters were same in both the approaches.

The mixture density was modeled as ideal gas law. It was assumed that the recombiner vessel does not exchange heat with the outside gas mixture. Recombiner boundaries have been modeled as adiabatic no-slip wall. Heat transfer by radiation from plate to surrounding has not been modeled in the present work. Low Reynolds number k- $\epsilon$  model was used for modeling turbulence. All other properties like thermal conductivity, specific heat etc. were computed based on appropriate mixing law. Power law scheme was utilized as differencing scheme for the convective terms for all equation, whereas the central difference scheme was used for the diffusive terms. A constant time step of 0.05 s was used for entire computation. Since source term was present in the equations a low value of under-relaxation was used. Under-relaxation was set as 0.05 for momentum, 0.1 for pressure and 0.005 for energy and species equation. The convergence criterion was set  $10^{-8}$  based on overall mass residual. Simulation required approximately 100 hrs run time on a P-IV machine with 4GB RAM.

## RESULTS AND DISCUSSIONS

The model was used to predict the performance behaviour of the PAR for PARIS-1 benchmark exercise. The results were predicted in terms of total hydrogen mass, average pressure and temperature of representative containment volume and temperature at PAR inlet/outlet and plate temperature. Initially the hydrogen concentration was high, the conversion rate is directly proportional to hydrogen concentration thus hydrogen removal takes place sharply. Figure 2 shows the amount of total hydrogen in the representative containment as a function of time with both the approaches. As the hydrogen is consumed heat is generated and a flow field is developed and hydrogen moves from bottom of the recombiner towards top outlet. The outlet is  $90^{\circ}$  turned and leaves the mixture in horizontal direction from where the heated mixture moves upward at an angle and touches the top wall of the representative containment. A symmetric flow condition prevails in the domain during initial time period. Weak convective loops are formed from both the recombiners. These loops do not penetrate in the mixture below the recombiner height. As the reaction takes place, the pressure inside the closed representative containment increases. Figure 3 shows the pressure variation during the simulation of the PARIS benchmark with both the approaches. In the initial period there is a relatively steep increase of pressure, which corresponds to a relatively high hydrogen recombination rate. In the later stages of the simulation the pressure increases slowly as the hydrogen recombination rate is less during 1000 to 3000 s.

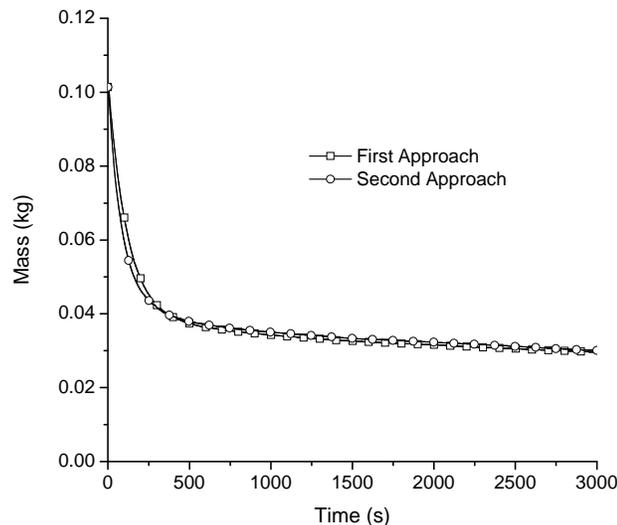


Fig. 2: Average hydrogen mass in the representative containment

Figures 4, 5 and 6 show the variations of the average temperature in the containment vessel, average inlet and outlet temperature of the PAR during the simulation with both the approaches. The second approach based on porosity predicted a higher temperature. Figure 7 shows average temperature of the recombiner plates with first approach. Since recombiner plates were not been modelled in second approach, it is not possible to evaluate the plate temperature. Figures 8 and 9 show the typical hydrogen mass fraction contour inside the representative

containment at 20 s and 300 s respectively. In the initial time period the flow was nearly symmetric and resulted in a stable thermal stratification in the domain after about 300 s. The thermally driven convection loops don't have sufficient strength to mobilise the cold hydrogen-rich gas located near the floor below the PAR inlet, this scenario develops into a diffusion controlled situation. The recombiner reduces the hydrogen concentration in the upper region significantly in nearly 900 s. After that only diffusional control recombination takes place and concentration reduces very slowly. The stratification pattern shows the influence of elevation of the PARs on the thickness of the bottom layer. The main reason for the formation of the bottom layer was the assumption of adiabatic walls from all sides. However in actual case containment wall will be at finite temperature and due to mixing promoted by steam condensation stratification will rarely occur. Figure 10 shows vertical temperature profiles at different times along the containment vertical centre line. After the initial homogeneous temperature field, a steep temperature front occurs, which divides the containment into two distinctly different regions with different temperature: the upper region with higher temperatures and the lower region with lower temperatures. The temperature in the upper region increases with time while in the lower region the temperature rise is less. Figure 11 shows vertical profiles of dry hydrogen mole fraction at different times along the containment vertical centre line.

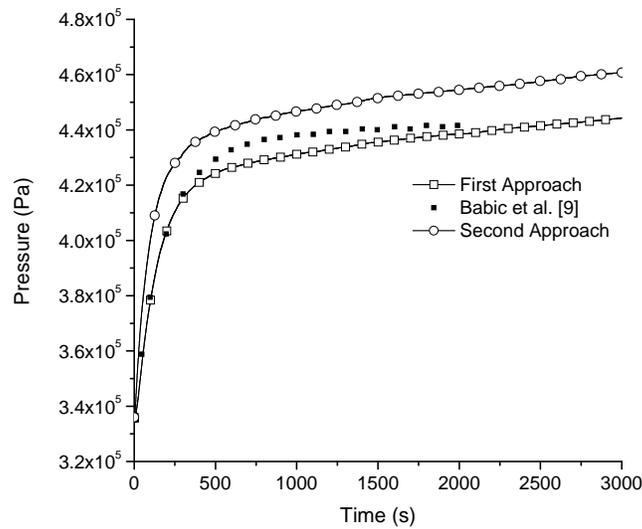


Fig. 3: Average pressure in the representative containment

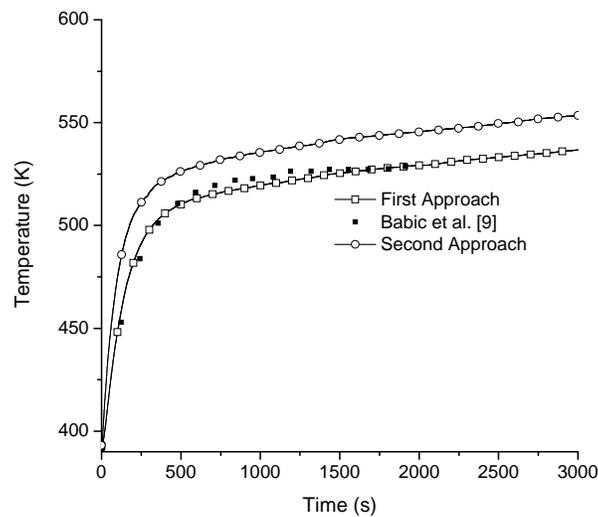


Fig. 4: Average temperature in the representative containment

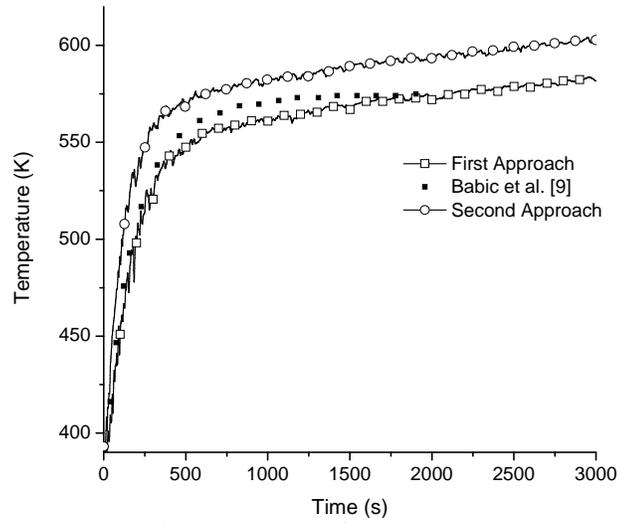


Fig. 5: Average left PAR inlet temperature

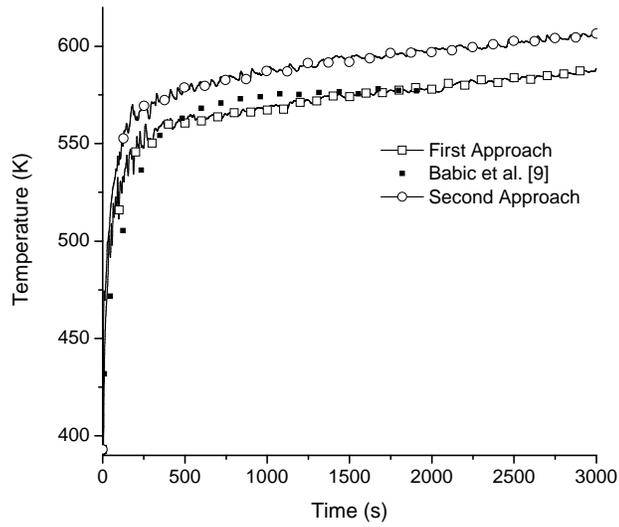


Fig. 6: Average left PAR outlet temperature

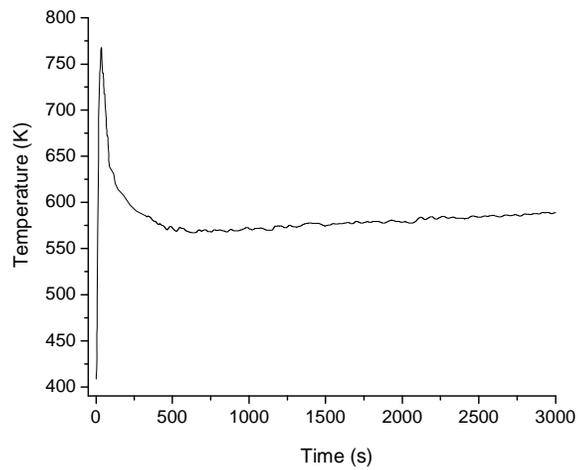


Fig. 7: Recombiner plate temperature

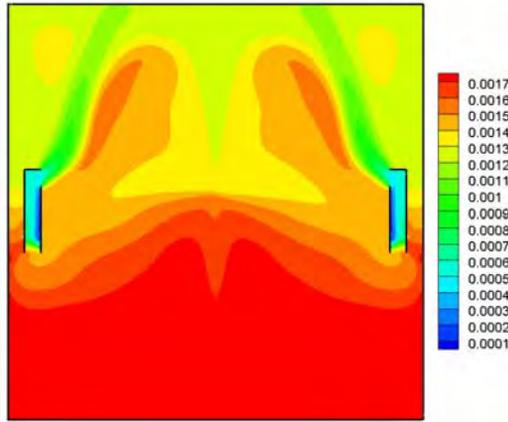


Fig. 8: Hydrogen mass fraction at 20 s

The Fig. 11 shows that hydrogen stratification occurs in the containment atmosphere. The dry hydrogen mole fraction evolves in a similar way as the temperature: first a uniform distribution, then a steep front and finally a gradual transition zone. The upper region of the containment atmosphere intensively interacts with the PAR, resulting in hydrogen consumption, whereas below elevation 1.0 m, the hydrogen concentration remains more or less constant.

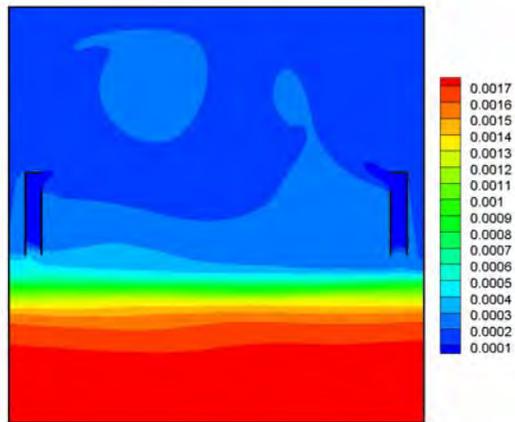


Fig. 9: Hydrogen mass fraction at 300 s

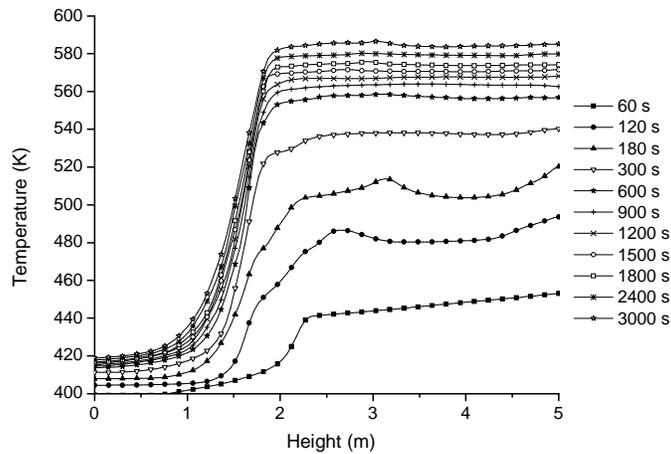


Fig. 10: Vertical temperature profiles at centerline at different times

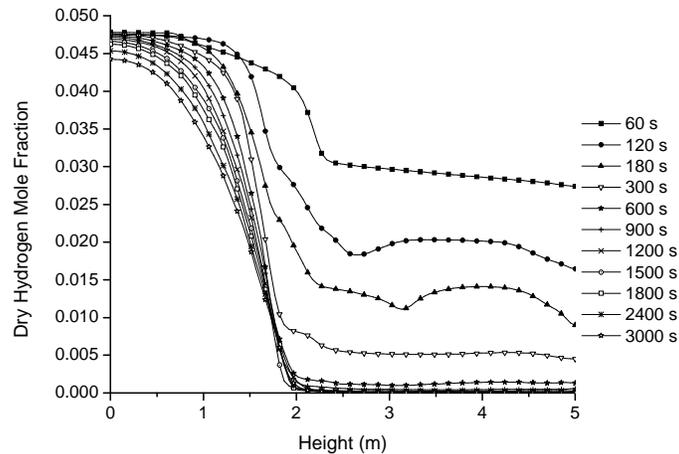


Fig.11: Dry hydrogen mole fraction vertical profiles at centreline at different times

## CONCLUSION

A simplified simulation of interaction of a passive autocatalytic recombiner with the containment atmosphere in a nuclear power plant was performed with the CFD code CFD-ACE+. A two-dimensional model was developed, and simple physical and geometrical conditions were assumed. A low-Re turbulence model was used. The beginning of the transient is characterised by a high hydrogen recombination rate and a steep increase of pressure and average atmosphere temperature. As hydrogen is consumed, the hydrogen recombination rate decreases and the pressure and average temperature stabilise. However, the simulation reveals that the interaction with PARs causes a partitioning of the atmosphere in two different regions: a lower region, with lower temperature and higher hydrogen concentration, and an upper region, with higher temperature and lower hydrogen concentration. At this instant, the only assessment of the validity of the obtained results is the comparison with published numerical results of the participants in the first phase of the PAR benchmark. The present approaches can be used for optimizing PAR location inside the reactor containment and assessing PAR efficiency in different scenarios. Modelling of steam condensation at containment walls can be considered for more realistic analysis. However for using such approaches the correlation for hydrogen conversion rate should be accurately known for the particular conditions.

## REFERENCES

- [1] IAEA, "Mitigation of hydrogen hazards in water cooled power reactors", IAEA-TECDOC-1196, 2001.
- [2] Bachellerie, E., Arnould, F., Auglaire, M., Boeck, B. de, Braillard, O., Eckardt, B., Ferroni, F., Moffett, R., "Generic approach for designing and implementing a passive autocatalytic recombiner PAR-system in nuclear power plant containments", *Nuclear Engineering and Design*, Vol. 221, 2003, pp. 151–165.
- [3] Kudriakov, S., Dabbene, F., Studer, E., Beccantini, A., Magnaud, J.P., Paillere, H., Bentaib, A., Bleyer, A., Malet, J., Porcheron, E., Caroli, C., "The TONUS CFD code for hydrogen risk analysis: Physical models, numerical schemes and validation matrix", *Nuclear Engineering and Design*, Vol. 238, 2008, pp. 551–565.
- [4] Travis, J.R., Spore, J.W., Royl, P., "GASFLOW: a Computational Fluid Dynamics code for gases, aerosols, and combustion," User's Manual, LA-13357-MS, FZKA-5994, 1998.
- [5] Implementation of simple passive autocatalytic recombiner models in ASTEC V1.0, Technical Note TN-ARN-1/2002, GRS Berlin, 2002.
- [6] Allelein, H.J., Arndt, S., Klein-Hebling, W., Schwarz, S., Spengler, C., Weber, G., "COCOSYS: status of development and validation of the German containment code system", *Nuclear Engineering and Design*, Vol. 238, 2008, pp. 872-889.
- [7] George, T.L., Wiles, L.E., Claybrook, S.W., Wheeler, C.L., McElroy, J.D., "GOTHIC-containment analysis package: Technical Manual", Numerical Applications Inc., 2001.
- [8] SARNET - Severe Accident Research NETwork of Excellence, <http://www.sar-net.eu/>
- [9] Babić, M., Kljenak, I., Mavko, B., "Numerical study of interaction between NPP containment atmosphere and passive autocatalytic recombiners", *International Conference Nuclear Energy for New Europe 2006*, Slovenia, September, 2006.
- [10] CFD-ACE+ V2009.2, User Manual, ESI CFD Inc., Huntsville, AL 35806