



CFD modeling of passive autocatalytic recombiners*

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Abstract. This study deals with numerical modeling of passive autocatalytic hydrogen recombiners (PARs). Such devices are installed within containments of many nuclear reactors in order to remove hydrogen and convert it to steam. The main purpose of this work is to develop a numerical model of passive autocatalytic recombiner (PAR) using the commercial computational fluid dynamics (CFD) software ANSYS-FLUENT and tuning the model using experimental results. The REKO 3 experiment was used for this purpose. Experiment was made in the Institute for Safety Research and Reactor Technology in Julich (Germany). It has been performed for different hydrogen concentrations, different flow rates, the presence of steam, and different initial temperatures of the inlet mixture. The model of this experimental recombiner was elaborated within the framework of this work. The influence of mesh, gas thermal conductivity coefficient, mass diffusivity coefficients, and turbulence model was investigated. The best results with a good agreement with REKO 3 data were received for $k-\varepsilon$ model of turbulence, gas thermal conductivity dependent on the temperature and mass diffusivity coefficients taken from CHEMKIN program. The validated model of the PAR was next implemented into simple two-dimensional simulations of hydrogen behavior within a subcompartment of a containment building.

Key words: nuclear reactor • hydrogen • passive autocatalytic recombiners • CFD modeling

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Introduction

A rupture of the primary circuit and leakage of the coolant into the reactor building (containment) is one of the most serious accidents that may happen in a water-cooled nuclear reactor. It is so-called loss-of-coolant accident (LOCA). The worst scenario assumes the break of the main coolant pipeline and double-sided leakage of water. This causes the loss of nearly whole coolant inventory in a few dozens of seconds. A rapid increase in temperature and pressure caused by a mixture of air and steam filling up the containment would be the result in a case of a controlled LOCA (assuming proper functioning of all the emergency systems). The situation would be much more severe if the emergency core cooling system were to fail. In the later phase of an uncontrolled LOCA, the mixture of steam, air, and hydrogen fills

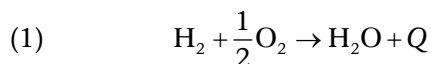
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up all the containment and, as a result, explosive conditions may appear. The following phenomena taking place during LOCA are the sources of the hydrogen [1–3]:

- Steam–zirconium reaction – it poses the most significant source of hydrogen in case of a large break LOCA. The rate of this reaction enlarges distinctly when the zirconium cladding temperature becomes greater than about 1100°C.
- Decomposition of water by radiolysis – caused by the complex interaction of ionizing radiation and water or dilute aqueous solutions.
- Corrosion of metals in a post-accident atmosphere – the knowledge about this process is still insufficient and the available data are, in the most cases, empirical.

It is obvious that situations leading to hydrogen generation should be avoided and this is the task of an emergency core cooling system. However, the risk associated with hydrogen should be taken into account and therefore, nuclear reactors must be equipped with systems for mitigating such risk. Many light water reactors (LWR) are equipped with passive autocatalytic recombiners (PAR) installed in reactor containments. The aim of their installation is to remove the hydrogen and convert it to the steam. A simple chimney shape construction (Fig. 1) and high efficiency of hydrogen removal is an advantage of this kind of devices.

The basis of a PAR operation is the exothermic reaction of hydrogen and oxygen:



that takes place on a catalysts surface. The amount of heat Q produced in this reaction is 120 kJ/g of hydrogen reacted [3]. Noble metals (platinum or palladium) coated structures, having the form of plates or spheres [4, 5], are used to form the catalyst bed. The recombination reaction starts automatically when a mixture of inlet gases containing hydrogen at given concentration come in contact with the catalysts' surface. The minimum concentration of hydrogen required to start the reaction usually is 1–2% by volume [4]. This is far below the flammability limit (4%) and detonation limit (10%) [3]. As the reaction is exothermic one, it heats up the gases and thus promotes natural circulation.

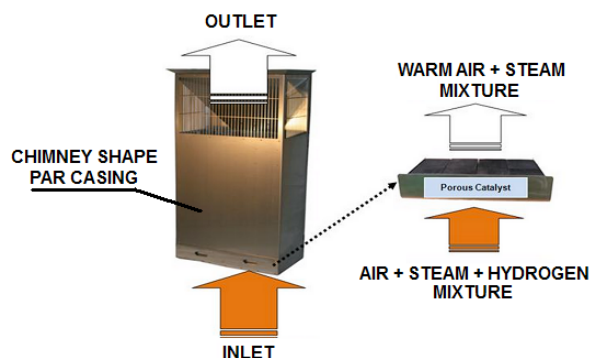


Fig. 1. Construction scheme and operating rule of a passive autocatalytic recombiner.

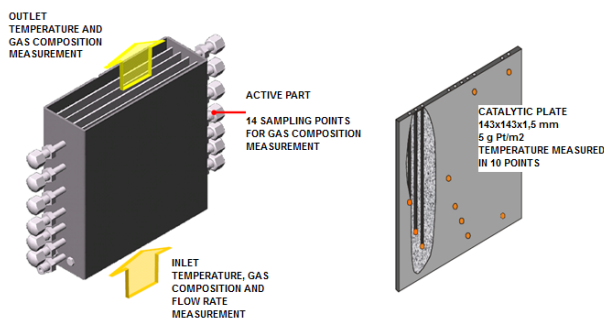


Fig. 2. Main part of the REKO 3 experimental station.

Development of the two-dimensional numerical model of a PAR

Characteristics of the analyzed object

There are many commercially available PAR designs [4]. However, this study concerns a specially designed PAR used in REKO 3 experiment. This is because a lot of data is available for this PAR and thus verification and validation of the numerical model under development is possible.

The main element of the test station is a plate-type PAR. Four steel plates are coated with platinum and placed parallel to the direction of gaseous mixture flow forming the vertical flow channels of rectangular cross-section area. The catalysts plates are enclosed in a rectangular casing of the shape close to the most popular commercial PARs. The diagram of the REKO PAR arrangement is shown in Fig. 2.

A mixture of steam and hydrogen has been supplied to the PAR during experiments. Parameters of the inflow mixture may be changed during the experiments. The basic measurements have been conducted for the inlet velocity equal to 0.25, 0.5, and 0.8 m/s, temperature 25, 70, and 110°C and for the inlet hydrogen concentration ranging from 0.5 to 4% [6].

Temperature of the plates and hydrogen concentration along the plates' height were measured and recorded. Thermocouples for temperature measurements were placed in holes drilled in the plates in order to avoid the mixture flow disturbances, as shown in Fig. 2. The gas samples were acquired from different measuring points, cooled down, and delivered to the analyzer chamber. The analysis of the mixture composition was performed by two independent systems operation in parallel [7].

General assumptions and governing equations of the model

Two-dimensional numerical model of the PAR has been elaborated in the presented stage of the work. Geometry of the active part of the PAR under consideration and types of boundary conditions are shown in Fig. 3.

Chemical reactions and physical processes take place inside the PAR. In the model, it is assumed that the kinetics of the recombination reaction (Eq. (1)) is described by the one-step Schefer's mechanism [8]:

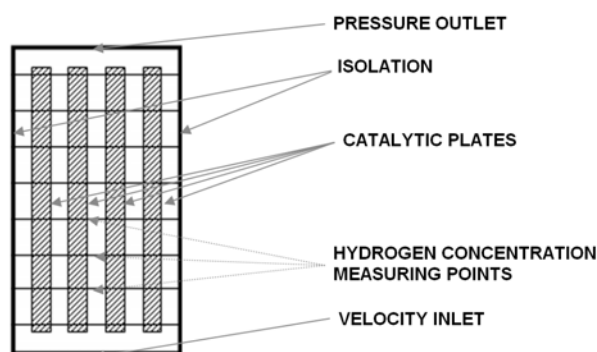


Fig. 3. Geometry of the active part of the PAR and types of boundary conditions.

$$(2) \quad k = AC_{H_2} \exp\left(\frac{-E}{(MR)T}\right)$$

where: A – pre-exponential factor, m/s; E – activation energy, J/kmol; MR – universal gas constant, J/kmol·K; T – temperature, K; C_{H_2} – molar concentration of H_2 , kmol/m³.

The elaborated two-dimensional CFD model of heat and mass transfer in the area of PAR utilizes species transport (four gases) model implemented in the FLUENT code. The mixture of air (nitrogen and oxygen), steam and hydrogen is treated as Newtonian fluid, ideal gas and its flow is assumed to be one-phase. Convective and diffusive heat transfer is considered in the model.

Taking into account the above-mentioned assumptions, a set of governing equations of the model consists of the following relationships:

- continuity equation for the gaseous mixture,
- momentum equation,
- species transport equations,
- energy balance equation,
- turbulence model equations,
- thermal equation of state,
- equation of the chemical reaction kinetics.

The geometry of the whole PAR and its active part is presented in Fig. 4. The initial analysis concerned the numerical mesh tests. Four types of structural meshes were applied to the model (see Fig. 4) and temperature profile along the catalytic plates was checked after testing computations. The experimental and numerical temperature profiles for the second catalytic plate are compared in Fig. 5. One can observe that temperature values predicted at the

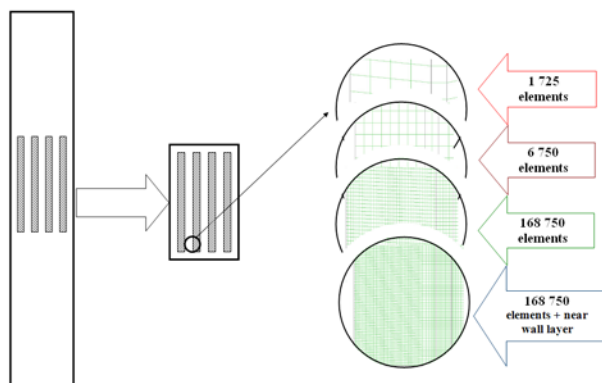


Fig. 4. Numerical meshes applied to the model.

beginning and the end of the plate while applying the coarse mesh are much higher than experimental ones. The medium-density numerical mesh allowed for better prediction of the ending part of the temperature profile. The temperature profiles obtained using dense meshes (without and with the near wall layer) are quite satisfying. Too big elements of the numerical mesh in the area of catalytic plates are not sufficient for proper reproduction of heat transfer to the fluid. The dense mesh with the near wall layer has been chosen for further analyses.

Setting up the main parameters of the model

This part of the study presents some kind of sensitivity of the developed model due to characteristic parameters describing physical and chemical processes taking place within considered recombiner. The simulations were carried out setting constant values of all the parameters, except the one tested at the moment. The results concern initial gaseous mixture temperature of 25°C and velocity at the inlet to the recombiner equal to 0.8 m/s.

Choosing a proper turbulence model while analyzing fluid flow with chemical reactions is very important. In the analyzed case, the gaseous mixture inflows to the bottom part of the PAR (relatively large flow path area) and next, flows through narrow spaces between the catalytic plates. This suggests that initially turbulent flow may become laminar in the active part of the catalyst bed. On the other hand, the intensity of chemical reactions taking place on the catalysts' surface introduces some irregularity to the flow. The next step in the numerical model development has been devoted to turbulence models tests. The results are presented in Fig. 6. The REKO 3 experiment results have shown that the temperature difference at both ends of the plate should be about 150 K. Such a value may be observed for the standard and the realizable k - ϵ turbulence models. Other turbulence models gave results differing significantly from the experimental ones. Assuming the laminar flow of the gaseous mixture also leads to wrong predictions. Taking the above into consideration, the standard k - ϵ model has been chosen for further analyses.

Important problem related to turbulence modeling is setting the turbulent Schmidt number (Sc_t), which is used for calculation of the turbulent diffusion coefficient. According to Laudner [9], this parameter should be equal to 0.75, while Kays and Crawford [10] set the value to 0.9 and received results close to experimental. The analyses carried out by the authors of [11] have shown that the Schmidt number may vary from 0.2 to 1.3. The actual value is dependent on the case under consideration and it may significantly influence the species transport results. Taking this into account, simulations have been realized in order to evaluate the Schmidt number influence. The values of this parameter ranged from 0.8 to 1.8 and the results are presented in Fig. 7. One may observe that setting higher values of the Schmidt number causes overestimation of temperature of the plate at its beginning, and setting lower values

results in overestimation of temperature at the end of the plate. Finally, it was decided that the Schmidt number equal to 1.3 allows for the best prediction of the temperature profile in the considered case.

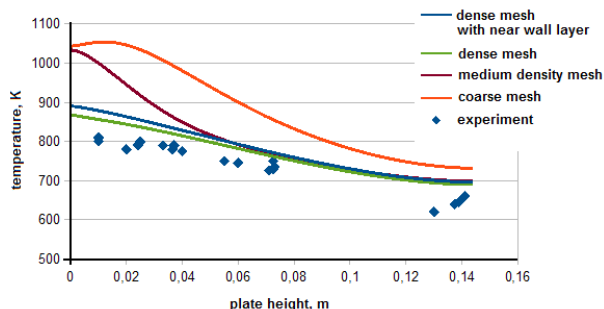


Fig. 5. Comparison of temperature profile along the centre catalytic plate for different size of numerical mesh.

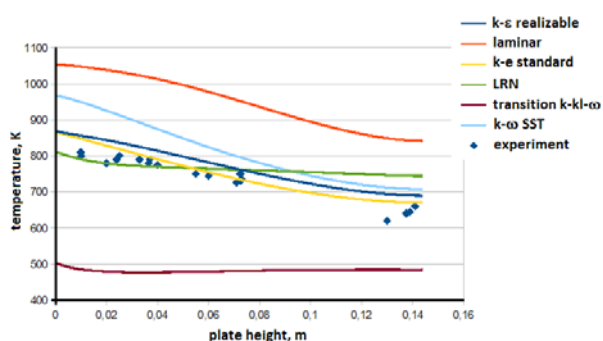


Fig. 6. Comparison of temperature profile along the centre catalytic plate for different turbulence models.

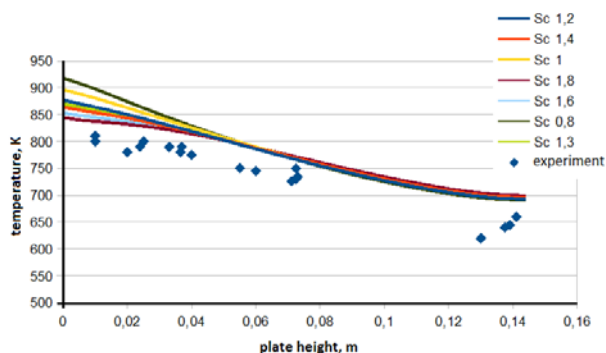


Fig. 7. Comparison of temperature profile along the centre catalytic plate for different turbulent Schmidt number values.

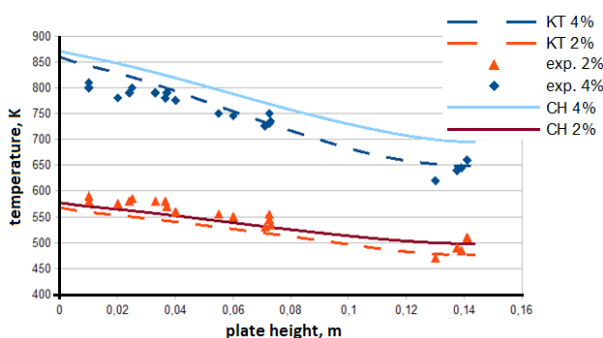


Fig. 8. Temperature profiles along the centre catalytic plate calculated using the kinetic theory of gases and CHEMKIN code libraries as the sources of molecular diffusion coefficients.

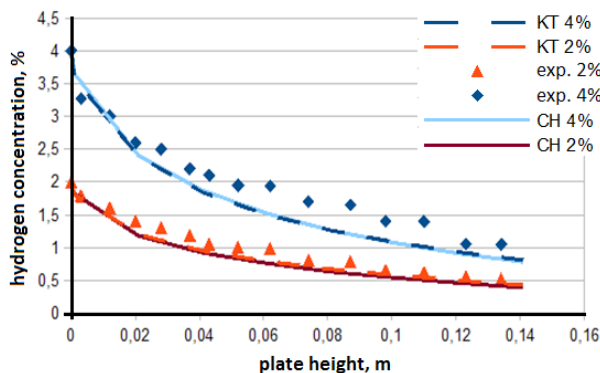


Fig. 9. Variations of hydrogen concentration along the centre catalytic plate calculated using the kinetic theory of gases and CHEMKIN code libraries as the sources of molecular diffusion coefficients.

The chemical reaction of hydrogen oxidation strongly influences the PAR operation. Therefore, special attention has been paid to parameters describing this process. The mass diffusivity coefficient is one of them and it has been analyzed in the study. Values of this coefficient have been determined according to the CHEMKIN code [12] libraries or based on the kinetic theory of gases [13]. Results of simulations obtained for initial hydrogen concentrations equal to 2 and 4% are compared with experimental data in Figs. 8 and 9. Figure 8 presents temperature profile along the plate. It can be observed that application of the kinetic theory of gases (line marked as KT) gave results closer to the experimental ones than in the case of using CHEMKIN database (line marked as CH). The results for this second case also may be recognized as satisfactory. Predictions of hydrogen concentration along the plate for both considered mass diffusivity coefficient sources are almost the same and remain in quite good agreement with experimental data.

Thermal conductivity of the gaseous mixture is an important factor affecting heat transport. This parameter is usually assumed as constant; however, in the considered case, the temperature gradient in the catalytic part is quite high (about 150 K along the plates). Therefore, a relationship for thermal conductivity was taken into account in the following form:

$$(3) \quad \lambda = \lambda_0 \sqrt{\frac{T}{T_0}}$$

where $\lambda_0 = 0.022 \text{ W}/(\text{m}\cdot\text{K})$ and $T_0 = 279 \text{ K}$. Figures 10 and 11 present the temperature profiles and hydrogen concentrations along the plate. It seems that estimation of hydrogen concentration does not depend on thermal conductivity model. Better compliance of experimental and numerical outcomes is achieved for 2% initial hydrogen concentration. It can be seen that for higher initial concentration of hydrogen, computational results are compliant at the beginning of the plate and are a little bit underestimated in the remaining part. Analyzing the temperature profiles shown in Fig. 10, one may note that assuming thermal conductivity dependent on temperature results in quite good agreement for

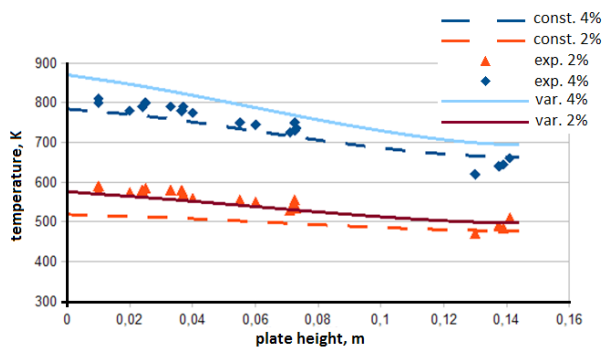


Fig. 10. Influence of thermal conductivity determination method on temperature profile along the centre catalytic plate.

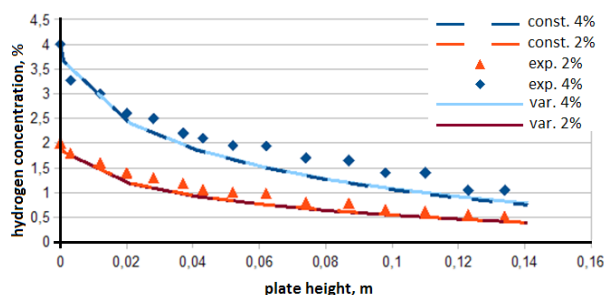


Fig. 11. Influence of thermal conductivity determination method on hydrogen concentration profile along the centre catalytic plate.

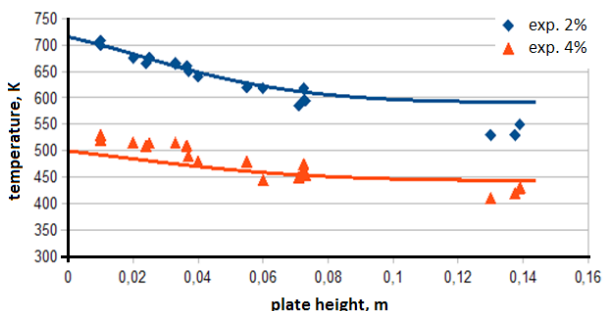


Fig. 12. The optimal numerical model: temperature profile prediction along the centre catalytic plate (inlet velocity of the gaseous mixture 0.25 m/s; inlet temperature 25°C).

lower initial concentration of hydrogen, while they are overestimated for higher initial concentration.

Validation of the model

All the analyses carried out before an optimal numerical model was created was taken into account. This model is based on following assumptions:

- the dense mesh with the near wall layer is used for discretization of the geometry,
- standard $k-\epsilon$ turbulence model is used,
- turbulent Schmidt number $Sc_t = 1.3$,
- mass diffusivity coefficients are taken from CHEMKIN code libraries,
- thermal conductivity is dependent on temperature according to Eq. (3),
- Schefer’s kinetics model with pre-exponential factor equal to 1.4×10^5 cm/s describes the recombination reaction.

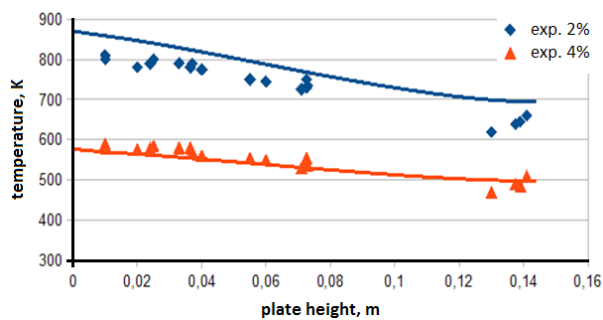


Fig. 13. The optimal numerical model: temperature profile prediction along the centre catalytic plate (inlet velocity of the gaseous mixture 0.8 m/s; inlet temperature 25°C).

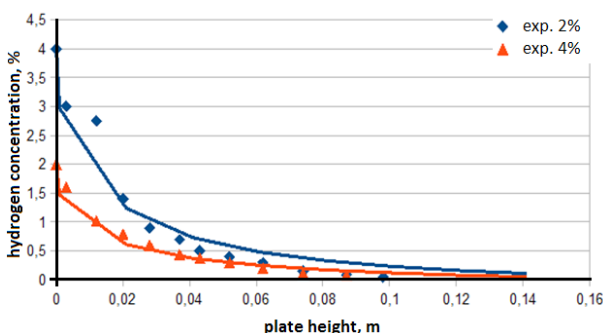


Fig. 14. The optimal numerical model: hydrogen concentration profile prediction along the centre catalytic plate (inlet velocity of the gaseous mixture 0.25 m/s; inlet temperature 25°C).

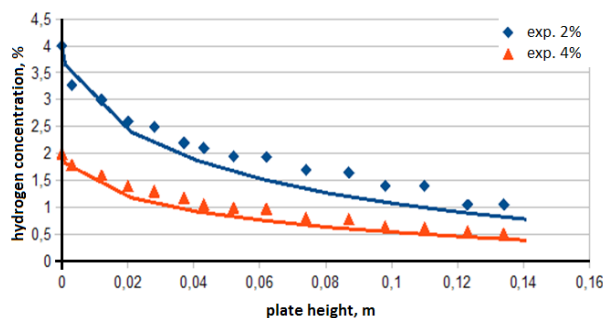


Fig. 15. The optimal numerical model: hydrogen concentration profile prediction along the centre catalytic plate (inlet velocity of the gaseous mixture 0.8 m/s; inlet temperature 25°C).

REKO 3 experiment was simulated applying all the assumptions listed above. Achieving convergence of results required running unsteady state simulations for 10 s, and next, the computations were switched to a steady state mode lasting about 10 min.

The results obtained by using the optimal numerical model are presented in Figs. 12–17. Comparisons of temperature profile and hydrogen concentration shown in Figs. 12–15 allow to state that the elaborated numerical model of the PAR gives results compliant with the experimental ones and reproduces physical and chemical processes with satisfying accuracy. The hydrogen concentration and temperature contours presented in Figs. 16 and 17 were obtained for inlet hydrogen concentration equal to 1, 2, 3, and 4%.

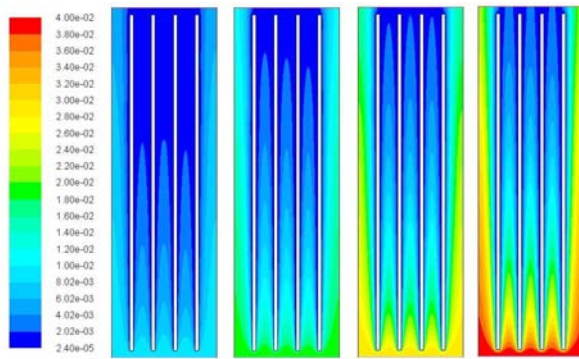


Fig. 16. Contours of hydrogen concentration in the active area of the modeled PAR for inlet hydrogen concentrations 1, 2, 3, and 4% (inlet velocity of the gaseous mixture 0.8 m/s; inlet temperature 25°C).

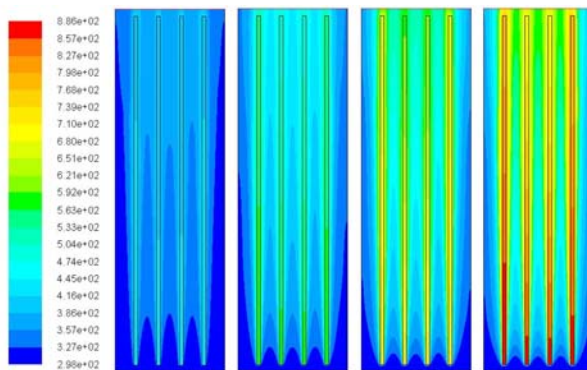


Fig. 17. Contours of temperature in the active area of the modeled PAR for inlet hydrogen concentrations 1, 2, 3, and 4% (inlet velocity of the gaseous mixture 0.8 m/s; inlet temperature 25°C).

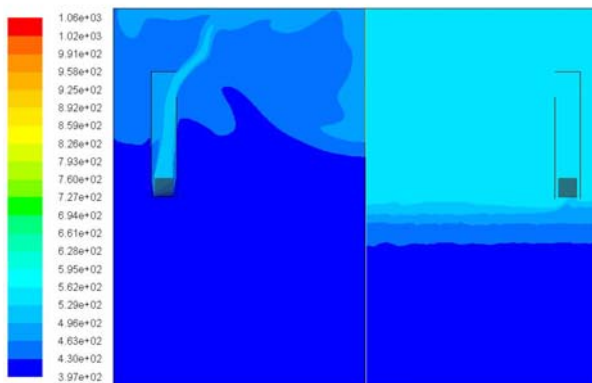


Fig. 18. PARIS-1 benchmark simulation: contours of temperature in the computational domain after 20 s (left side) and 100 s (right side) of process.

Sample application

Two-dimensional model of a containment subcompartment was elaborated with two passive recombiners placed symmetrically. Considered object is some kind of benchmark exercise called PAR interaction studies – PARIS [14] and it was previously investigated by authors of [15] and also REKO 3 research team [7]. The subcompartment height was 3 m and width 4 m. PARs physics and chemistry were modeled as described earlier, but the geometry was prepared for AREVA's FR 1-150 PAR type: casing

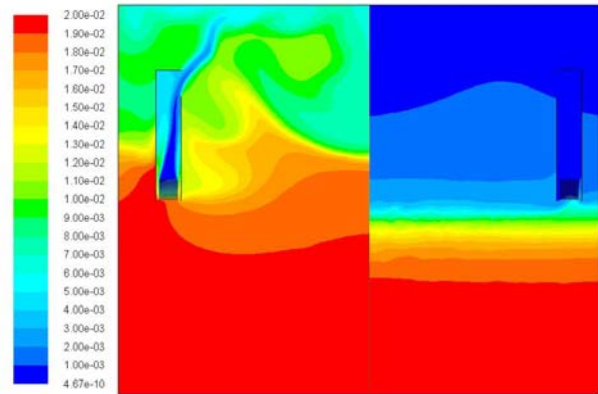


Fig. 19. PARIS-1 benchmark simulation: contours of hydrogen molar fraction in the computational domain after 20 s (left side) and 100 s (right side) of process.

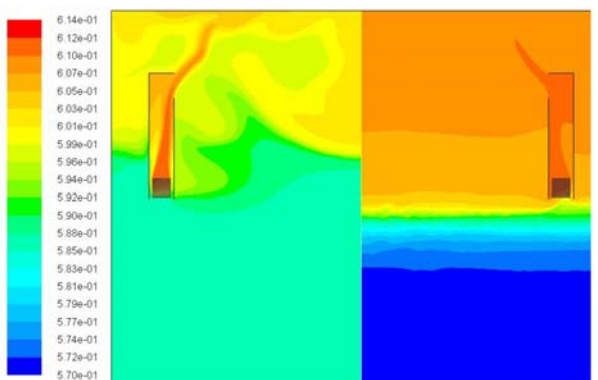


Fig. 20. PARIS-1 benchmark simulation: contours of steam molar fraction in the computational domain after 20 s (left side) and 100 s (right side) of process.

height 1.0 m and width 0.2 m. The catalytic bed contains 15 plates with dimensions $0.15 \times 0.15 \times 0.0001$ m [16].

Initial conditions of the considered case were as follow: mass fraction of hydrogen 0.018; mass fraction of oxygen 0.12; mass fraction of steam 0.48; the rest is nitrogen; temperature 393 K, absolute pressure 336 kPa. The case was analyzed as an unsteady state. Temperature contours as well as the hydrogen and steam concentration contours after 20 s (left side) and 100 s (right side) of analyzed process are shown in Figs. 18–20.

Final remarks and conclusions

The paper presents the main part of computational analyses carried out in order to elaborate a CFD model of a passive autocatalytic recombiner. It has been proven that numerical modeling of such a device is possible by using a commercial CFD software; however, it needs a lot of work to tune up the most important parameters of the model. The most important conclusions concerning performed analyses are as follow:

- the one-step Schefer's model describes well the catalytic oxidation of hydrogen in the recombiner, but an important issue is choosing the correct values of parameters describing the reaction ki-

netics with the catalyst present (mass diffusivity coefficients, pre-exponential factor),

- more accurate results have been obtained for lower hydrogen concentrations at the inlet.

The important issue is the creation of numerical mesh that should be fine enough to catch wall effects. This poses a serious problem in application of detailed CFD models of PARs to containment simulations – the scale problem would appear in such case. Nevertheless, some simplified simulations, regarding a part of the containment are possible.

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