

A model of hydrogen passive autocatalytic recombiner and its validation via CFD simulations

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Abstract Passive autocatalytic recombiners (PAR) is the only used method for hydrogen removal from the containment buildings in modern nuclear reactors. Numerical models of such devices, based on the CFD approach, are the subject of this paper. The models may be coupled with two types of computer codes: the lumped parameter codes, and the computational fluid dynamics codes. This work deals with 2D numerical model of PAR and its validation. Gaseous hydrogen may be generated in water nuclear reactor systems in a course of a severe accident with core overheating. Therefore, a risk of its uncontrolled combustion appears which may be destructive to the containment structure.

Keywords: Nuclear reactor; Containment; Hydrogen; Numerical modeling; CFD

1 Introduction

In a severe accident or a beyond-design-basis accident (BDBA), the reaction of steam with zirconium alloy fuel cladding, radiolysis of water, molten corium-concrete interaction and post-accident corrosion can generate gaseous hydrogen. The total mass of hydrogen produced in-vessel depends on several factors and for most reactors is of the order of 1000 kg [1].

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High peak rates for hydrogen release to the containment can result from discontinuous releases from the reactor pressure vessel. The detonation of this gas can result in damage to such structures as containment buildings or spent fuel buildings. To avoid the risk of containment damage due to hydrogen detonations, passive autocatalytic recombiners (PARs) can be utilized in various reactor designs.

Passive autocatalytic recombiners are widely used for hydrogen removal systems design in modern nuclear reactors, but existing nuclear power plants with older reactors started to utilize such solution also. The knowledge on hydrogen quantities and distribution within containment volume is necessary for proper localization of these devices [2]. The rates and quantities of hydrogen produced and the location of its release into the containment depend on a variety of conditions largely independent of each other (accident scenarios, core degradation progress, unavailability of engineered safety systems, etc.) and also independent of the reactor type [2].

As it has been mentioned earlier a serious problem for the containment integrity may result from hydrogen combustion. The flammability limit in gaseous mixtures such as steam, air and hydrogen, is defined as the minimum concentration of hydrogen required to propagate a flame in the environment where oxygen is present in excess. The experimentally determined flammability limits in steam-saturated air at room temperature and pressure are [3]:

- 4.1% vol. for upward propagation,
- 6.0% vol. for horizontal propagation,
- 9.0% vol. for downward propagation.

Significant hydrogen concentration could be reached locally in a short time, leading to a flammable gas mixture, but because of the presence of high steam concentrations, hydrogen burn may be prevented. If the atmosphere is undergoing rapid condensation – e.g., by spray initiation – a potentially detonable mixture could form rapidly in case of a high concentration of hydrogen [4]. Therefore it is crucial to gain a detailed information concerning the hydrogen behavior within the containment building. This may also help in more realistic predictions of hydrogen removal system performance.

Specific numerical models have been developed in the last twenty years to simulate the behaviour of catalytic recombiners in accidental conditions. These models include: empirical model or correlation of module hydrogen

recombination rate, theoretical 1D model for simulating the internal behavior [5]. Such models may be coupled with two types of computer codes, namely the lumped parameter codes, and the computational fluid dynamics codes.

The use of a correlation in a lumped parameter code seems to be sufficient for a fast evaluation of the hydrogen amount versus time in the containment or in the different subcompartments. Such approach has been applied in [6]. On the other hand, for the assessment of the efficiency of individual catalytic recombiners (start-up behaviour, temperature distribution) local conditions at the entrance of the recombiners are needed [7], which are, at the beginning of hydrogen release, different from the averaged compartment values. For such an evaluation the computational fluid dynamics (CFD) models are necessary.

This work deals with a 2D numerical model of PAR and its validation via CFD computations. The simplified model of PAR has been elaborated for realizing coupled analyzes with the lumped parameter code and the CFD code. The lumped parameter code HEPICAL has been described and already used for simulations of hydrogen combustion and its thermodynamics consequences to the containment structure loads [8].

2 Characteristics of the catalytic hydrogen removal method

A catalytic recombiner is passive device – no external energy is needed for its operation, and is self starting also at low temperatures and wet conditions. The recombiner consists of a vertical channel or stack equipped with a catalyst cartridge in the lower part, see Fig. 1. Such design creates the so-called ‘chimney effect’ – a gas mixture flows through the recombiner by means of natural circulation.

The catalyst bed has the form of plates of spheres coated with an active material. An exothermic reaction occurs at the surface of the catalyst cartridge when hydrogen and oxygen are present in the atmosphere. The heat of the reaction, combined with the vertical arrangement and spacing of the catalyst plates or spheres promote natural convective flow through the recombiner. Warm humid air and unreacted hydrogen are exhausted through the top grating while steam-air mixture and hydrogen are drawn through the bottom. The active catalyst materials include the noble metals: platinum or palladium.

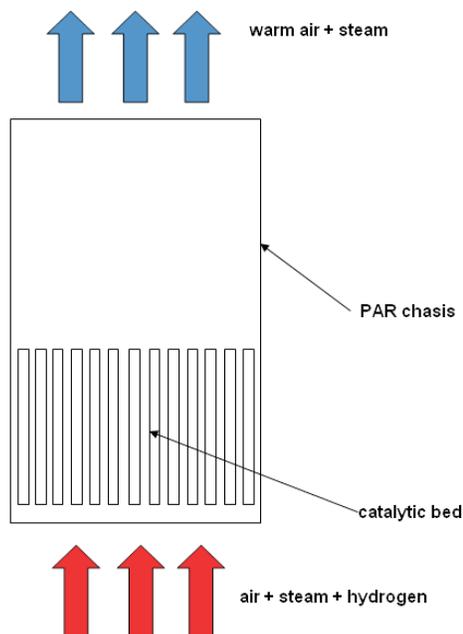


Figure 1. Passive autocatalytic recombiner – scheme of construction and operating rules.

A PAR based system for hydrogen removal consists of catalytic recombiners – from 25 to 60 [5] for a typical pressurized water reactor containment to accommodate a wide range of hydrogen release scenarios. The arrangement of the individual catalytic recombiners inside the reactor building is determined by a variety of parameters such as planned hydrogen release rates, location and distribution, geometry and operational constraints on maintenance areas, accessibility.

3 Computational analysis

3.1 Numerical model of PAR

First numerical model of the passive recombiner comprises the two-dimensional model of a device of perpendicular shape. The geometry of that model, basic dimensions and boundary conditions types are presented in Fig. 2. Dimensions in this figure are given in mm. It should be noted here that this model does not refer to any real PAR. The aim of this work is to establish

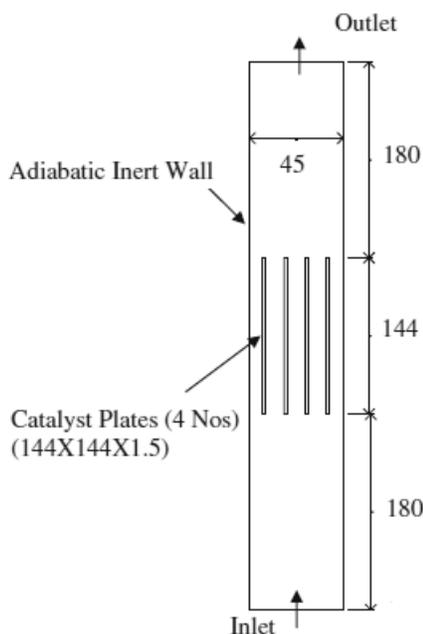


Figure 2. Geometry of the numerical model of PAR with basic dimensions and boundary conditions set, in mm.

a certain base for modeling of such devices by means of CFD approach and for further development of the models. However, the model under consideration has been created in order to make possible its validation according to the REKO test facility, what is described in the following section.

The numerical mesh of this model consists of 22.5 thousand of quad-type cells. The model is supplemented with governing equations of energy balance equation, continuity equation and momentum equation [9]. As the natural flow is assumed to occur in the passive recombiner the flow has been modeled as laminar in the first simulations.

An important issue in the recombiner model is the approach for modeling the reaction of hydrogen oxidation. The one-step Schefer's mechanism for this reaction with the catalyst present in the atmosphere has been applied:



The rate of this reaction was determined according to Arrhenius equation [10], which is a simple formula for the temperature dependence of reaction

rates:

$$R = 14 \times \exp(-14.9 \times 10^6 / R_u T) \times [\text{H}_2] \quad \text{mol/m}^2/\text{s}, \quad (2)$$

where R_u is the universal gas constant, T is the absolute temperature, and $[\text{H}_2]$ is the hydrogen mole fraction.

3.2 Results of initial simulations

First simulations have been accomplished applying the following boundary conditions:

- inlet gaseous mixture temperature $T = 343$ K;
- inlet velocities 0.25, 0.5 and 0.8 m/s;
- inlet hydrogen concentrations 1, 2, 3 and 4% (volumetric fractions).

The problem has been solved as the steady state one. Selected results of computations are presented in Figs. 3 and 4.

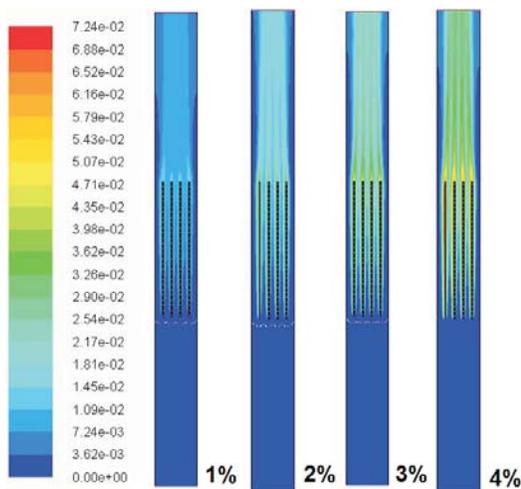


Figure 3. Distributions of hydrogen volumetric fraction in the computational domain for different inlet hydrogen concentrations.

3.3 Model validation

The experiments at the REKO-3 test facility have been used for validation of the numerical model of PAR in this work. This testing rig serves mainly

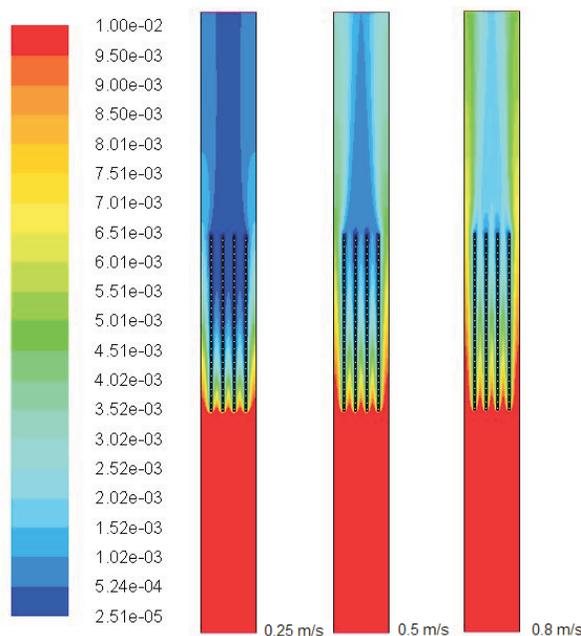


Figure 4. Distributions of hydrogen volumetric fraction in the computational domain for different inlet velocities.

to clarify the basic phenomena taking place within the PAR, that is interactions of reaction kinetics, heat and mass transfer, and the flow conditions inside the recombiner. The experimental set-up is shown in Fig. 5. It allows to investigate the catalyst samples inside a vertical flow channel under the well defined conditions such as composition of gas mixture, its flow rate and inlet temperature. The catalyst sheets (in the form of stainless steel coated with platinum catalyst material) are placed parallel forming the vertical rectangular flow channels [11].

The experiments have been performed for three different flow rates (0.25, 0.5 and 0.8 m/s), for three different inlet gas temperatures (298.15, 343.15, and 383.15 K), and for hydrogen concentrations between 0.5 and 4.0%. The main parameters measured during tests were the distribution of the catalyst temperatures and the gas compositions in the vertical flow direction. The gaseous mixture composition at the outlet was also measured. Selected tests performed at the REKO test station have been simulated using the numerical model described earlier. The results are shown in Figs. 6 and 7.

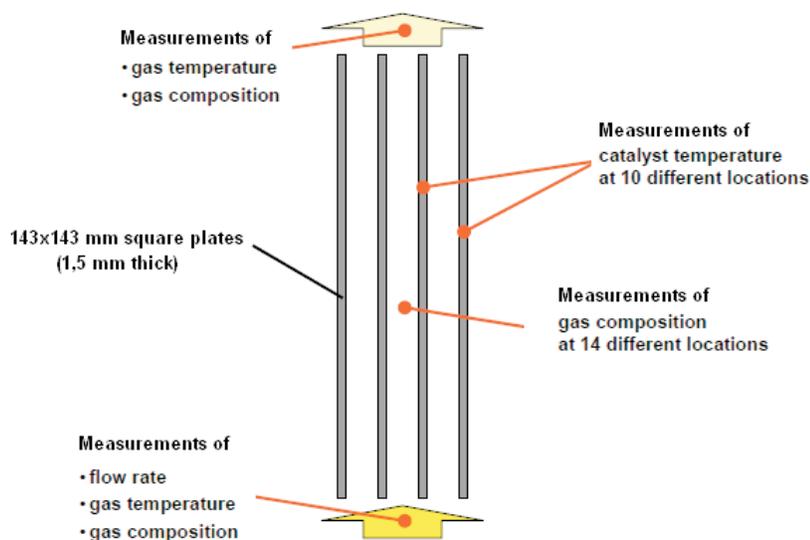


Figure 5. Simplified scheme of the REKO-3 test rig active zone with measurements information (prepared according to [11]).

It can be seen from Figs. 6 and 7 that computational results remain in

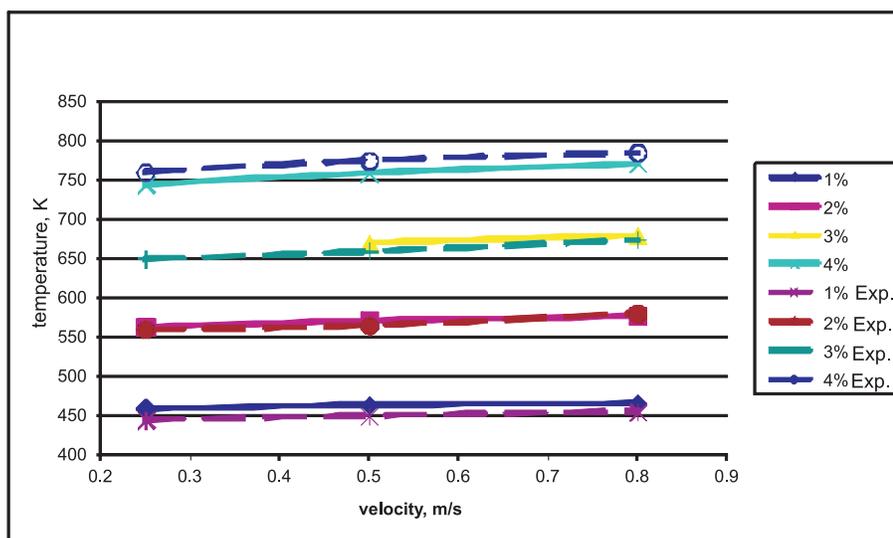


Figure 6. Maximum temperature of catalytic plates as a function of inlet velocity.

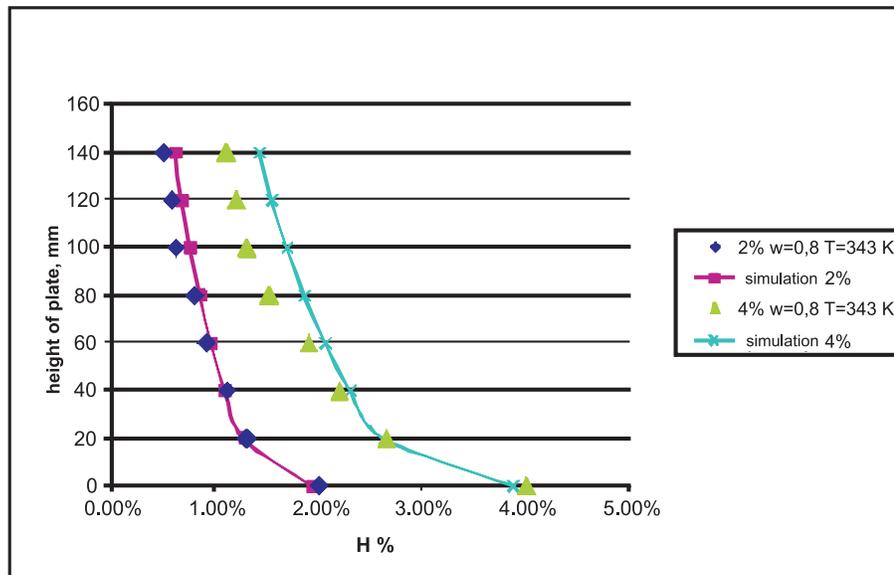


Figure 7. Hydrogen concentration along the height of plates for different inlet concentrations.

quite a good agreement with the experimental ones. Temperature in active zone depends on the oxidation reaction rate. This reaction rate is higher for higher hydrogen concentrations, and this is clearly observed in Fig. 6. Looking at Fig. 7 points out that the higher fraction of hydrogen at the inlet makes the recombination process more efficient.

4 Conclusions

The work presents initial results of application of CFD based methods for the passive autocatalytic recombiners modeling. 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,
- an important issue is choosing the correct values of parameters describing the reaction kinetics with the catalyst present,
- a more accurate results have been obtained for lower hydrogen concentrations at the inlet.

Summarizing it could be noted that the results obtained in this initial phase of modeling are promising, however, the presented model needs some further work in its development and validation.

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