DEVELOPMENT OF A CFD BASED ANALYTICAL MODEL FOR THE PASSIVE CATALYTIC RECOMBINERS FOR HYDROGEN MITIGATION IN INDIAN NUCLEAR POWER PLANTS

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ABSTRACT

Release of hydrogen as a consequence of severe accident in a nuclear power plant can be a potential threat to the integrity of the containment building. The use of passive catalytic recombiners is one of the best known options being considered for hydrogen management in nuclear power plants (NPPs). The modelling of recombiners using CFD tools is being attempted worldwide. The objective of the present study is to build and validate a 3-D analytical model for the passive recombiner using the governing mass, momentum, energy and species conservation equations from first principles. CFD code FLUIDYN-MP has been used to setup the governing equations for modelling of the associated multi-physics phenomena. The present paper brings out the salient features of the analytical model of the passive recombiner incorporated in the CFD code FLUIDYN-MP and its validation against test data from open literature.

INTRODUCTION

Many methodologies and technologies have been investigated and developed in the past to mitigate the consequences of hydrogen buildup inside the containment of NPPs under severe accident scenarios. One such most promising method is to deploy passive catalytic recombiners, which work on the principle of recombining hydrogen with oxygen from ambient air on catalytic surfaces to form steam and release of the exothermic heat of reaction. These devices are known to recombine hydrogen at much lower concentrations and at near-ambient temperatures, thus eliminating the risk of gas phase ignition/deflagration/detonation of the hydrogen–air-steam mixtures.

The development of an analytical model for the passive recombiner needs consideration of various multi-physics phenomena, such as buoyant gas transport, conjugate heat transfer between catalytic plates and gas mixture, combined convective and radiative heat transfer mechanisms, species transport by combined convective and molecular diffusion and the reaction kinetics. Experimental as well as analytical studies on hydrogen behaviour in nuclear reactor containments are being performed worldwide. As a part of analytical modelling of the recombiner behaviour, initially the codes that were developed were based on either empirical/semi-empirical/lumped parameter approaches or one-dimensional modeling approaches for representing various phenomena that take place in the recombiner box. However, this approach lacked the capability to generate several important data necessary to ensure safe and efficient performance of the passive catalytic recombiners. In the recent times, CFD based modelling for hydrogen behaviour in closed geometries has gained importance because of its capability to accurately model and predict many such missing information, viz., (i) transport of hydrogen within the recombiner and also into the surrounding (outside the recombiner) gas mixture, (ii) local flow patterns, and (iii) detailed catalyst plate and gas mixture temperatures.

Most recently, Heitsch [1] has presented a detailed study on CFD calculations using GASFLOW, FLUENT and CFX for the Paks NPP. The purpose of this work was to demonstrate that CFD codes can be used to model gas movement inside the containment during a severe accident. Kudriakov [2] applied the TONUS code for hydrogen risk analysis. Breitung [3] has presented a detailed step by step procedure for mechanistic analysis of hydrogen behavior and mitigation during accidents and highlighted the application of CFD codes for hydrogen behaviour modelling. Royl [4] has performed three dimensional analysis of steam/hydrogen transport with catalytic recombiners in a nuclear reactor containment using computer code GASFLOW.

As part of the technology development programme for hydrogen management in Indian Pressurised Heavy Water Reactors (IPHWRs), experimental as well as analytical studies are underway at BARC. A CFD based model has been developed and integrated in the Code FLUIDYN-MP. The model has been validated against experimental data available in open literature. The recombiner model has also been used to predict the behavior of the prototype recombiner designed in BARC.

This paper presents salient features of the model, its validation against data from open literature and results of parametric studies performed to investigate the influence of various process and design parameters.

ANALYTICAL MODEL DEVELOPMENT

Salient Features of FLUIDYN-MP

FLUIDYN-MP is a general purpose multi-physics CFD code which has a fully-implicit, pressure based, collocated solver for transient incompressible/compressible flows with heat transfer, turbulence, chemical reactions, free surfaces, etc. The code solves for Cartesian velocity components using a collocated finite volume method. The conservative forms of compressible Navier-Stokes equations, the equations for conservation of energy and transport for individual species are solved along with the ideal gas equation of state of the form $\rho = f(p,T)$. Pressure is computed using a Poisson solver for the pressure correction using three different methods. Convection fluxes are computed by blending of pure upwind scheme and higher order schemes. The time integration is performed using an Euler implicit scheme. Three methods of pressure-velocity coupling are available: SIMPLE, SIMPLEC, and PISO. Some of the above-mentioned features of the code have been used to model the catalytic recombiner along with a simplified model for the kinetics of the catalytic reaction and using SIMPLE and SIMPLEC algorithms of the code.

Modelling of Reaction Kinetics

A chemical reaction can be modelled in the code FLUIDYN-MP in the following general form

$$\sum_{i=1}^{i=j} \nu_{1i} R_{1i} \leftrightarrow \sum_{i=1}^{i=k} \nu_{2i} R_{2i}$$

with ν's representing the stoichiometric coefficients and R_1 and R_2 the reactants and products respectively. In the present model, the rate is expressed in terms of the concentrations of the reactants and/or products, temperature, etc. The source terms for the species concentrations and temperature are expressed as follows

$$S_i = \sum_j R_j (\nu_{2i} - \nu_{1i}) M_i$$

$$S_T = \frac{1}{Cp} \sum_j Q_{rj} R_j$$

S_i = source term for species i
R_j = reaction rate of reaction j in terms of leading reactant [kmol/m^{3}s]
M_i = molecular weight of species i [kg/kmol]
S_T = source term for temperature
Q_{rj} = heat of reaction of j in terms of leading reactant [J/kmol]
$= \frac{1}{\nu_{11}} \sum_i \Delta H f_i (\nu_{2i} - \nu_{1i})$ where ΔH_f = heat of formation

The NSNT solver of FLUIDYN-MP models finite-rate chemical reactions. The rate of a reaction, R_j, can be computed in 3 ways:

(i) Rate completely determined by chemical kinetics
(ii) Rate controlled by the mixing of the reactants
(iii) Combination of the above two

In the Arrhenius model used for the present work, the reaction rate, R_j, is completely determined by chemical kinetics. The generalised Arrhenius form as implemented in NSNT is as follows:

$$R_j = -A_j T^{\zeta_j} e^{-E_j/\Re T} \prod_i x_i^{\nu_{ji}}$$

where A_j is the pre-exponential factor, ζ_j is temperature exponent, E_j is the activation energy, R is the universal gas constant, x_i denotes the mole fraction of species i and ν_{ji} depicts the species rate exponents. Aj, ζj, Ej, νji are experimentally determined parameters that need to be given by the user. However, the solution of this equation requires the mesh to be fine enough to resolve reaction zones appropriately. The factors A_j and E_j have a major role in determining the mesh size and quality.

The above reaction has been modelled earlier by a detailed reaction mechanism as reported by Heitsch [5] and Appel [6]. For the present work, a single step global reaction rate expression as reported by Schefer [7] has been used. This approach assumes a single step reaction from the stable reactants to the stable products without considering any of the intermediate reactions. So the species considered in the reaction are hydrogen, oxygen and water vapour. The rate of the catalytic reaction in the present case is given by the following expression [7].

$$R'' = 14.\exp.\left(\frac{-16.1x10^{6}}{R_u.T}\right).[H_2]$$

This expression is reported to have been obtained by fitting the data from the experiments performed for hydrogen mole fraction in the range of 7% to 20% and temperature varying from 450 K to 1200 K. The region covering the plate is modelled as a solid domain and rest of the domain is modelled as fluid cells. The reaction source and sink terms for the species are applied in the very first fluid cell adjacent to the solid catalyst surface. Since the reaction is taking place at the surface, the heat release due to the reaction occurs directly on the plate surface. It is assumed that the reaction mechanism is same for all the catalytic plates and hence, the reaction rate will depend on the prevailing catalyst temperature and the species concentration in the adjacent fluid cells.

Modes of Heat Transfer

The heat flow between the grid points is calculated as displayed in Fig. 1. Inside the catalyst plates, heat is transported due to heat conduction. Inside the flow channels, the heat flow is due to conduction through the gas phase. Conjugate heat transfer model for representing convective heat dissipation from solid plates to gas phase as well as radiation heat transfer mechanism between the walls have been considered. Heat sources which result from the exothermic reaction between hydrogen and oxygen are added directly to the catalytic walls.

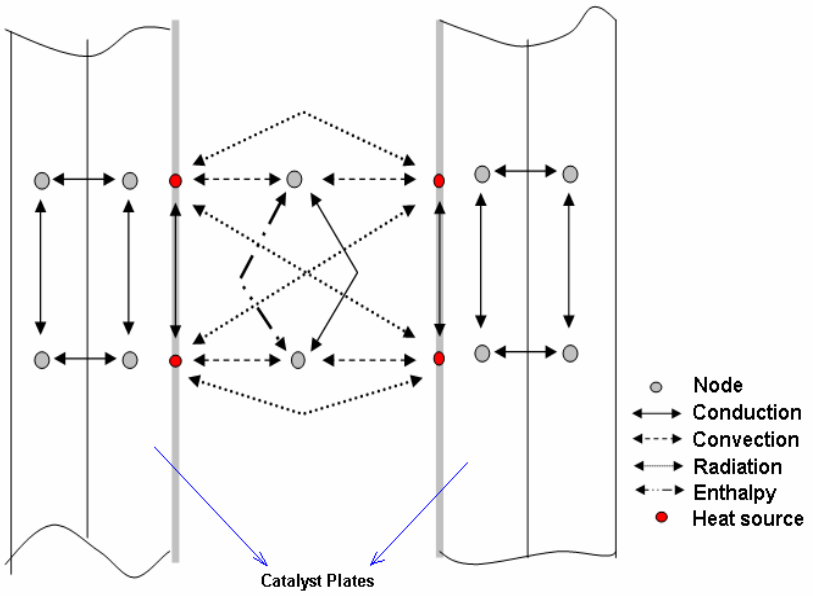


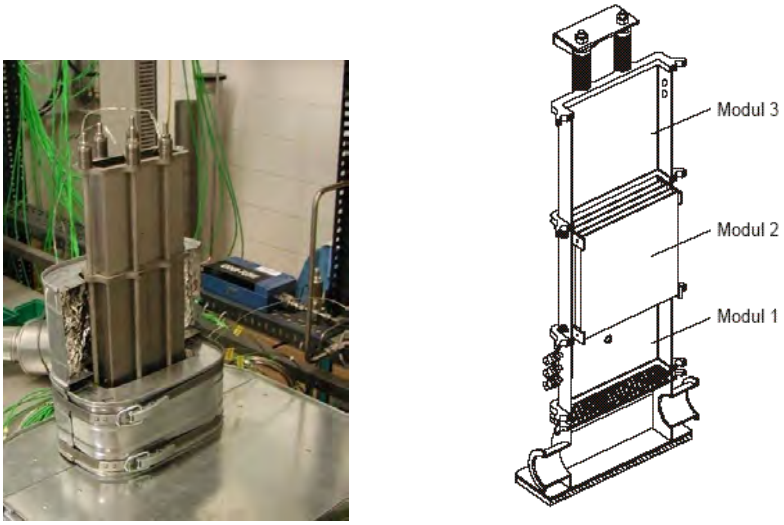
Fig. 1: Modes of Heat transfer in the recombiner model

Flow Configuration

Flow is assumed to be compressible, laminar and viscous. The fluid is a gas mixture comprising nitrogen (carrier fluid), hydrogen, oxygen and water vapor. The data for the gas specific heat and other transport and thermal properties are taken from reference [8] and [9].

VALIDATION

The model for catalytic recombiners as described above was validated by comparing the results of the analysis against the experimental results obtained in REKO-3 facility (Fig. 2). A brief description of the facility is given below. This facility has three modules (Module1, Module2, and Module3). Module1 works as inlet and feeds gas mixture to Module2, which contains four catalytic plates where hydrogen recombines with oxygen to form water. Module3 works as outlet. The whole unit is thermally insulated on the sides to prevent heat losses. Four catalyst sheets (stainless steel sheets coated with wash coat/platinum catalyst material) having dimensions of 143mm x 143mm x 1.5 mm are used in the experiments. The facility has been used to investigate the behavior of a recombiner under controlled conditions [10, 11, 12].



(a) Partially Insulated (b) Schematic

Fig. 2: REKO-3 Test facility

Summary of Validation Cases

The geometry for numerical computations is shown in Fig. 3. For the model development and validation, various simulations were performed with increasing complexity of simulation in which a dry gas mixture of air and hydrogen was fed to the recombiner. These simulations differed in number of channels, gas transport properties and heat losses to the ambient.

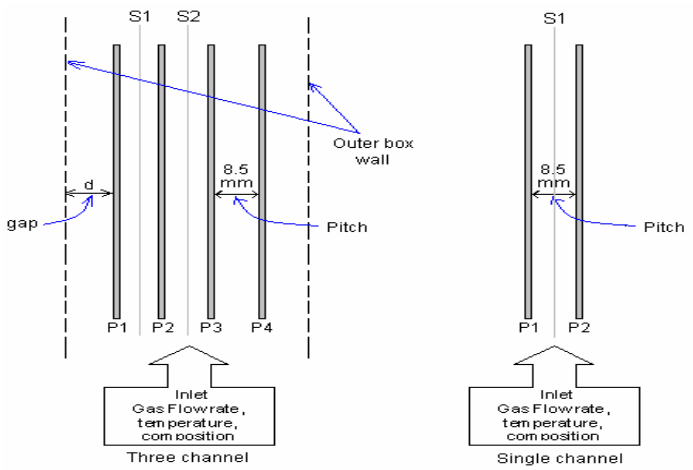


Fig. 3: Geometry for three channel and single channel cases

The various analyses performed are as listed below:

- Single channel (2 parallel plates)
  - With and Without heat losses from the box to the surroundings

    Simulations to investigate the effect of transport properties of the gas mixture at different gas mixture temperatures (room temperature, minimum, maximum and average plate temperature) on the recombiner behaviour.

    Simulation of convective and radiative heat losses in the recombiner along with grid sensitivity studies.

- Three channel (4 parallel plates)
  Simulation of the 4-plate recombiner experiments having finalised the boundary conditions on the basis of single-channel studies.
- Parametric studies on single channel configuration to assess the influence of various parameters like inlet concentration of hydrogen, inlet flow velocity, plate spacing and height, and diffusivity of hydrogen on the recombination behaviour.

RESULTS AND DISCUSSION

Single Channel Cases (with and without heat loss)

In the first set of calculations, heat losses from the plates to the ambient were neglected. These simulations were performed for inlet hydrogen concentration of 2% H_2 (v/v), inlet velocity of 0.25 m/s and using gas transport properties at room temperature. Since there was a deviation observed between the calculated results and experimental data, further calculations were performed by considering the gas transport properties (viscosity, Prandtl Number) at different temperatures (minimum, maximum and average plate temperatures). It was concluded that the transport properties at average catalyst plate temperature simulated the experimental conditions in the best manner. Subsequent simulations were performed with the properties at average plate temperatures. It was also noted from these studies that though the concentration profiles were closer to experimental results, the plate temperature profiles were showing deviation from experimental results, especially in the upper region of the plate. To find out the reason for this deviation, grid sensitivity studies were performed with the grid sizes as given in Table 1.

Table 1: Description of grid sizes used for calculations

Grid	Element size	
	Along the flow direction (mm)	Perpendicular to flow direction (mm)
G1	1	0.5
G2	0.5	0.25
G3	0.25	0.125

Typical results for the different mesh sizes are given in Fig. 4 and Fig. 5. The numerical results matched closely with the experimental results for the finer mesh (G2 and G3). For this simulation, the heat losses from plates to ambient were also considered, as described in the previous section.

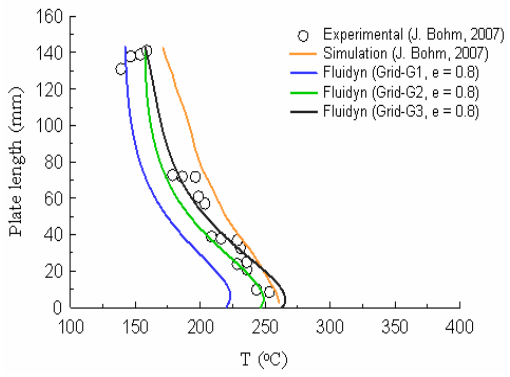


Fig. 4: Catalytic plate temperature

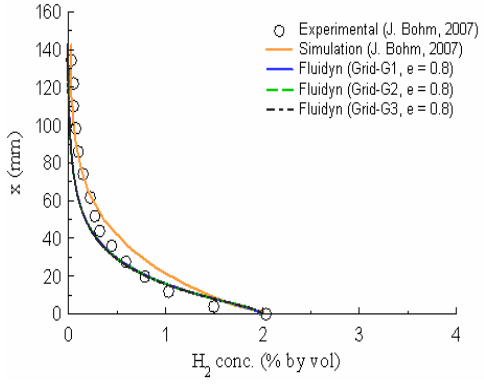


Fig. 5: Hydrogen concentration profiles

Three Channel Cases

In order to simulate REKO-3 prototype results using the present analytical model, 4 catalyst plates along with walls of the facility were considered. For simulation purpose, only plates P1, P2 and the left box wall were chosen, since the system has symmetry with axis S2 as shown in Fig. 3. The temperature profiles were calculated at the surfaces of P1 and P2, while hydrogen concentrations were calculated along planes S1 and S2. From the earlier grid sensitivity studies, since the results with grid G2 were found to match reasonably well with experimental data

and Grid G3 was found to be computationally expensive, grid G2 was used for the present simulations. As part of the studies for three channel geometry, several simulations were performed by changing the gap between the catalyst plate and the box wall as this dimension is not reported in the respective references. The outer wall of the box was modelled as an insulated boundary. Typical results for the temperature and concentration profiles are given in Fig. 6 and Fig. 7.

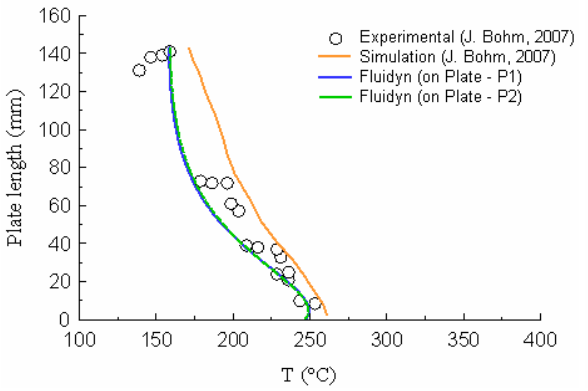


Figure 6: Catalytic plate temperature profile

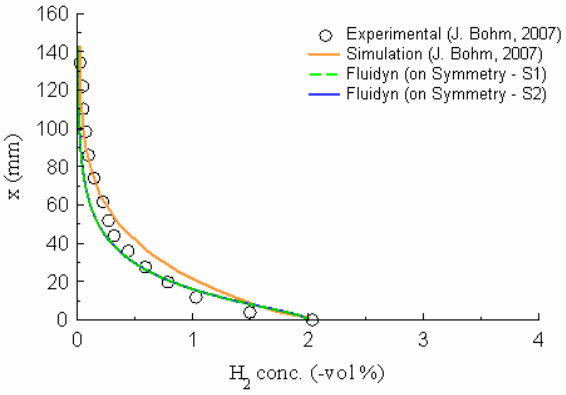


Fig. 7: Hydrogen concentration profile

These results also present the analytical results published by Bohm [10]. Fig. 8 and Fig. 9 depict the typical contours for temperature on the catalyst plates and hydrogen concentration in the fluid space. As can be seen from these results, only the lower portion of the catalytic plate is seen to be participating in the catalytic reaction. This can be attributed to lower inlet H_2 concentrations (2%) used in the reported experiment.

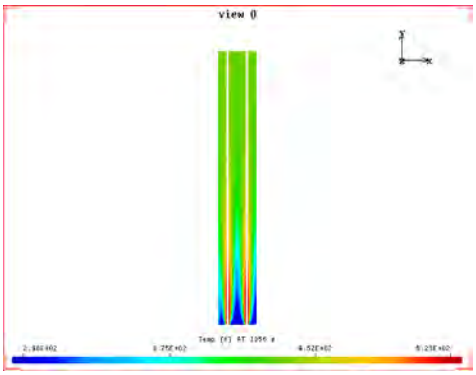


Fig. 8: Contour plot for Temperature

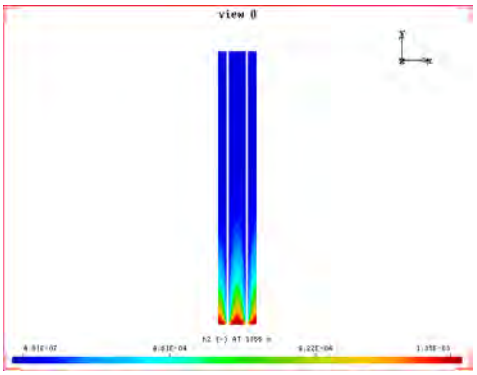


Fig. 9: Contour plot for H_2 concentration

Parametric Studies

Several parametric studies were performed using grid-G3 for a single channel to study the effect of parameters listed in Table 2. It may be noted that experimental results are reported only for different values of the (i) inlet concentration and (ii) inlet flow velocity of hydrogen given in Table 2.

Table 2

Parameter	Value
Concentration of H_2 at inlet	• 1% , 2%, and 4 % (v/v)
Pitch between plates	• 4.25 mm, 8.5 mm **(actual case),** 17.0 mm
Plate height	• 71.5 mm, 107.25 mm, and 143.0 mm **(actual case)**
Flow rate at inlet	• 0.25 m/s, 0.5 m/s and 0.8 m/s

Typical results to show the effect of variation of concentration and flow rate of hydrogen at the inlet against the experimental results are given in Fig. 10 to Fig. 13. It can be seen that the steady state concentration profiles are matching well with experimental results for all the values of concentrations at inlet. The temperature profiles for the cases with 1% (v/v) and 2% (v/v) are closer to experiments but this is not the case for 4% (v/v). The model predicts higher temperatures at the leading edge of the plate for the higher concentration case. This could be attributed to the dependence of gas diffusivity on temperatures which was not accounted for in the simulation. In the upper part of the plate, since the availability of hydrogen is less (Fig. 11), plate temperatures are less and as diffusivity does not change much, the results in this region of the plate match well with the experimental results.

For the simulations with varying inlet flow rates, the inlet H_2 concentration was maintained at 2% (v/v). The plate temperatures match with the experimental results whereas, as can be seen from Fig. 13, the hydrogen concentration profiles are deviating from experimental results at higher velocities. It was noted that the grid sensitivity study was performed only for the low velocity cases. For the higher velocity cases, gradients are expected to be steeper near the walls and hence the grid would need to be further refined for accurate modelling of the experiments at higher velocities.

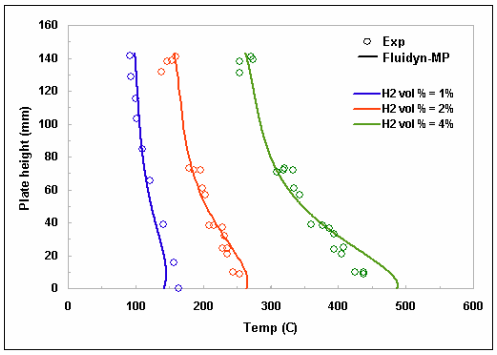


Fig. 10: Catalytic plate temperature profile with different concentrations of H_2

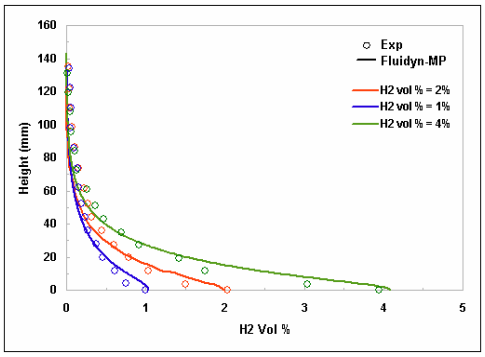


Fig. 11: Hydrogen concentration profile with different concentrations of H_2

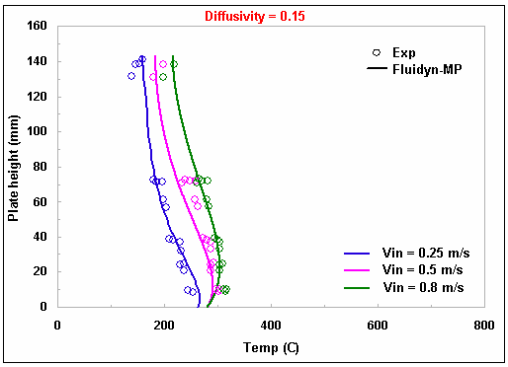


Fig. 12: Catalytic plate temperature profile for different flow rates

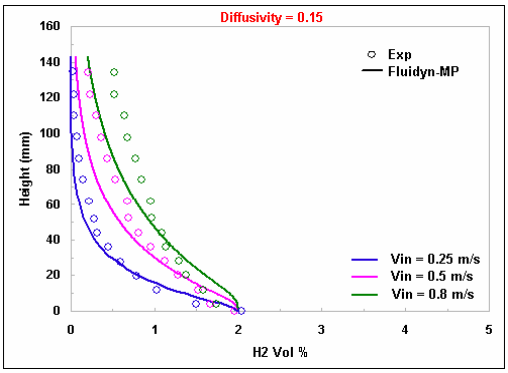


Fig. 13: Concentration profile for different flow rates

CONCLUSIONS

A model for catalytic recombination of hydrogen in box-type passive autocatalytic recombiner has been developed and validated for dry atmospheric conditions using experimental data from open literature. This model has been integrated into the CFD code FLUIDYN-MP. Validation studies have been performed for single channel and three channel configurations reported in literature. Additional parametric studies have been performed to check the effect of inlet flow rates and concentration, pitch between plates, diffusivity of hydrogen and height of the plates. The code predictions are generally in good agreement with the test data. For low velocity cases, less than half of the

plate height has been shown to be sufficient to achieve the experimentally observed recombination rates. The model can be further used to optimize the recombiner design parameters.

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