Hydrogen Recombiner

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Hydrogen Recombiner CFD Model: Development, Benchmarking and Performance Evaluation

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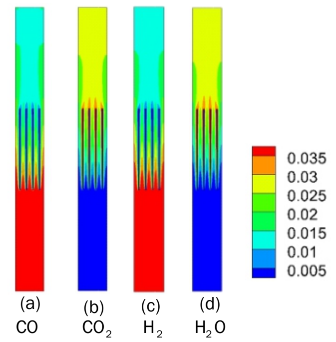

Mole fraction contours of various species (CO= 4% (v/v) & H_2=4% (v/v) at inlet)

ABSTRACT

A significant quantity of hydrogen may be released into the containment of water cooled nuclear power reactors in case of postulated loss of coolant accident followed by non-availability of emergency core cooling system and moderator cooling in PHWR. This hydrogen gets distributed in the containment and may form combustible mixture with the available air. As a part of accident management program, hydrogen recombiners are being deployed worldwide in all water-cooled power reactors to manage the hydrogen concentration inside containment. The recombiners must be evaluated for their performance under various conditions such as their placement at various positions, their efficacy in presence of CO gas. Detailed experimental and modeling work of recombination process is required for their performance evaluation under different conditions. In this work, development of recombiner model and its integration to general purpose CFD code is reported. The developed model has been benchmarked against national and international experimental data. The benchmarked model has been further used to evaluate performance of recombiner under different conditions.

KEYWORDS: *Hydrogen recombiner, Computational Fluid Dynamics (CFD)*

Introduction

Passive Autocatalytic Recombiners (PARs) are installed in water-cooled nuclear power reactor containments to manage the combustible gas released during a postulated severe accident condition. In a loss of coolant accident with unavailability of emergency core cooling system and moderator cooling system, the high temperature Zircaloy cladding oxidizes with available steam and hydrogen is formed. The generated hydrogen gets distributed in the containment and depending on the concentration of air, steam and hydrogen and available small ignition source, uncontrolled combustion can occur in the containment building, which represents a threat to the integrity of the last radiological barrier. Various techniques have been developed by industries depending upon the requirements to mitigate the hydrogen consequences such as pre-inerting, post-accident inerting, electrically operated recombiner, catalytic recombiner, igniters, mixing by use of fans and layout/geometry which promote mixing etc. Catalytic recombination of hydrogen with oxygen is the most popular method adopted by many countries (IAEA-TECDOC-1196) [1]. PARs are passive as they are self-starting and self-feeding having no moving parts and no external energy is required. Plate type PAR consists of a vertical channel where plates coated with catalyst are arranged in parallel. Platinum or palladium are used as catalyst as they are noble metals and have the ability to adsorb hydrogen and oxygen. The recombination reaction occurs spontaneously at the catalyst surfaces and the heat of reaction produces a natural convection flow through the enclosure. Water vapor as a product of reaction along with remaining hydrogen air mixture moves upward because of buoyancy and fresh hydrogen air mixture enters through the bottom inlet section (Bachellerie et al., 2003) [2]. Most of the PARs are plate type; CFD modelling of plate type PAR, validation with national and international data and integration of CFD model with general purpose CFD code have been discussed in the subsequent sections. Recombiner performance has also been evaluated for (i) placing it near to wall or central region (ii) placing it at different elevations and (iii) presence of carbon monoxide (CO) in addition to H_2.

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Mathematical Model of Plate Type Recombiner

For PAR modelling, the interaction of reaction kinetics, heat transfer and associated fluid flow in the catalytic recombiner and radiation heat loss from plates to surroundings are modeled. The simplified geometry of plate type PAR is shown in Fig.1. The governing mass, momentum, energy and conservation equations for hydrogen, oxygen, nitrogen and water vapor have been solved. Surface reaction takes places at catalytic plates. This reaction can be modeled by detailed reaction mechanism as mentioned by Deutschmann and another one proposed by Kasemo both reported by Appel et al., 2004 [3]. However, detailed reaction mechanism is computationally expensive, hence in present work the reaction has been modeled as a one-step reaction mechanism as proposed by Schefer, 1982 [4].

$$H_2 + 1/2O_2 \rightarrow H_2O \qquad (1)$$

Where reaction rate (k) in kmol/m^2/s is given as

$$k = 14\exp(-14.9 \times 10^6 / R_u T)\,[H_2] \qquad (2)$$

Where R_u is Universal gas constant, (8314 J/kmol K), T is Absolute temperature of the mixture, (K) and [H_2] is Molar Concentration of Hydrogen, [mol/m^3]. The above reaction

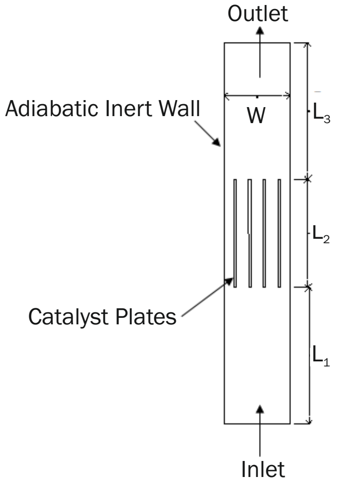

Fig.1: Simplified geometry of Plate Type PAR for 3D CFD computation.

provides additional source term for energy equation. The heat of reaction of the above reaction is 119.96 MJ/kg. The equation also provides source term in species transport equation for water vapour and sink term in species transport equations for hydrogen and oxygen. All these source or sink terms were applied at the very first fluid cell adjacent to solid plate. The high energy output from the reaction results in a considerable heat transmission from surfaces by convection and radiation to environment. Catalyst plates have been modelled as conducting walls to model the conjugate heat transfer in solid plates. The external wall has been modelled as no-slip adiabatic wall (Gera et al., 2011a [5]).

Benchmarking of Plate Type PAR Model

Plate type PAR model has been benchmarked with experimental data of REKO test facility (Reinecke et al., 2004) [6]. REKO test facility consists of a vertical flow channel with a rectangular cross-section 46 mm wide (W) and 146 mm deep. The channel is 504 mm long (L_1+L_2+L_3 (Fig.1)) with about 180 mm of channel length above the catalyst plates (L_3 (Fig.1)). For the reported experiments four plates made of stainless steel and coated with wash coat/platinum catalyst material were arranged in parallel inside the flow channel. The plates used were 1.5 mm thick and 143 mm long (L_2 (Fig.1)) and 146 mm deep.

Experiments have been performed for different inlet flow velocities (0.25, 0.50, and 0.80 m/s) at different inlet temperatures (298, 343 and 383 K). Inlet hydrogen concentrations were varied between 0.5 and 4% v/v. Fig.2 and 3 show the comparison of numerical results with experimental data for hydrogen mole fraction and catalyst plate temperature, respectively. Results are compared for two different inlet hydrogen concentration of 2% and 4% having inlet temperature of 343 K and velocity as 0.8 m/s. Results are found to be in good agreement with the experimental data. Fig.3 shows the temperature and hydrogen concentration contour plots for air hydrogen mixture of 2% v/v hydrogen concentration entering at 383 K for REKO test facility.

Mathematical Model for Integration of Recombiner with Containment model

A detailed analysis is required to optimize the passive autocatalytic recombiner locations and their numbers in nuclear containment. 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 complex. This is because the mesh size requirement to fully resolve the recombiner plates requires 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. Thus, a simplified approach is required to model recombiner with containment CFD model. Numerous experiments have been performed on various types of recombiners to study the aspects like efficiency, start-up condition, hydrogen conversion rate, effect of operating pressure, temperature and other inlet conditions. Based on these experiments, empirical correlations have been developed to compute the hydrogen removal rate of particular make recombiners. 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 empirical correlations (Bachellerie et al., 2003) [2]. For solution, as a simplified approach to integrate the recombiner model with containment CFD calculation, these correlations can be used to define localized volumetric sink of hydrogen, oxygen and source of energy and water vapour in the CFD codes. The transient Navier-Stokes equation with the energy

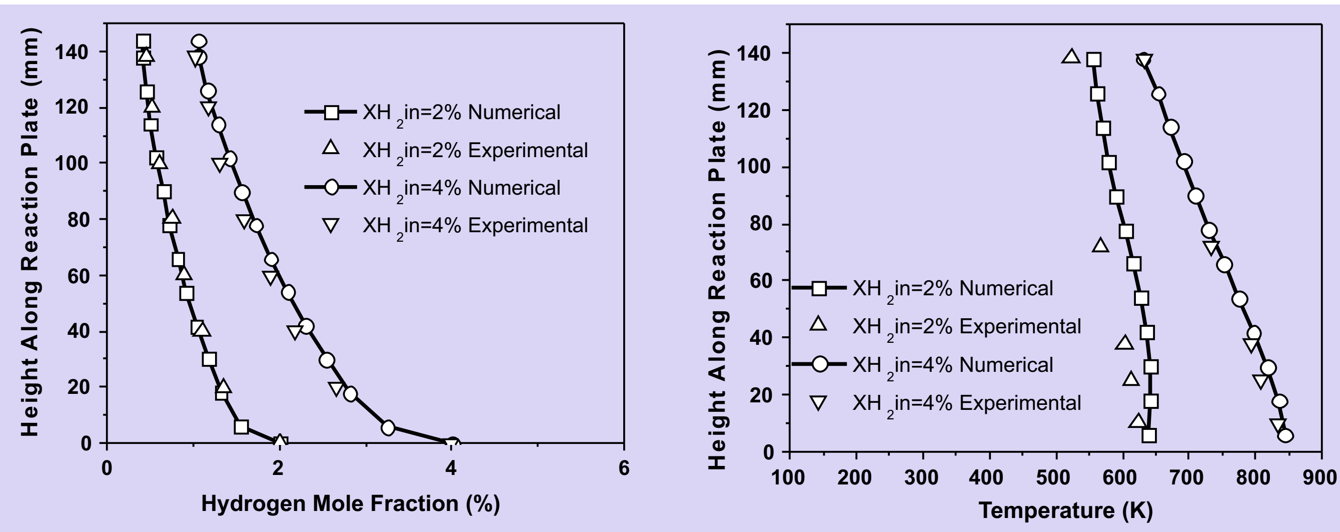

Fig.2: Steady state average hydrogen mole fraction in the recombiner section of REKO facility (left), catalyst surface temperature along the catalytic plate (right), (Inlet hydrogen mole fraction 2% & 4% and inlet temperature 343 K).

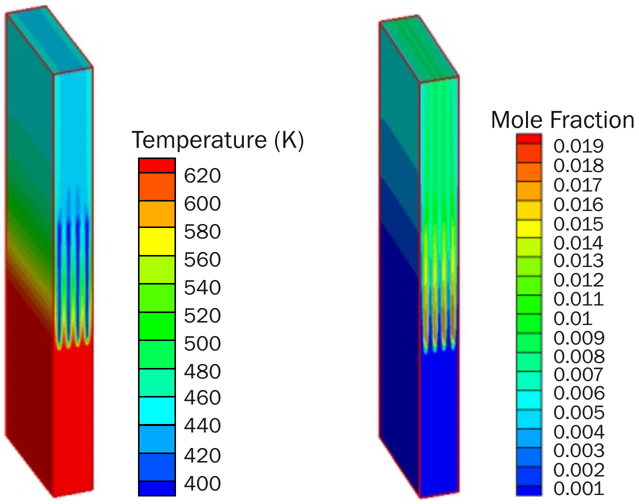

Fig.3: Temperature contours (left) and hydrogen mole fraction contour (right) for simulation performed for REKO facility (Inlet hydrogen mole fraction 2% v/v and inlet temperature 383 K).

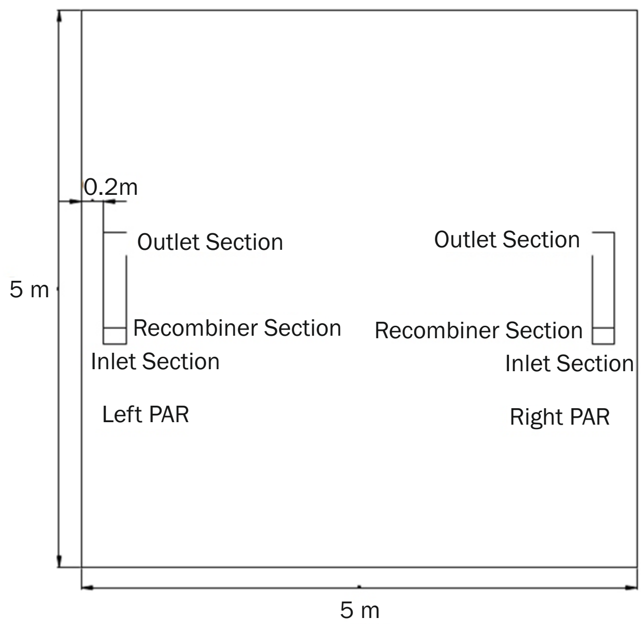

Fig.4: PARIS-1 model geometry with 2 PARs.

and species transport was solved using commercial CFD software (CFD-ACE+, 2009) [7]. To model the mixture behaviour of hydrogen, air and steam, a continuum approach was used, in which only one velocity field was defined using the average density of gas mixture. The independent behaviour of hydrogen, oxygen and steam was considered using species transport equation. The recombiner section was modelled as fluid medium. The hydrogen conversion was modelled by means of the empirical correlation. 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. First the hydrogen conversion rate was evaluated based on the reported manufacturer's correlation then it was used to obtain the solid plate temperature based on heat balance. Then the heat convected by fluid mixture through the recombiner section was computed and has been used as source term in energy equation at recombiner section. Based on the hydrogen conversion rate corresponding sink term for hydrogen and oxygen and source term for steam was incorporated through user coding. Recombiner boundaries have been modelled as adiabatic no-slip wall (Gera et al., 2011b [8]).

Benchmarking of Integrated Recombiner Model

The integrated recombiner model was benchmarked with PARIS-1 (PAR interaction studies) model geometry. As part of this study, two PARs of the AREVA design were considered in a 2D rectangular domain as shown in Fig.4. Height of PAR (h) was 1 m, and width (w) was 0.2 m. PAR entry and exit section widths were also equal to 0.2 m. There were two PARs located in the containment. The main purpose of the benchmark simulation was to observe the containment atmosphere mixing phenomena during a postulated severe-accident scenario. The results were compared with the CFD results obtained by Babić et al. (2006) [9].

Fig.5 shows the amount of total hydrogen and the pressure variation in the representative containment. As the hydrogen is consumed energy gets generated because of recombination process. A low pressure region is created at the recombiner section and hydrogen moves from bottom of the recombiner towards top outlet. Fig.6 shows the hydrogen mass fraction contour inside the representative containment at 20 and 300 seconds. Initially the hydrogen concentration is high, the reaction rate is fast hence consumption of hydrogen mass is rapid and there is fast pressure rise. The rate of hydrogen

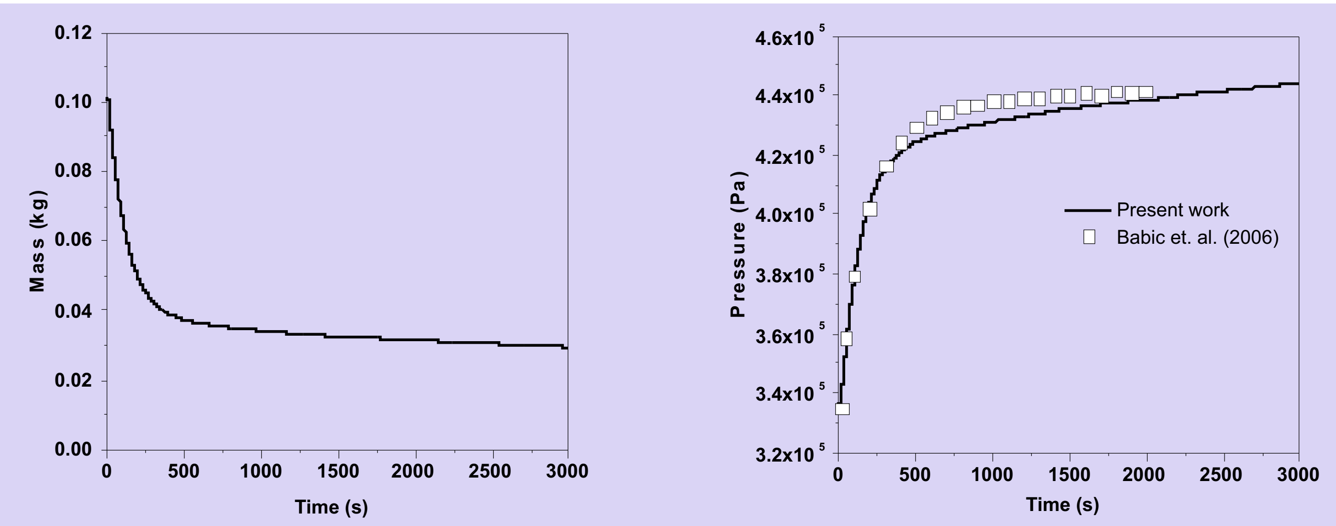

Fig.5: Average hydrogen mass (left) and average pressure (right) in the representative containment.

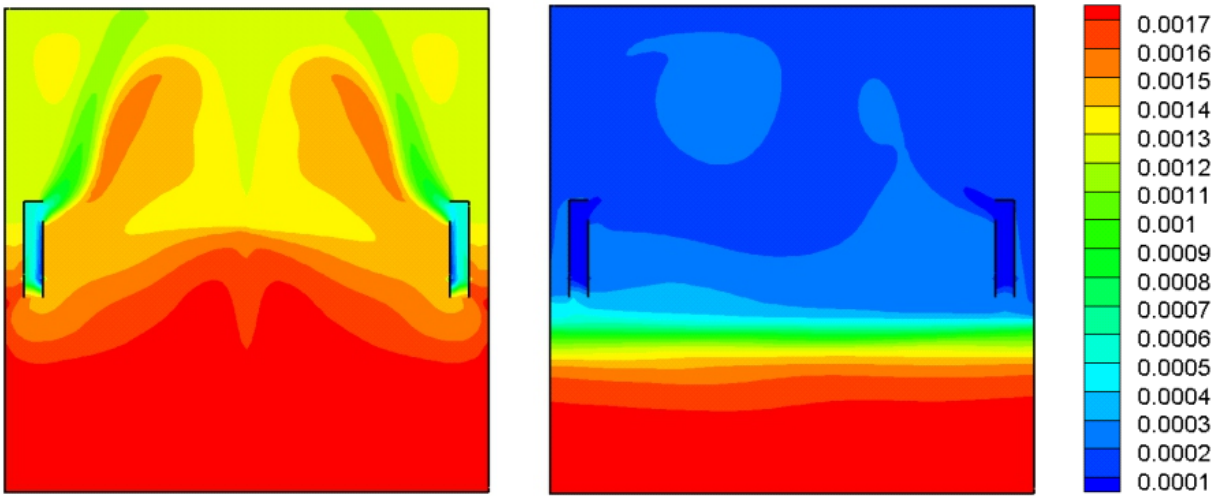

Fig.6: Hydrogen mass fraction at 20 s (left) and 300 s (right).

consumption and rate of pressure rise decreases as the hydrogen is consumed. After 300 seconds the upper region has low density due to high temperature. The cold unconsumed hydrogen remains in lower part and diffuses slowly towards PAR.

Benchmarking with National Data

The numerical approach was validated against experiment conducted at Hydrogen Recombiner Test Facility (HRTF) at Tarapur. HRTF is a cylindrical vessel of diameter 2.98 m with torospherical heads. Its total height is 9.12 m. The geometry and mesh of the vessel with PAR and injection pipe is shown in Fig.7. The PAR box has the dimensions 1.02 m (height) X 0.88 m (length) X 0.425 m (width) and is oriented above the injection pipe such that the length dimension is along the pipe direction. The recombiner box has been modelled as no-slip walls and is interacting with the vessel through top and bottom open section. As a part of validation, hydrogen injection experiment corresponding to 7.5% (v/v) of vessel volume was considered. The top and bottom sections of recombiner box are open and all sidewalls have been considered as no slip wall. No heat or mass transfer (steam condensation) occurs on the vessel walls. The simulation was

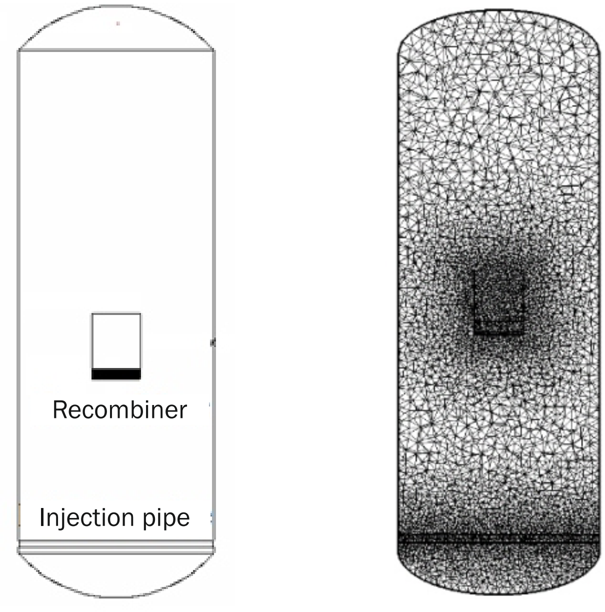

Fig.7: HRTF geometry (left) and mesh (right), PAR at center configuration.

carried out and the results were predicted in terms of total hydrogen concentration variation at the inlet and the outlet with respect to time. These results were then compared with the results obtained from experiment performed at the test facility. As is seen from the Fig.8, there is good agreement in the rate at which hydrogen is getting consumed which is indicative of the rate of reaction between the experiment and CFD model. Due to proprietary nature of HRTF data, Y-axis numerical values are not shown. Thus, one can safely say that this particular CFD model can predict the reaction rate without much deviation from the experimental result.

Effect of Recombiner Placement

As part of the regulatory approval for the installation of PARs in the nuclear containment, it has been highlighted that the basis for number and location of the PARs is based on lumped parameter analysis, where effect of turbulence and buoyancy cannot be accurately modelled. It is required to have detailed assessment of the realistic flow fields around the PAR so that the adequacy of the location of the PARs can be confirmed. PARs are generally qualified in small to medium scale facilities worldwide. However, due to the fixed "recombiner size to vessel volume ratio" in these tests, the behaviour of the recombiner in large-sized containment volumes cannot be accurately predicted. It is required to understand the PAR behaviour in a large size room and to investigate the zone of influence of a PAR. Detailed CFD calculations in this regard will help to optimize the location of the PARs in different compartments of the containment. In the present section CFD simulations have been performed to evaluate the PAR performance in 210 m^3 enclosure and PAR was positioned at centre or side location. CFD simulations have also been performed to evaluate the PAR performance in 2250 m^3 enclosure and PAR was positioned at side location at top, bottom or central position. Different cases were studied by varying (i) computational volume (ii) hydrogen injection duration corresponding to total hydrogen concentration (iii) recombiner position (side wall and center region) (iv) effect of placing recombiner at different heights.

Case 1: Injection of hydrogen in 210 m^3 enclosure having 60% (v/v) steam and rest air. Initial pressure and temperature 2.0E+05 Pa and 388 K. Recombiner is placed at the center of enclosure. Total mass of 2.1 kg of hydrogen was injected in 1800 s (this corresponds to 8% v/v uniform hydrogen concentration in the volume if hydrogen is properly mixed in the volume).

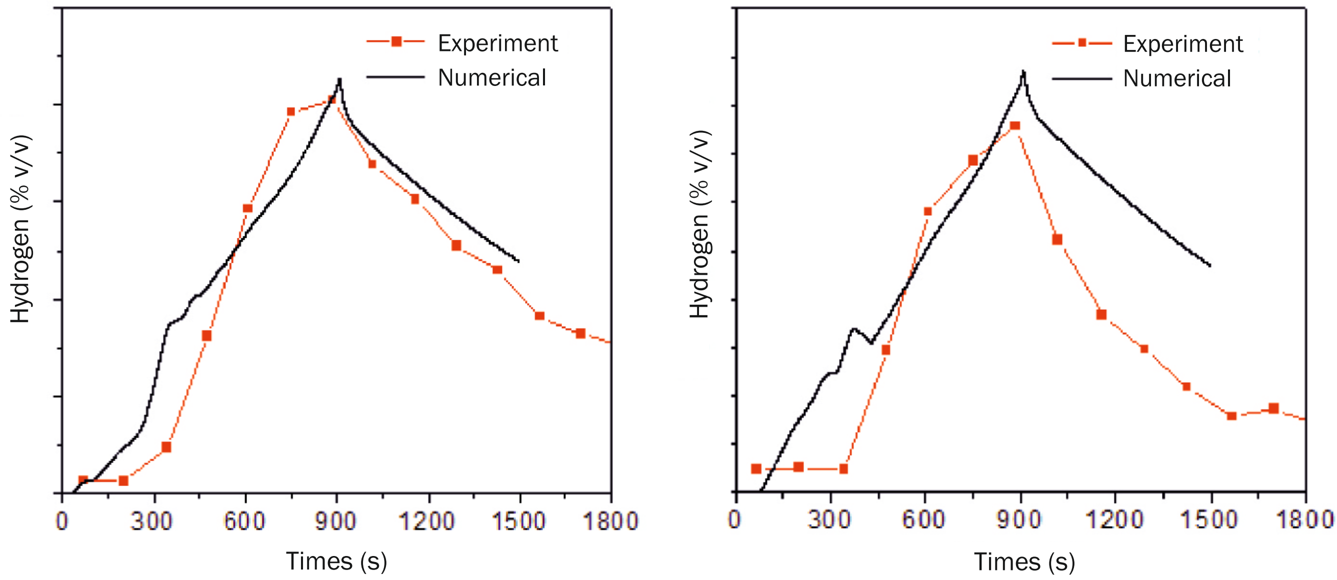

Fig.8: Temporal variation of the hydrogen concentration at PAR inlet (left) and outlet (right).

Case 2: Recombiner is placed at the side location in the enclosure. Other conditions are similar to Case 1.

Case 3: Injection of hydrogen in 2250 m^3 enclosure having 60% (v/v) steam and rest air. Initial pressure is 2.0E+05 Pa and temperature is 388 K. Recombiner is placed at the side location at bottom position in enclosure. Total mass of hydrogen injected 22.3 kg in 1800 s, (this corresponds to 8% v/v uniform hydrogen concentration in the volume if hydrogen is properly mixed in the volume).

Case 4: Recombiner is placed at the side location at middle position in enclosure. Other conditions are similar to Case 3.

Case 5: Recombiner is placed at the side location at top position in enclosure. Other conditions are similar to Case 3.

Recombination of hydrogen and oxygen takes place at recombiner section and due to heat generation natural circulation loop is established in enclosure. The mass of hydrogen decreases with time as the recombination rate and plate temperature increase. The average temperature inside the vessel increases with time as the recombination progresses. Due to this natural circulation hydrogen in the top section of enclosure is depleted at much faster rate than the section below the recombiner. It can also be seen from the Figs.9-10 where contours of mole fraction of hydrogen are seen at different time intervals. The temperature of upper layers is higher. The pressure and temperature inside the vessel increase with time as the recombination process progresses. If the PAR is placed at side location, its performance is slightly deteriorated as compared to centre location as shown in Fig.11. This can also be seen from Figs.9 and 10. However in actual reactor; most of the PARs are placed near wall. There is slightly delay in reaction to start in side wall placed recombiner (case-2) in comparison to centrally placed recombiner (case-1). All these results show that PAR is able to remove hydrogen in 210 m^3 enclosure. After that its performance was evaluated for big enclosure size 2250 m^3. The PAR performance is evaluated by placing recombiner at side at three different elevations (case 3 to 5). It was found out that PAR is able to mobilize the gases in such large volumes. In case 3 better mixing is observed, while in case 5 stratified layer is formed with higher concentration of hydrogen near injection level as depicted by Figs.12-13. In case of bottom configuration (Case 3) a better mixing is observed.

Effect of Presence of CO in Addition to H_2

Carbon monoxide (CO) may be generated inside the containment due to molten corium concrete interaction (MCCI) during a severe accident condition when vessel fails and corium comes in contact with raft concrete. The generated CO will interact with passive autocatalytic recombiners (PARs), which are installed inside nuclear reactor containments for H_2 removal. Depending on the conditions, CO may either react with oxygen to form carbon dioxide (CO_2) or act as catalyst poison, reducing the catalyst activity and hence the hydrogen conversion efficiency. CO is catalyst poison for platinum if

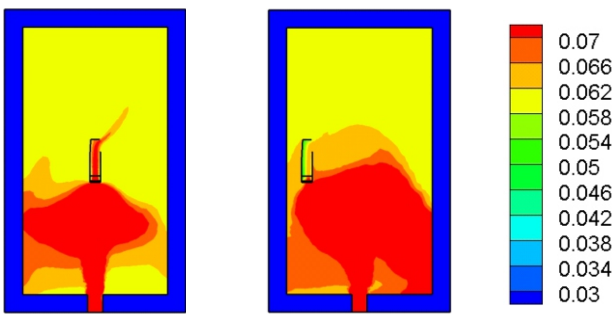

Fig.9: Hydrogen mole fraction contours at 1800 second for case 1 (left) and case 2 (right).

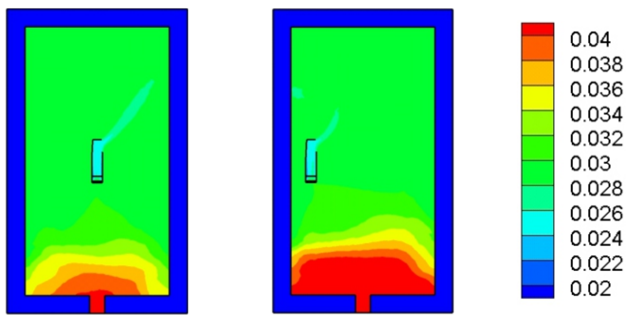

Fig.10: Hydrogen mole fraction contours at 6000 second for case 1 (left) and case 2 (right).

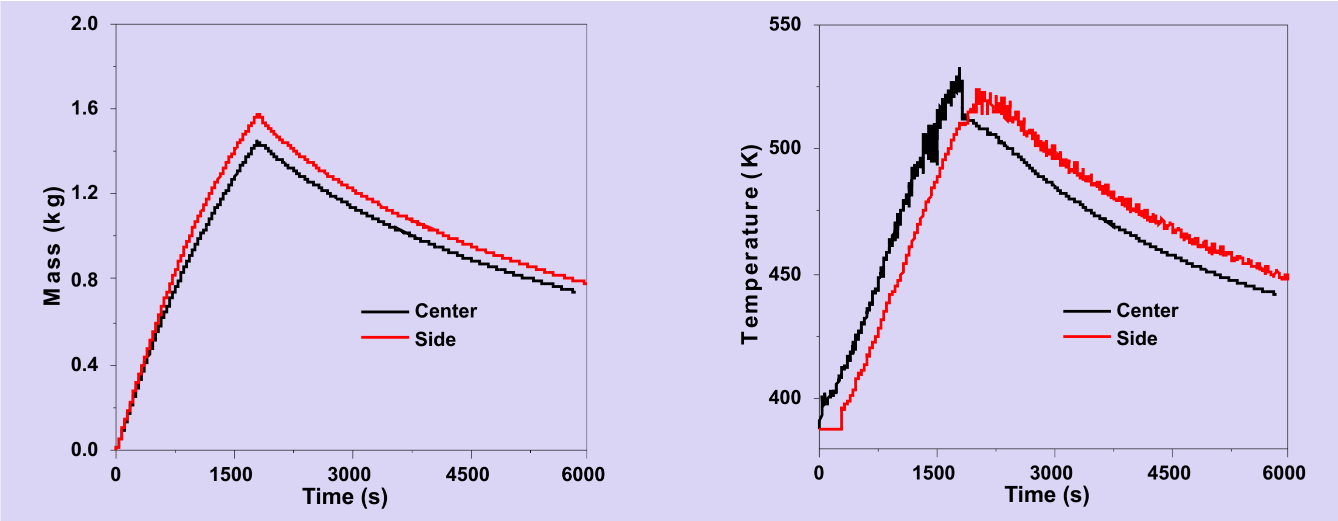

Fig.11: Temporal variation of total hydrogen mass (left) and PAR outlet temperature (right) in enclosure for case 1-2.

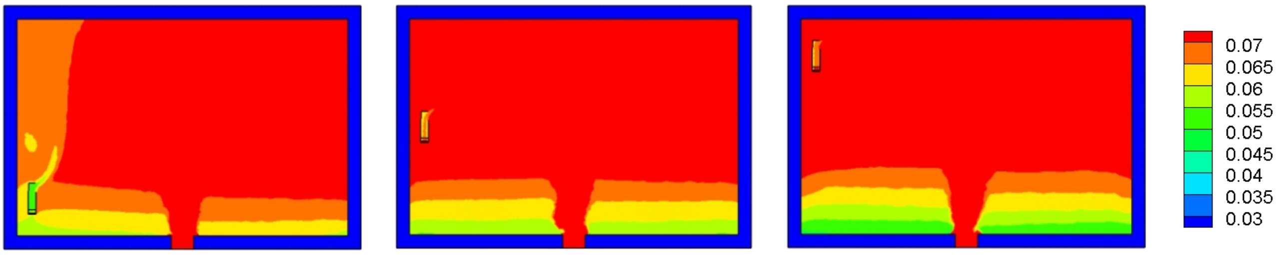

Fig.12: Hydrogen mole fraction contours at 1800 second for case 3-5.

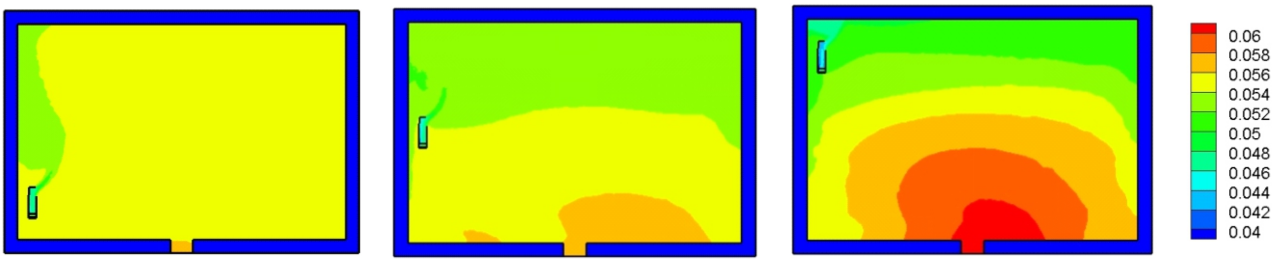

Fig.13: Hydrogen mole fraction contours at 18000 second for case 3-5.

injected before hydrogen, but if hydrogen is injected first, CO gets recombined due to high temperature and its role is to further increase the gas and plate temperature. Thus, CO has the potential to further increase the catalyst plate temperature, which may lead to the self-ignition temperature of combustible mixture. Hence, the effect of CO on catalyst plate temperature and on PAR removal rate needs to be investigated. For the present numerical study, a 2D CFD model has been developed using commercial software CFD-ACE+. Simulations were performed using this model to determine the effect of CO on catalyst plate temperature with 2 & 4 % (v/v) H_2 and 0-4 % (v/v) CO in air at the recombiner inlet for a reported experiment conducted at the REKO-3 test facility (Klauck et al., 2014) [10]. Fig.14 shows the result of the computation and comparison with experimental measurement. In these cases, H_2-air-CO mixture enters at a velocity of 0.5 m/s at 293 K with inlet H_2 concentration of 2 and 4% v/v and CO concentration 0-2% v/v. The results of present computation are in very good agreement with available data. As the mixture enters in the recombiner section the reaction occurs at the leading edge of the catalyst plate. The reaction rate is highest at the leading edge of the plate. This is manifested by maximum catalyst surface temperature near the leading edge of the plate. As flow takes place over catalytic plate, boundary layer is formed over the plate surface and H_2/CO diffuses from bulk of the mixture towards plates for recombination. With the flow along the catalyst plate, concentration gradient decreases thus reaction rate also decreases along the catalyst plate.

Both the recombination reactions are exothermic and increase the plate temperature. The addition of CO leads to further increase in plate temperature. The maximum temperature of plate is 600 K when H_2 only for 2% v/v at inlet and 720 K for 2% v/v H_2 and 2% v/v CO at inlet. The depletion of H_2 and CO and formation of H_2O and CO_2 are shown in Fig.15 for one of the case. The species boundary layer formation is clear from this Fig.15 (Gera et al., 2023 [11]).

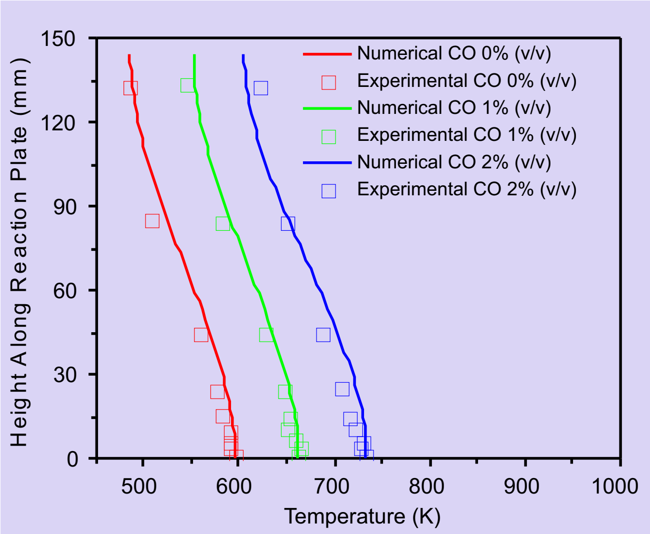

Fig.14: Comparison of computed plate temperatures with experiments for different CO concentration at inlet (H_2 2% (v/v) at inlet).

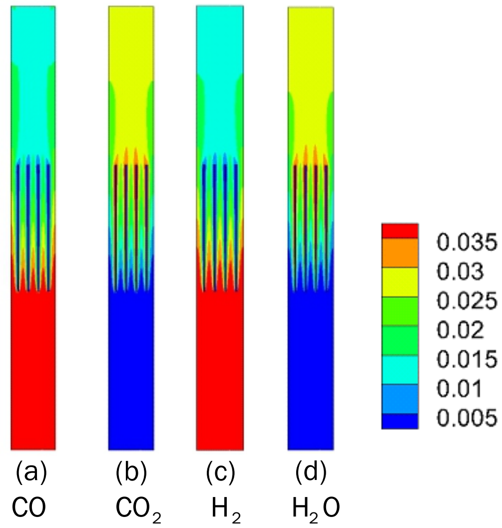

Fig.15: Mole fraction contours of various species (CO= 4% (v/v) & H_2=4% (v/v) at inlet).

Conclusions

A CFD model for plate type hydrogen recombiner has been developed and validated against REKO experimental data. The simplified model of PAR has been integrated with general purpose CFD code used for simulating hydrogen distribution and the integrated model is verified with PARIS benchmark. The integrated code has also been validated against national data. The developed model has been utilized to evaluate the recombiner performance for different conditions such as (i) placing it near to wall or central region (ii) effect of placing recombiner at different elevations and (iii) presence of CO in addition to H_2. It was found that performance of PAR at central region is better than placement of PAR at near wall region. In case of placing PAR at bottom elevation better mixing is observed. It was also found that recombiner maximum plate temperature is further increased by adding CO.

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