

Session 3: Containment issues, Paper N° 3.1**Review of LP&CFD recombiner modelling and experiments:
applicability to reactor simulations and open issues**

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Summary

In case of a severe accident in light water reactors, a large amount of hydrogen could be generated from the reaction between steam and zirconium at high fuel clad temperature and also from reactions of molten core debris with concrete. The hydrogen generated will be released in the containment atmosphere, mix with air and steam and possibly create local flammable conditions. In order to prevent loads resulting from a possible hydrogen combustion, French and German reactor containments are equipped with passive autocatalytic recombiners (PARs) which recombine hydrogen with oxygen already at concentrations below the flammability limit. In common PAR designs, catalytic materials (platinum and palladium on ceramic washcoat) are housed in a metallic structure whose purpose is to optimize the circulation of gases in contact with the catalyst.

Numerous tests have been conducted in the past to investigate the PAR behaviour in situations representative of severe accidents (Battelle Model Containment in Germany, H2PAR and KALI H2 in France, AECL Whiteshell Laboratories in Canada, etc.). Furthermore, these tests demonstrated that, provided that special care is paid to the design and construction of the catalysts, catalyst poisoning by materials such as carbon monoxide, iodine and aerosols present in the containment atmosphere will not reduce fundamentally the effectiveness of the PARs.

Some of the mentioned tests show also that PARs could ignite the flammable gas mixture at elevated hydrogen concentrations. These experimental results need however to be corroborated by more detailed experiments and by refined modelling of phenomena occurring in PARs. In order to better characterise the PARs induced ignition risk, a series of dedicated experiments has started at the REKO-3 facility located in Forschungszentrum Jülich (FZJ). In parallel, a refined modelling of the recombiners has been developed by IRSN and will be used to have an insight of phenomena occurring at the PAR catalyst plates.

Furthermore, previous tests indicated that the position of recombiners could have an impact on their overall efficiency. The installation of PARs in the reactor building is influenced by geometric and operational constraints; numerical models were developed from the experimental data for codes like COCOSYS or ASTEC in order to optimize the PAR location and to assess the efficiency of the PAR implementation in different scenarios. However, these models are usually simple (black-box type) and based on the manufacturer correlation to calculate the hydrogen depletion rate. Recently, enhanced CFD models have been developed at IRSN and FZJ in order to take into account phenomena such as PARs location effect, gas mixture ignition induced by PARs, and oxygen starvation effect. A new specific instrumented facility is also under construction at FZJ to investigate these phenomena in more detail.

Session 3: Containment issues, Paper N° 3.1**A. INTRODUCTION**

The nuclear safety is based on the “defence-in-depth” concept, introduced by the International Atomic Energy Agency (Ref. [1]). This concept consists of hierarchical levels of equipment and procedures to preserve the integrity of the physical barriers. The reactor containment constitutes the last physical barrier to the release of radioactive fission products into the environment in case of a light water reactor (LWR) hypothetical severe accident.

During severe accidents (SA) in LWR, hydrogen is generated by the exothermal oxidation of the fuel claddings, hot metallic components, and after failure of the reactor pressure vessel and melt relocation to the reactor pit due to decomposition of concrete as well. The release rate and the total amount of hydrogen depend on the particular severe accident scenario and also on the reactor type. These processes produce hydrogen with such high rates and for such a long term in the course of a SA that the local volumetric concentrations inside the containment exceed the lower flammability limit. Both major nuclear accidents of Three Mile Island (TMI) in 1979 and Chernobyl in 1986 resulted in world-wide investigations on SA and the hydrogen risk. The protection of the containment against the consequences of a hydrogen deflagration or even explosion is an integral part of the forth level of defence-in-depth concept (“prevention of accident progression and mitigation of the consequences of severe accidents”).

In order to prevent the hydrogen explosion hazard in the reactor building, risk management strategies can be implemented by one or a combination of the following methods:

- the deliberate ignition of the mixture as soon as the flammability limit is reached,
- the consumption of hydrogen,
- the removal of oxygen,
- the dilution of the atmosphere to prevent the formation of flammable mixtures either by the increase in the volume of the containment, or by the injection of an inert gas.

The choice of a mitigation strategy depends primarily on the design of the containment. For PWRs with large dry containment, the strategy usually consists in combining large free volume to allow dilution, a high value of the design pressure and the use of means, as passive autocatalytic recombiners (PARs), to consume hydrogen. This strategy has been adopted in a large number of European pressurized water reactors (PWR).

Hydrogen and oxygen react exothermally on the catalytic surfaces inside a PAR generating steam and heat beyond conventional flammability limits (Ref. [2]). The reaction heat creates buoyancy driven flow which makes a PAR a completely passive operating device. Present commercially available and implemented PAR designs are box-type PAR with catalyst sheets from AREVA, France/Germany and AECL, Canada, or with porous catalyst beds from NIS, Germany. Figure 1 shows a PAR of AREVA design, mainly back fitted to European PWR containments.

In the frame of SA analysis, reliable numerical models are a vital element for the assessment of PAR performance and efficiency in accident scenarios. International research activities in the European FP-6 Network of Excellence SARNET (Severe Accident Research NETwork) investigate related aspects. The important modelling issues are on the one hand the phenomena inside the PAR, e.g. the buoyancy driven flow and hydrogen depletion due to the catalytic surface reaction. On the other hand, the interaction of the recombiner with the containment atmosphere in particular the PAR outflow and its influence on the mixing

Session 3: Containment issues, Paper N° 3.1



Figure 1: AREVA-design PAR inside a German PWR containment

process, the inducing of natural convection by the hot PAR box housing, and the effect of forced flow conditions need to be addressed. Safety related criteria such as possible gas-phase ignitions caused by hot parts due to the exothermal reaction have to be considered as well.

B. STATE OF THE ART: PAR MODELLING IN CONTAINMENT CODES

Numerous integral experiments have been carried out to study manifold aspects of recombiners like the efficiency of hydrogen removal, start-up conditions, poisoning, oxygen starvation, steam and water impact (Table 1).

Table 1: Facilities list

Facility	Country-institution	Shape	Volume	Material
LSVCTF	Canada-AECL	Rectangular	120 m ³	Steel
BMC	Germany-BATTELLE	Cylinder	625 m ³	Concrete
H2PAR	France-IPSN	Cylinder	8 m ³	Terphane
KALI H2	France-CEA	Cylinder	15.6 m ³	Steel

These experiments investigated the global behaviour of a recombiner in a larger environment in order to demonstrate the effectiveness and to facilitate the derivation of simplified ('black-box') models for long-term severe accident analyses. These empirical correlations describe the hydrogen consumption rate for a reference PAR type as a function of the gas composition, temperature and pressure. The hydrogen consumption rates proposed by the different manufacturers are given by the following empirical correlations (Ref. [2]):

Session 3: Containment issues, Paper N° 3.1

$$\text{AECL} \quad \dot{r}_{H_2} = (0.15196 \cdot C_{H_2} + 0.0126 \cdot C_{H_2}^2) \cdot \left(\frac{298}{T}\right)^{1.10974} \cdot p^{0.57769} \quad \left[\frac{\text{kg}}{\text{h}}\right]$$

$$\text{AREVA} \quad \dot{r}_{H_2} = \eta \cdot \min(X_{H_2}, 2 \cdot X_{O_2}, 0.08) \cdot (A \cdot p + B) \cdot \tanh(X_{H_2} / 0.5) \quad \left[\frac{\text{g}}{\text{s}}\right]$$

$$\text{NIS} \quad \dot{r}_{H_2} = 1.134 \cdot C_{H_2}^{1.307} \cdot \frac{p}{R \cdot T} \quad \left[\frac{\text{g}}{\text{s}}\right]$$

where X_i is the volume fraction and C_i the volumetric concentration (mole/m³) of the hydrogen and oxygen respectively, p is the pressure (bar), T the absolute temperature (K) and A and B and the other constants respectively are model parameters which include all residual influences and conditions at the validation experiments and depend on the recombiner model. In the AREVA approach η can be used to describe the PAR efficiency under different conditions (e.g. oxygen depletion or spray). As a first approach these correlations can be implemented by means of volumetric sinks and sources of energy, mass and momentum or as a 'black-box' model directly in LP and CFD codes as described for example in Refs. [3, 4].

In addition to the simplified models based on manufacturer correlations and used e.g. in the GASFLOW code developed by FZK, a one dimensional detailed model has been implemented in ASTEC and COCOSYS (Ref. [5]) codes. The adopted approach subdivides the PAR into three main segments along the height of the box: entry, catalytic plates and chimney (Figure 2).

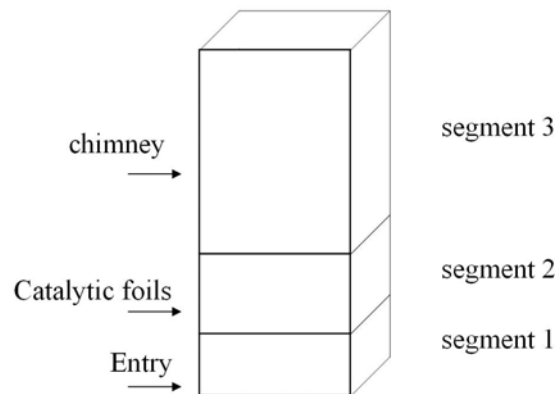


Figure 2: Detailed 1D recombiner modelling approach

For each segment, the thermal hydraulic conditions are calculated. The model considers the catalytic plates as well as the structure of the housing. The heat transfer between the surrounding gas and the structures is calculated by means of free and forced convection, condensation and radiation. The form of reaction equation corresponds to the Arrhenius like expression (Ref. [6]):

$$r = aA c_{H_2}^{n_1} c_{O_2}^{n_2} \exp\left(-\frac{\Delta E}{RT}\right)$$

where $c_{H_2}^{n_1}$ and $c_{O_2}^{n_2}$ stands for the hydrogen and oxygen concentrations. The constants a , n_1 , n_2 , the plate surface A and the activation energy ΔE are adjusted according to experiments.

Detailed and simplified models implemented in ASTEC were validated by IRSN on the basis of H2PAR tests (Ref [4]). Figure 3 shows the good agreement between measured and calculated hydrogen concentrations.

Session 3: Containment issues, Paper N° 3.1

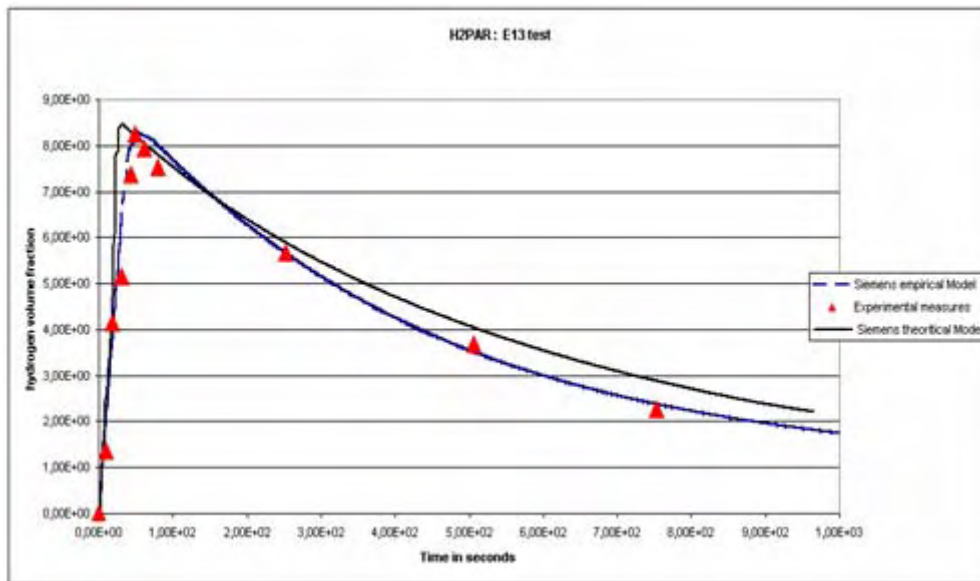


Figure 3: Comparison of AREVA (Siemens) recombiner model with the H2PAR-E13 test

The implementation of a manufacturer correlation into different codes has been applied by participants in the SARNET PAR Interaction Studies (PARIS benchmarks) (Ref. [7]). In the frame of SARNET WP12-2 (containment atmosphere mixing, CAM), PAR interaction studies have been performed in order to generally investigate adopted models and approaches. The first benchmark (PARIS-1) focused on the impact of PAR elevation on the phenomena of stratification. In the frame of this study an AREVA FR90/1-150 like PAR was considered in a 2D rectangular domain. The hydrogen conversion was modelled by means of the empirical AREVA correlation. In PARIS-1 a closed 5 m x 5 m domain fitted with two PARs was addressed. In order to compare the results of different codes solely with regard to the PAR-containment interactions, heat and mass transfer to the walls was not considered.

The starting point for the 3000 s calculation was a homogeneous mixture at 100°C, containing 5 vol.% hydrogen in dry air and steam at saturation conditions. In the first 300 s the flow was nearly symmetric (Figure 4 left) and resulted in a stable thermal stratification in the domain after about 400 s. As thermally driven convection loops have difficulties to mobilise the cold hydrogen rich gas located near the floor below the PAR inlet (Figure 4 right), this scenario develops into a diffusion controlled situation. The elevation of the PARs had an influence on the thickness of the bottom layer, but could not resolve it anyway.

The main reason for the formation of the bottom layer was the artificial assumption of adiabatic walls. In a real situation heat transfer to the compartment walls and condensation would result in an enhanced mixing of the atmosphere. This was demonstrated in a further calculation with constant wall temperatures revealing that the bottom layer is dissolved after 500 seconds. Nearly adiabatic situations are however imaginable and the phenomenon of thermal stratification has occurred in some integral containment experiments. Consequently, the relevance of detailed studies has been affirmed by this benchmark exercise. Currently, PARIS-2 is in progress investigating the impact of forced downward flow conditions on the PAR operation.

Session 3: Containment issues, Paper N° 3.1

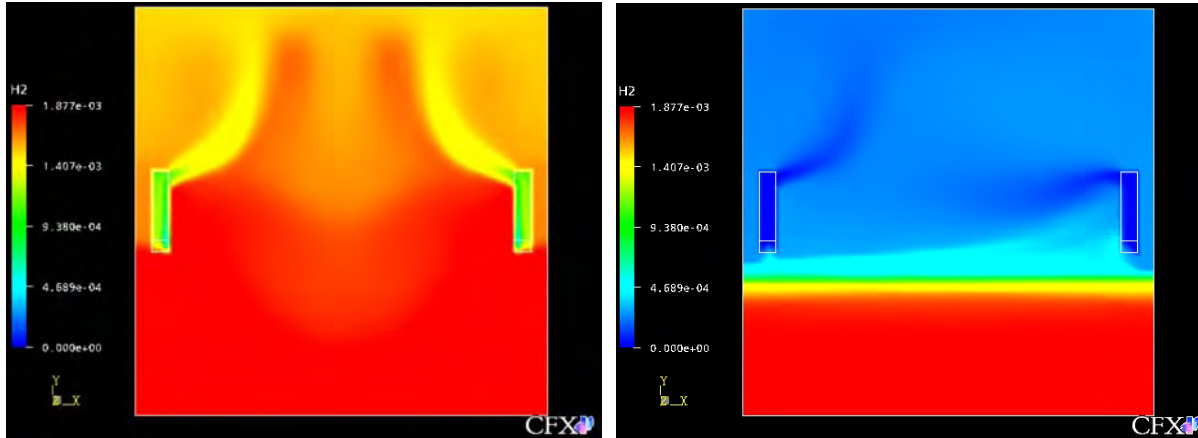


Figure 4: PARIS-1, hydrogen mass fractions: PAR operation after 10s and diffusion controlled scenario after 400s (CFX calculations by FZJ)

The described approaches are suited for optimizing PAR location inside the reactor containment and assessing PAR efficiency in different scenarios. However, the applicability is dependent on the experimental situation from which the correlations were derived and phenomena as e.g. the ignition risk are not covered.

C. RECENT DETAILED STUDIES ON PAR OPERATIONAL BEHAVIOUR

C.1 REKO-3/REKO-4 experiments at FZJ

In order to fill existing knowledge gaps and to complement the existing data base, detailed experiments are performed at the Institute for Safety Research and Reactor Technology (IEF-6) at FZJ. In the small-scale test facility REKO-3, experimental studies on the operational behaviour of a recombiner section are performed under well-defined and steady-state conditions. Currently, a new large test vessel REKO-4 is under construction for testing PAR behaviour under natural convection conditions.

The experimental set-up of the REKO-3 facility (Figure 5) allows the investigation of catalyst samples inside a vertical flow channel under well-defined conditions comprising gas mixture, flow rate and inlet temperature. The catalyst sheets (stainless steel coated with washcoat/platinum catalyst material) are arranged in parallel forming vertical rectangular flow channels. Such a set-up represents a box-type recombiner section of AREVA design. The correlation of the hydrogen conversion and catalyst temperatures with the experimental parameters serve basically to clarify the interactions of reaction kinetics, heat and mass transfer, and the flow conditions inside the recombiner. Inside this configuration, the distribution of the catalyst temperatures and the gas composition in vertical flow direction are measured (Figure 6).

For measuring the distribution of the catalyst temperature the catalyst sheets are equipped with thermocouples. In order not to disturb neither the gas flow nor the catalyst coating, drillings were manufactured inside the rear edge of the plates by means of spark erosion (0.6 mm) enabling thin thermocouples (0.5 mm) to be inserted at different locations inside the samples. These temperature measurements provide very robust data due to optimal contact with the catalyst and avoided flow interaction. For the measurement of the gas concentrations in the flow channel along the catalyst sheets, a probe head is introduced at different vertical positions allowing measurement of the hydrogen and oxygen depletion in

Session 3: Containment issues, Paper N° 3.1

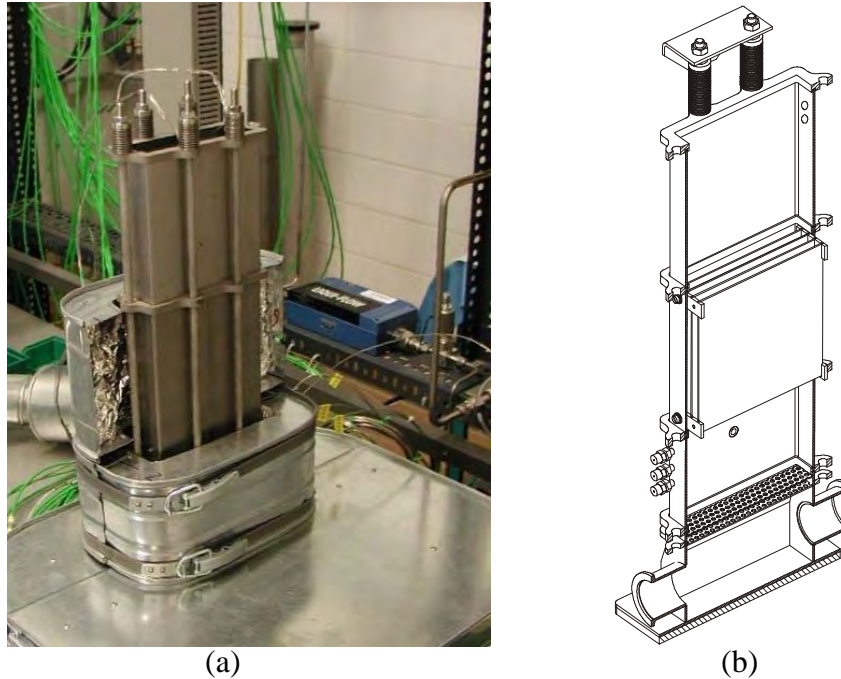


Figure 5: REKO-3 test facility: (a) partially isolated, (b) schematic view

flow direction. A total of 14 sample points has been implemented. The measurement gas is conducted from the sample point through a cooler and condensate trap. Hydrogen and oxygen concentrations are measured in line by separate systems.

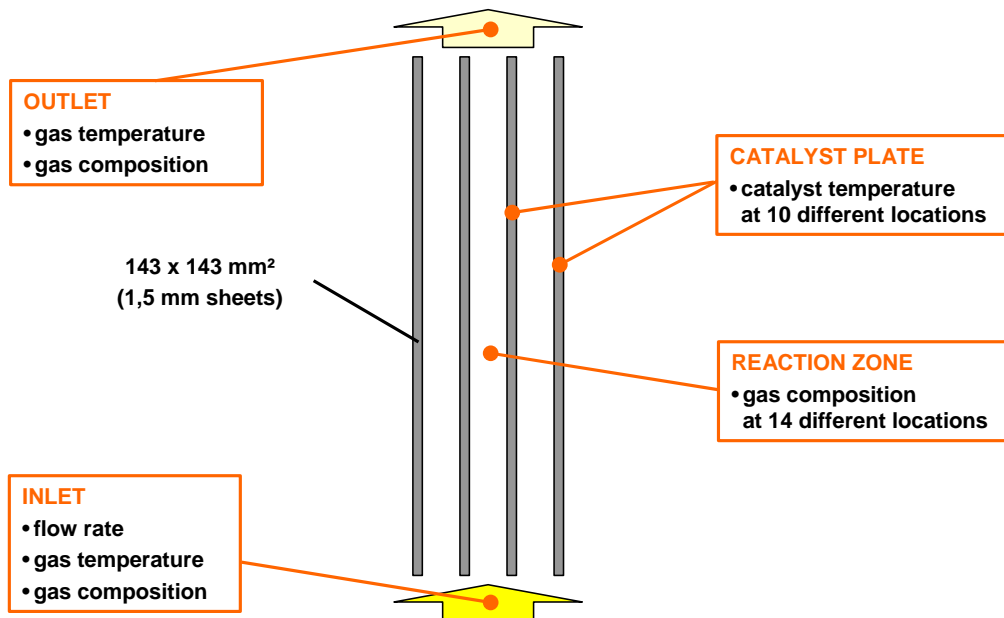


Figure 6: REKO-3 measuring points in a 4-sheet arrangement

Some exemplary measurement results are given in Figure 7. These steady-state distributions of the hydrogen concentration (left side) and the catalyst temperature (right side) were obtained at 2 vol.% and 4 vol.% inlet hydrogen concentration, the inlet gas temperature of 25°C and a flow rate of 0.8 m/s. The symbols represent measuring values while the lines are added for the sake of clarity. The measured values are plotted on the horizontal axis in order to illustrate the vertical arrangement of the catalyst plates.

Session 3: Containment issues, Paper N° 3.1

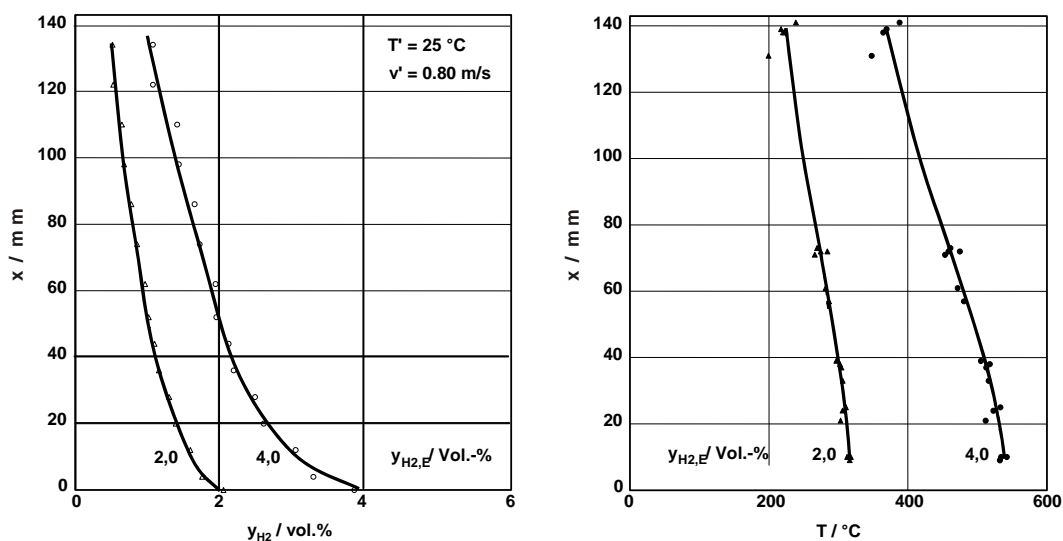


Figure 7: Steady-state hydrogen depletion (left side) and catalyst temperatures (right side) along the catalyst sheets for two different hydrogen inlet concentrations at 25°C inlet temperature and a flow rate of 0.8 m/s

The main part of the conversion reaction takes place in the very first area of the catalyst surface. This is reflected by the first sharp concentration drop as well as by the temperature profile where the maximum temperature is located close to the leading edge of the catalyst sheet ($x = 0$ mm). The temperature rise closer to the trailing edge is to some extent caused by heat conduction and convective heat transport. Maximum temperatures reach the conventional ignition limit of about 560°C already at a hydrogen concentration of 4 vol.% hence representing the risk of an unintended ignition.

In general, the experiments have been performed for flow rates between 0.25 and 1.0 m/s, inlet gas temperatures up to 110°C, steam concentration up to 60 vol.%, and for hydrogen concentrations between 0.5 and 6.0 vol.% (Ref. [8]). Specific attention has been paid to the effect of steam and oxygen starvation on the conversion rate. On-going experiments deal with the ignition of the gas mixture on hot catalyst sheets.

In order to study the operational behaviour of PAR under natural flow conditions and to study the chimney effect on the PAR operation in detail, a new facility (REKO-4) is presently constructed at FZJ. The pressure vessel with an inner diameter of 1.4 m and a height of 4 m is designed for a pressure of 25 bar. A total of 25 flanges will allow the accessibility of the vessel with different measurement techniques. Foreseen measurements inside the vessel are basically pressure, gas temperature and hydrogen/oxygen concentration. The PAR device will be equipped with thermocouples for catalyst and gas temperature measurement and hydrogen/oxygen sensors. Most challenging measurement will be the use of particle image velocimetry (PIV) for the measurement of the flow field at the inlet and the outlet of the PAR. On the one hand, these information are essential in order to determine the PAR throughput and efficiency. On the other hand, this high-resolution measurement technique provides data suited for CFD validation.

In general the experiments will deal with aspects of the operational behaviour of small PAR devices when the flow rate is solely induced by the heated catalyst sheets. For this purpose, a defined amount of hydrogen will be released into the air-filled vessel. After start-up of the exothermal catalytic reaction the buoyancy effect is expected to fuel the PAR device. The hydrogen conversion will be monitored by sensors inside the vessel. Due to the option of positioning the PIV device in different locations it is expected to visualise the flow pattern in different relevant areas around the PAR. First experimental results are expected for late 2008.

C.2 Detailed model REKO-DIREKT

The empirical models described in B are validated for the specific experimental conditions present in the original tests. For detailed studies, however, these correlations do not provide relevant parameters, such as catalyst or box temperature in order to assess e.g. the ignition risk. For this reason, REKO-DIREKT, a 2D detailed mechanistic model based on Fortran 90 was developed at FZJ (Ref. [9]). The principal structure of REKO-DIREKT is given in Figure 8.

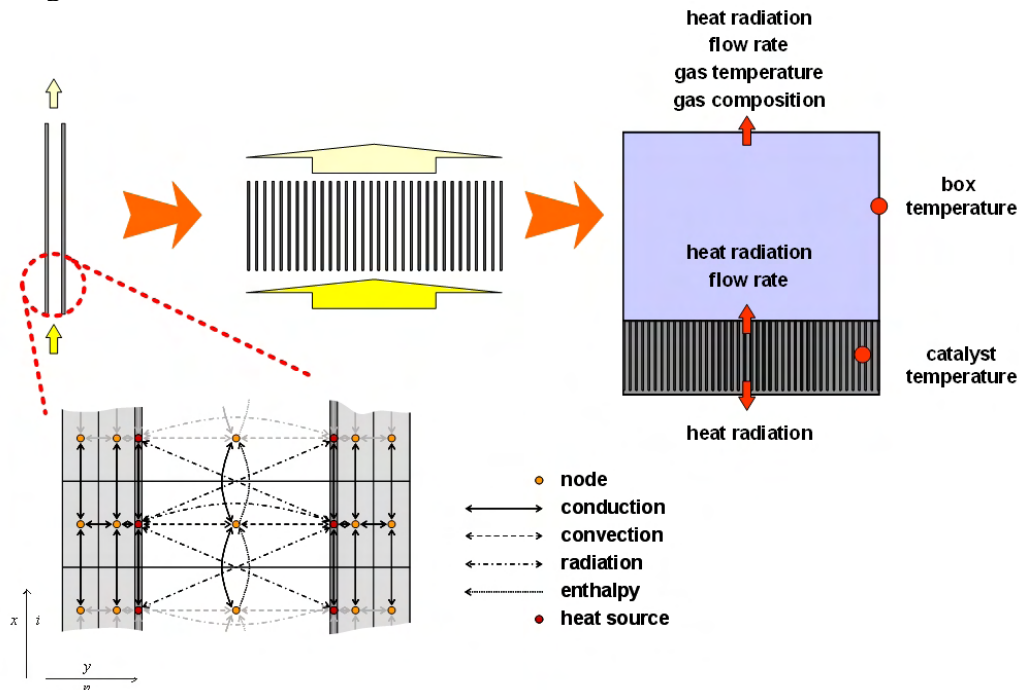


Figure 8: Structure of REKO- DIREKT code

Local reaction rates inside a single channel formed by two catalyst sheets are calculated by means of a mechanistic transport approach

$$\dot{r}_i = Sh \cdot \frac{D_i}{d} \cdot \Delta C_i, \quad i = H_2, O_2$$

where Sh is the local Sherwood number, D_i the effective diffusion coefficient of the species i in the mixture, d the hydraulic diameter and ΔC_i the molar concentration of the species i . This single channel is expanded to a full recombiner catalyst section to calculate in a last step the buoyancy driven flow through the PAR box as well as the radiative and convective heat fluxes from the PAR to its environment. REKO-DIREKT provides the local gas composition as well as the efficiency of the PAR, and the catalyst temperature profile which can be used for further risk and safety assessment.

REKO-DIREKT has been validated against the REKO-3 database (Table 2). Figure 9 shows as an example the temperature profiles and hydrogen concentration decay for different hydrogen inlet concentrations and flow velocities. The code reproduces the temperature distribution and reaction kinetics for the experimental test cases quite well. Natural convection through the PAR box is already implemented in the code and predicts credible

Session 3: Containment issues, Paper N° 3.1

values for the flow velocity, compared to the integral large scale experiments. A validation data base for the chimney model is an expected result from future REKO-4 experiments.

Table 2: Experimental results and model validation matrix

Issue	Experimental data available	Validation performed
Standard reaction kinetics	X	X
Influence of steam	X	X
Lack of oxygen	X	X
Ignition criteria	on-going	

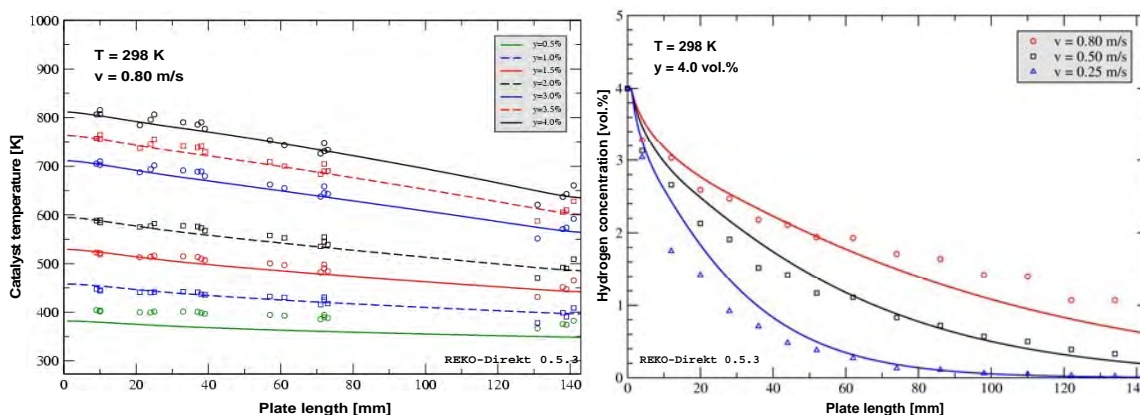
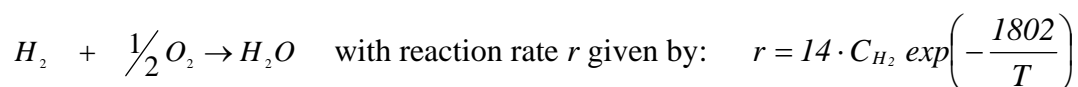


Figure 9: Temperature and concentration profiles: Experimental and REKO-DIREKT results

C.3 CFD modelling with different approaches

As mentioned previously, the main drawback of the empirical approaches, based on experimental correlations, are, in one hand, that PAR environment is neglected and, in the other hand that the validation of the correlation is limited to the field covered by the tests. To overcome these limitations, IRSN and FZJ have developed different CFD approaches to better describe the physical phenomena inside PAR (local flow, heat generation, ...) and the interaction between PAR and the environment.

The first IRSN approach consists in implementing specific User-Fortran routines into CFX 5.0 code. These code extensions deal mainly on catalytic surface reaction modelling. To perform this development, chemical reactions are supposed to be fast regarding species diffusion phenomena and one-step reaction from Ikeda et al in Ref. [10] is applied:



For surface reactions, only cells adjacent to it are allowed to take place. Gas reactions are ignored by a reaction rate of zero in all other fluid cells. Inside the recombiner, laminar flow is assumed. Heat conduction is allowed in the fluid and the reaction sheets. Buoyancy and diffusion of the species are considered.

Based on the model described above, 2D catalytic sheet model and full 3D PAR model including 15 catalytic sheets were developed (see Figure 10).

Session 3: Containment issues, Paper N° 3.1

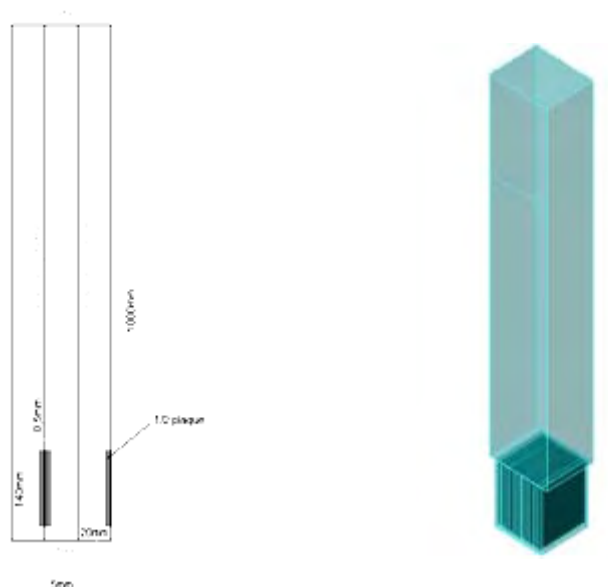


Figure 10: Schemes of the 2D catalytic sheet model (11 849 elements) and of the 3D recombiner model with 15 sheets (40524 elements)

These two models have been validated against the H2PAR experiments. Figure 11 shows the temperature time evolution at two locations (1.8 cm from the sheet bottom and at the PAR outlet).

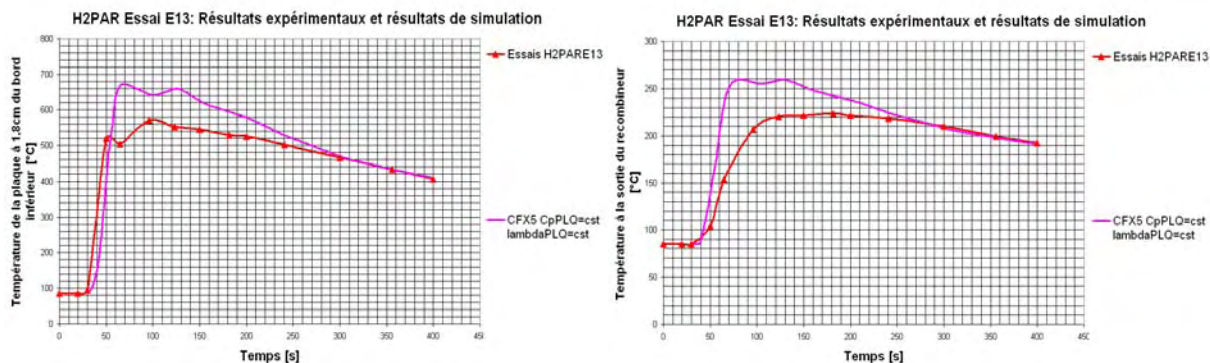


Figure 11: Time evolution of catalytic sheet temperature and gas temperature in the chimney

The code reproduces reasonably well the transient of gas and surface temperature inside the recombiner. Nevertheless, the adopted fast surface reaction hypothesis did not allow the investigation of transport and chemistry interaction which governs phenomena as ignition. For these reasons, IRSN has developed a more detailed CFD model including complex gas and surface chemistry, and multi-component transport. The dedicated SPARK code was developed by coupling CHEMKIN II (Ref. [11]), SURF CHEM (Ref. [12]) and EGLIB (Ref. [13]) libraries following the architecture presented in Figure 12.

Detailed chemical kinetics are then considered for gas chemistry and catalytic surface chemistry including adsorption, surface, desorption reactions on the elementary active sites (Figure 13).

Session 3: Containment issues, Paper N° 3.1

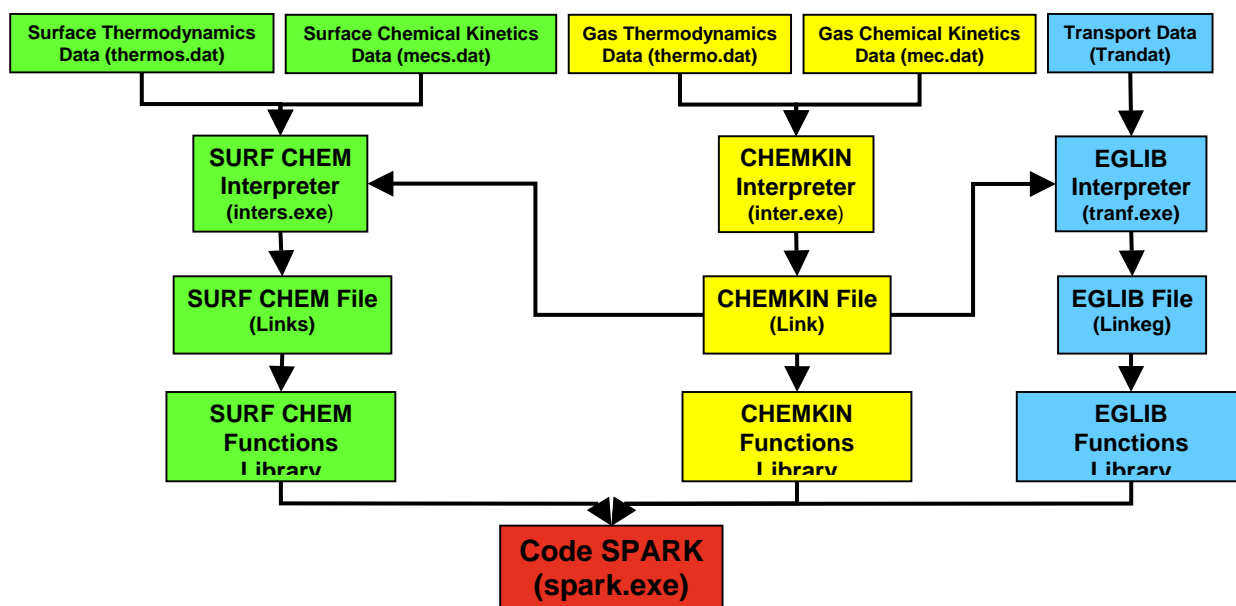


Figure 12: Coupling architecture strategy of SPARK code

H ₂ /O ₂ reactions				A	n	E (cal/mol)
1	H + O ₂ ⇌ O + OH	2.0010 ¹⁴	0.00	16802.10		
2	O + H ₂ ⇌ H + OH	2.0610 ⁰⁴	2.67	6285.85		
3	H ₂ + OH ⇌ H ₂ O + H	1.0010 ⁰⁸	1.60	3298.28		
4	2OH ⇌ O + H ₂ O	1.5010 ⁰⁹	1.14	100.38		
5	H + H + M ⇌ H ₂ + M	1.8010 ¹⁸	-1.00	0.00		
6	2O + M = O ₂ + M	2.9010 ¹⁷	-1.00	0.00		
7	H + OH + M ⇌ H ₂ O + M	2.2010 ²²	-2.00	0.00		
HO ₂ reactions				A	n	E (cal/mol)
8	H + O ₂ + M = HO ₂ + M	2.3010 ¹⁸	-0.80	0.00		
9	HO ₂ + H ⇌ H ₂ + O ₂	2.5010 ¹³	0.00	693.12		
10	HO ₂ + H ⇌ 2OH	1.5010 ¹⁴	0.00	1003.82		
11	HO ₂ + H ⇌ H ₂ O + O	3.0010 ¹³	0.00	1720.84		
12	HO ₂ + O ⇌ OH + O ₂	1.8010 ¹³	0.00	-406.31		
13	HO ₂ + OH ⇌ H ₂ O + O ₂	6.0010 ¹³	0.00	0.00		
H ₂ O ₂ reactions				A	n	E (cal/mol)
14	2HO ₂ ⇌ H ₂ O ₂ + O ₂	2.5010 ¹¹	0.00	-1242.83		
15	2OH + M ⇌ H ₂ O ₂ + M	3.2510 ²²	-2.00	0.00		
16	H ₂ O ₂ + H ⇌ H ₂ O + OH	1.0010 ¹³	0.00	3585.09		
17	H ₂ O ₂ + H ⇌ H ₂ + HO ₂	1.7010 ¹²	0.00	3752.39		
18	H ₂ O ₂ + O ⇌ OH + HO ₂	2.8010 ¹³	0.00	6405.35		
19	H ₂ O ₂ + OH ⇌ H ₂ O + HO ₂	5.4010 ¹²	0.00	1003.82		

Adsorption reactions			s	E
1	H ₂ + 2Pt ^(s) → 2H ^(s)	0.046	-	
2	H + Pt ^(s) → H ^(s)	1.000	-	
3	O ₂ + 2Pt ^(s) → 2O ^(s)	0.070	-	
4	O + Pt ^(s) → O ^(s)	1.000	-	
5	H ₂ O + Pt ^(s) → H ₂ O ^(s)	0.750	-	
6	OH + Pt ^(s) → OH ^(s)	1.000	-	
Surface reactions			A	E (cal/mol)
7	H ^(s) + O ^(s) ⇌ OH ^(s) + Pt ^(s)	3.7010 ²¹	2749	
8	H ^(s) + OH ^(s) ⇌ H ₂ O ^(s) + Pt ^(s)	3.7010 ²¹	4183	
9	2OH ^(s) ⇌ H ₂ O ^(s) + O ^(s)	3.7010 ²¹	11520	
Desorption reactions			A	E (cal/mol)
10	2H ^(s) → H ₂ + 2Pt ^(s)	3.7010 ²¹	16109 -1434 σ _H	
11	2O ^(s) → O ₂ + 2Pt ^(s)	3.7010 ²¹	50956 -14340 σ _O	
12	H ₂ O ^(s) → H ₂ O + Pt ^(s)	1.0010 ¹³	9632	
13	OH ^(s) → OH + Pt ^(s)	1.0010 ¹³	46080	

Figure 13: Gas phase chemical mechanism of Warnatz (Ref. [14]) and Surface chemical mechanism in Deutschmann (Ref. [15])

Session 3: Containment issues, Paper N° 3.1

The previous chemical reactions mechanisms were implemented in SPARK code and validated against several test. Figure 14 shows the good agreement between experiment (Ref. [16]) and simulation for hydrogen concentration, oxygen concentration and surface temperature along the catalytic sheet.

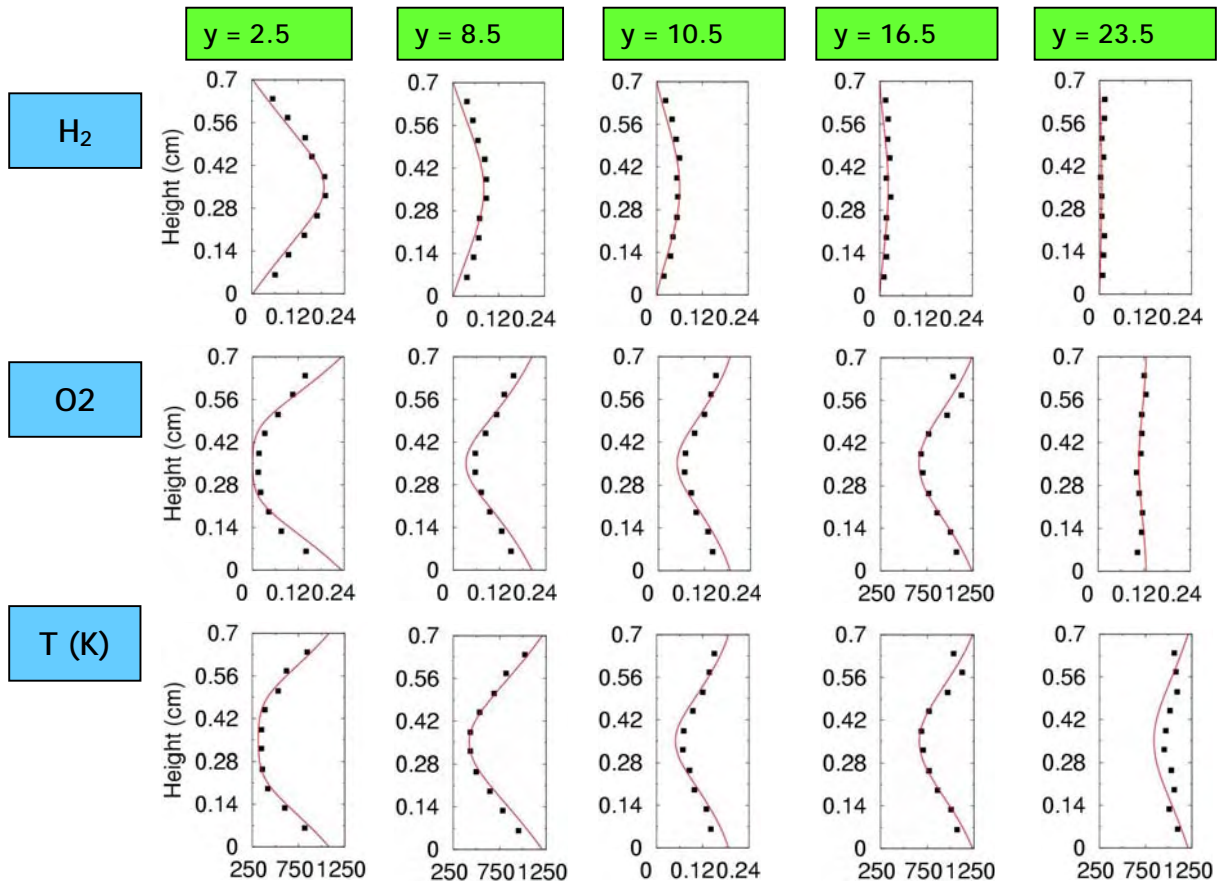
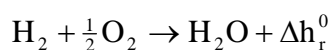


Figure 14: Gas concentration and surface temperature: comparison of experiment and SPARK results

The CFD model implemented by FZJ is based on the elementary understanding that in the present case the rate determining step of the catalytic recombination of hydrogen and oxygen is the transport of the products and educts from and to the surface. For the simulation of the catalytic process only the transport of the species is considered. The outer surface of the catalytic coating is modelled as a wall. The chemical reaction is implemented as a single step reaction



by means of sinks and sources of mass, enthalpy and momentum in the cells adjacent to the wall.

The prediction of the reaction rate is done by fully resolving the species boundary layer, which allows solving the reactants flux by Fickian diffusion only. Consequently, the reaction rate can be described as

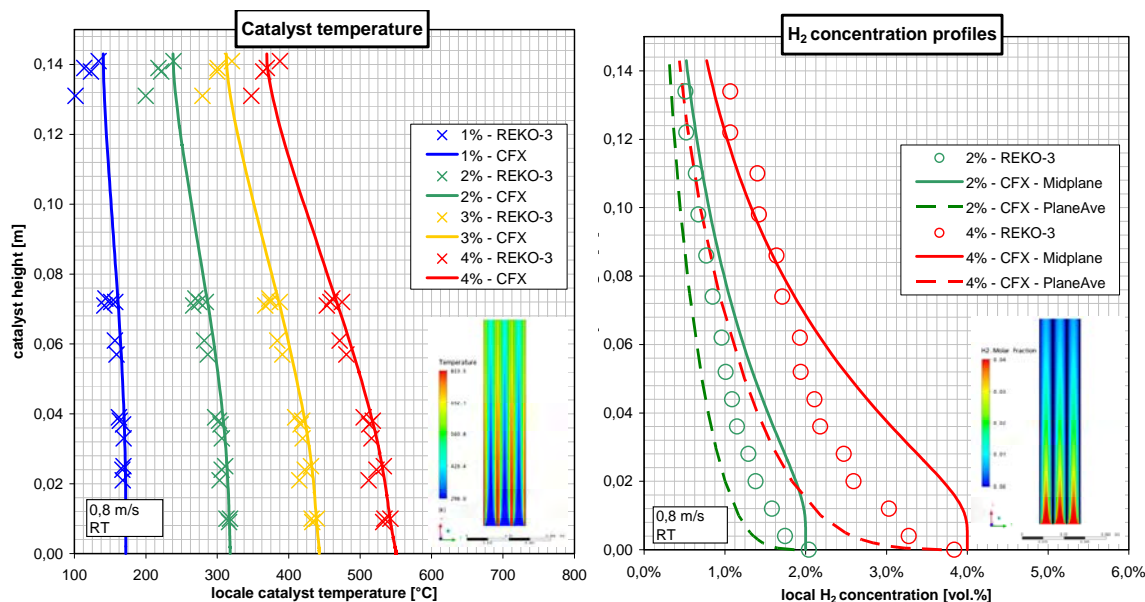
Session 3: Containment issues, Paper N° 3.1

$$\dot{r}_i = \bar{c} \cdot \left(D_i + \frac{v_t}{Sc_t} \right) \frac{\partial X_i}{\partial z} \Big|_w, \quad i = H_2, O_2$$

where c is the molar concentration of the mixture, D_i is the effective diffusion coefficient of the species i in the mixture, v_t/Sc_t is the turbulent diffusion coefficient and X_i the molar fraction of the species i . To enable the model to predict also the reaction rates under oxygen starvation conditions the transport of oxygen is also considered. The system of compressible Navier Stokes equations is closed by the $k-\omega$ based shear stress transport (SST) turbulence model in order to use this low Reynolds approach and integrate up to the wall and avoid the use of wall functions. The i -species are described by the ideal gas equation of state. As buoyancy is the major driving force, the full buoyancy model including the production and dissipation of turbulence is included. The radiative heat transfer between the plates and also with the environment is considered by a Monte Carlo model.

This modelling approach has been validated against the REKO-3 test series for plate type catalyst sheets (AREVA design). Figure 15 shows exemplarily the temperature profiles in the catalyst sheets and the hydrogen concentration decay in the flow channel for different hydrogen inlet concentrations and a typical flow velocity of 0.8 m/s in a PAR.

The catalyst temperature profiles are predicted quite well by the model, except a slight over prediction at the end of the catalyst sheet in some cases due to some uncertainties with regard to the radiative heat exchange with the REKO-3 channel and outlet. Comparing the concentration profiles with the experimental data is difficult as this measurement allows no point or profile measurement, so for validation purpose two extreme cases are considered: The values at the symmetry plane between the plates (solid line) as a quasi point measurement and an arithmetic average over the channel cross section (dashed line). The numerical model predicts the concentration profiles, and with it the entire reaction rate quite well in a reasonable agreement to the experimental results.



**Figure 15: Temperature and concentration profiles:
Experimental and CFD modelling results**

This modelling approach has its advantage in predicting the detailed thermal hydraulic and transport phenomena in a PAR channel and is used for the assessment of new catalyst designs. For the study of an entire PAR operational behaviour it is presently too cost intensive to model all catalyst elements in this degree of detail. For this reason, the coupling of REKO-DIREKT with CFX is presently considered at FZJ.

Session 3: Containment issues, Paper N° 3.1

D. SPECIFIC CHALLENGE: PAR IGNITION MODELLING

Concerning the possibilities of ignition by recombiners, several experiments were conducted on French facilities H2PAR, operated by IRSN, and KALI H2 operated by CEA. The conclusions of these experiments lead to define the following “flammability limits” depending on gas mixture (Ref. [17]):

- √ in the tests in dry air, a generalized ignition is observed between 5.5% and 6.8% of hydrogen molar fraction;
- √ at 9.2% steam initial molar fraction, a generalized ignition was detected for a molar fraction of hydrogen injected of 8.5%;
- √ at 31% steam initial molar fraction, an ignition appears for a hydrogen concentration of about 8.6%;
- √ at 45% steam initial molar fraction, the limit of ignition was given of about 10% in molar fraction of hydrogen;

The results relating to the limit of ignition by the tested recombiner model, obtained during KALI-H2 and H2PAR tests are gathered in Figure 16.

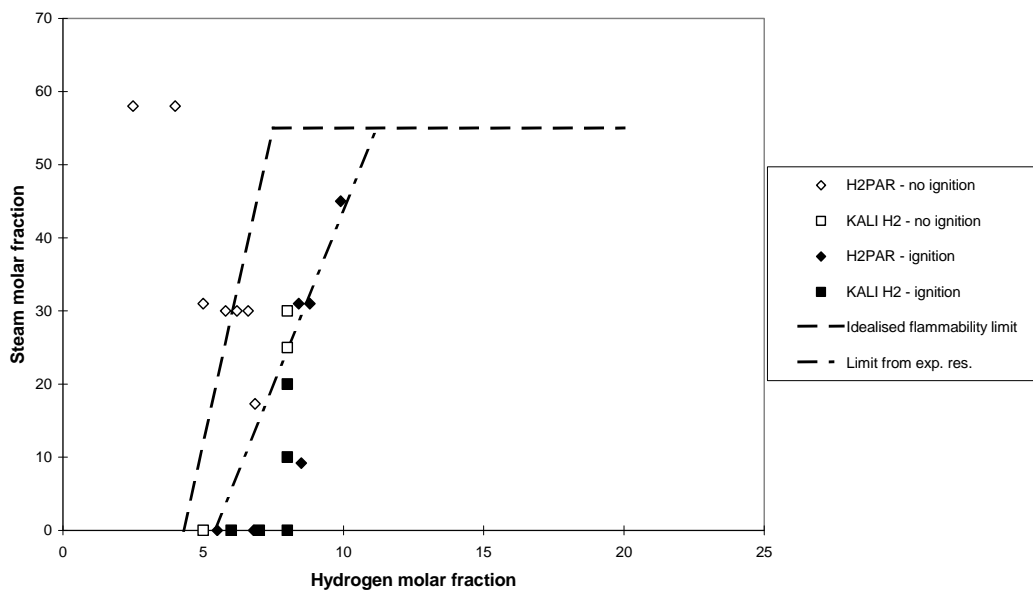


Figure 16: Ignition limit from KALI-H2 and H2PAR tests

The distribution of the experimental points makes it possible to define two zones in the diagram, one for which there is a potential generalised ignition, the other for which there is no generalized ignition. Nevertheless, these results need to be corroborated by well instrumented experiments as REKO-3.

In the course of the experimental matrix of the REKO-3 test programme (Table 1) ignition experiments are performed. The experiments are performed with the standard set-up given in Figures. 5 and 6. The course of a typical experiment is given in Figure 17: The measured temperature values (catalyst temperature, inlet gas temperature) are represented on the left vertical axis, while the inlet hydrogen concentration values can be taken from the right vertical axis. It has to be noted that the horizontal offset between measured concentration and

Session 3: Containment issues, Paper N° 3.1

the value provided by the flow controllers is caused by the delayed flow of the measurement gas through the probe line. In the shown experiment, a hydrogen-air mixture with 7 vol.% hydrogen at a flow rate of 0.25 m/s was present. Immediately after injection of hydrogen into the flow channel (320 s) the catalyst temperature (measured at the leading edge of the catalyst sheet) starts to increase. It can be noted that the thermocouples measuring the inlet gas temperatures are influenced by heat radiation from the catalyst sheets. As the catalyst temperature reaches nearly 600°C (580 s) ignition occurs followed by a downward combustion. As a consequence, the inlet gas temperature increases while the catalyst temperature decreases due to the fact that the continuous flame combustion below the catalyst sheet practically completely burns the hydrogen in the gas mixture. At 620 s the hydrogen supply is stopped and all temperatures decrease to ambient conditions.

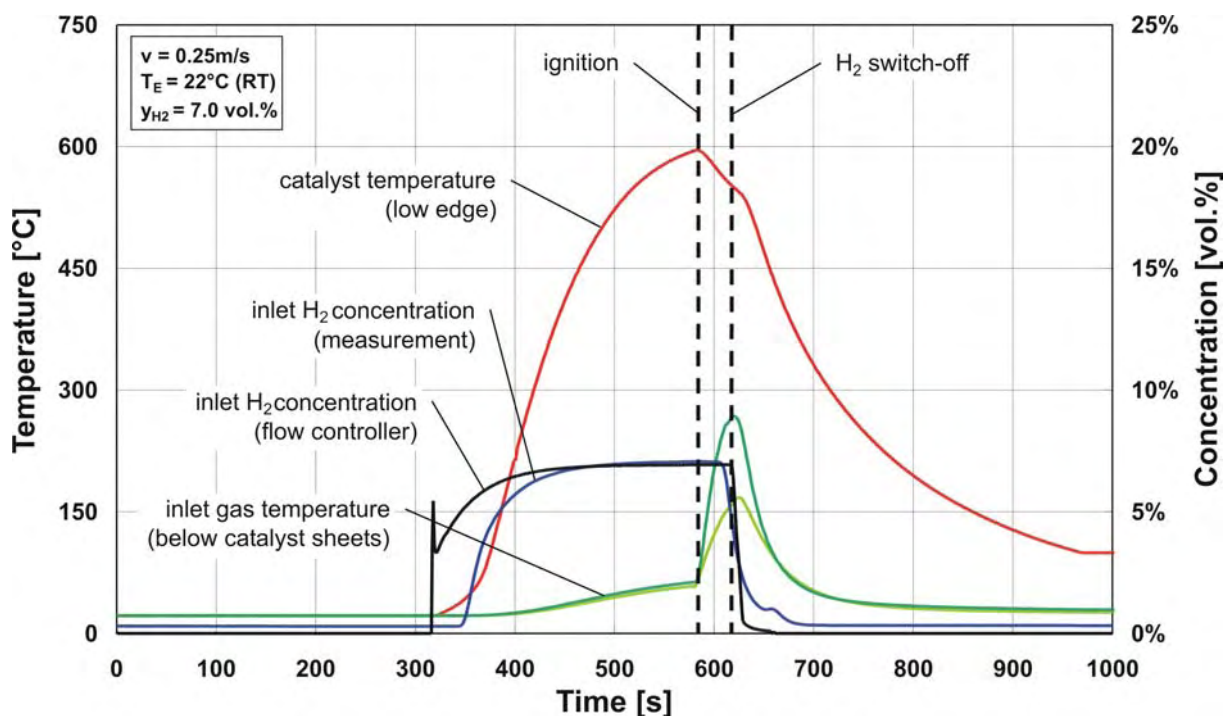


Figure 17: REKO-3 ignition experiment

Detailed modelling adopted in SPARK code by IRSN and presented in C is sufficient to predict ignition. Figure 18 shows the well agreement between calculation and experiments described in Ref. [16]. The location of ignition point is well predicted by the code 9.58 cm against 9.30 cm measured experimentally.

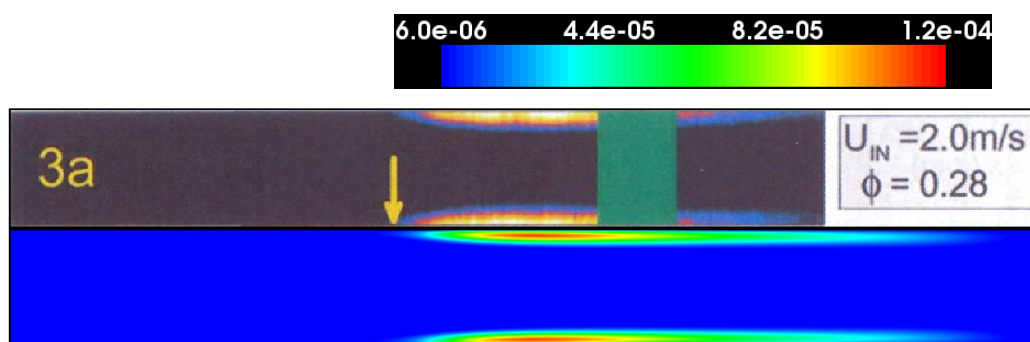


Figure 18: Comparison of experimental and numerical OH molar fraction

Session 3: Containment issues, Paper N° 3.1**E. CONCLUSIONS**

In the past, several experimental programmes were conducted and numerous models were developed to investigate the PAR behaviour in SA situations and to optimise the PARs location inside the reactor containment. However, most of these models are simple and based on empirical correlations issued from experiments. These correlations neglect the influence of the PARs environment and their validation is limited to the field covered by the experiments which do not include phenomena like gas mixture ignition induced by PARs, oxygen starvation, or PAR-environment interaction. To overcome these limitations, analytical and experimental works have recently been performed at FZJ and IRSN in order to enhance the understanding of the processes inside recombiners.

Complementing the existing integral database, FZJ has performed separate effect tests in the REKO-3 facility under well defined conditions in order to investigate the reaction kinetics on the catalyst elements of a PAR. Based on these experimental results a detailed model REKO-DIREKT has been developed describing the PAR operational behaviour. In order to assess the PAR-atmosphere interaction, two approaches are investigated: development of a detailed CFD PAR model and the implementation of the mechanistic model REKO-DIREKT in the CFX code by means of User-Fortran routines.

IRSN has developed two CFD approaches to describe the physical phenomena inside PAR (local flow, heat generation, ...) and the interaction between PAR and the environment. The first approach consists in implementing specific User-Fortran routines into CFX 5.0 code mainly devoted to catalytic surface reaction modelling. This model was validated against H2PAR test. Nevertheless, the adopted fast surface reaction hypothesis did not allow the investigation of transport and chemistry interaction which governs phenomena as ignition. For these reasons, IRSN has developed a more detailed CFD model including complex gas and surface chemistry, and multi-component transport. The dedicated SPARK code was developed by coupling CHEMKIN II, SURF CHEM and EGLIB libraries and validated against dedicated experiments.

In the frame of SARNET, several work has been done to improve the numerical modelling capacities. A first series of common recombiner interaction studies (PARIS benchmarks) has been performed affirming the need of detailed investigation of PAR operational behaviour. PARs location effect, gas mixture ignition induced by PAR and oxygen starvation are still open issues. With increasing time after installation of PARs in European LWRs (~ 10 years) the poisoning effects during normal NPP operation will play a more important role in the PAR maintenance. Detailed chemical models may be needed for dealing with these phenomena.

Hydrogen has been identified as high priority topic for further activities in the frame of SARNET-2. The activities in the field of PAR modelling will be continued with the goal of developing joint modelling approaches leading to a common approach for the ASTEC code.

CFD modelling will be jointly continued and improved in benchmark exercises. For this purpose, experimental data will be needed. On the one hand, a new test vessel REKO-4 at FZJ will be dedicated to the assessment of PAR behaviour under natural convection conditions. On the other hand, co-operations with on-going OECD projects (THAI, SETH II) could provide needed benchmark data.

Session 3: Containment issues, Paper N° 3.1

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