OUTCOMES OF THE EXPERIMENTAL AND NUMERICAL WORK ON THE OPERATIONAL BEHAVIOR OF PASSIVE AUTOCATALYTIC RECOMBINERS IN THE LATE PHASE OF A SEVERE ACCIDENT IN THE FRAMEWORK OF THE AMHYCO PROJECT

Ernst-Arndt Reinecke*
Forschungszentrum Jülich GmbH
52425 Jülich, Germany
e.reinecke@fz-juelich.de

Gabriela Nobrega, Ahmed Bentaib
Institut de Radioprotection et de Sûreté Nucléaire
92262 Fontenay-aux-Roses, France
ahmed.bentaib@irsn.fr

Araceli Dominguez-Bugarin, Gonzalo Jiménez
Universidad Politécnica de Madrid
28040 Madrid, Spain
araceli.dominguez@upm.es, gonzalo.jimenez@upm.es

Matthias Braun
Framatome GmbH
91052 Erlangen, Germany
matthias.braun@framatome.com

Stephan Struth, Claudia Druska, Michael Klauck
Forschungszentrum Juelich GmbH
52425 Jülich, Germany
s.struth@fz-juelich.de, c.druska@fz-juelich.de, m.klauck@fz-juelich.de

ABSTRACT

The molten core-concrete interaction in the ex-vessel phase of a severe accident in a light water reactor is a source of significant amounts of gaseous products including carbon monoxide. The European project AMHYCO addresses open issues related to the understanding of the impact of carbon monoxide on safety-relevant accident phenomena and on hydrogen mitigation measures. In order to support the simulation of accident sequences and the assessment of safety measures, Task 3.2 aimed to provide realistic models considering the influence of carbon monoxide on the operating behavior of passive auto-catalytic recombiners. The work program included conducting new experiments, analyzing existing experimental data, and further developing numerical models.

The experimental program performed within Task 3.2 involved the REKO facilities at FZJ to study the impact of predicted accident atmospheres including oxygen-lean mixtures and the presence of carbon

* corresponding author

monoxide on the hydrogen recombination efficiency of different generic catalysts. More specifically, criteria were derived to predict the atmospheric conditions leading to catalyst poisoning. At the same time, a unified database including selected datasets from the various OECD/NEA-THAI projects was developed. The available experimental data were used to further advance numerical models such as the manufacturer's engineering correlation for Framatome PARs as well as the scientific models SPARK (IRSN), PARUPM (UPM), and REKO-DIREKT (FZJ).

KEYWORDS
Severe Accident, Hydrogen Mitigation, Passive Auto-catalytic Recombiners, Carbon Monoxide

1. INTRODUCTION

The European AMHYCO project [1] aims at revising Severe Accident Management Guidelines, with focus on mitigating combustible gases in the late phase of a severe accident. In this late accident phase, the molten corium-concrete interaction (MCCI) is producing significant amounts of carbon monoxide, in combination to a further hydrogen release. The project addresses open issues related to understanding the impact of the presence of carbon monoxide on the hydrogen combustion risk and required mitigation strategies. The corresponding research activities are performed in WP3 of the project's work plan (see Fig. 1). Following the conclusions of the literature review created in WP1 and the bounding accident scenarios selected in WP2, the experimental program allows for a comprehensive validation of models implemented in relevant computational tools, which are used for scenario simulations in WP4.

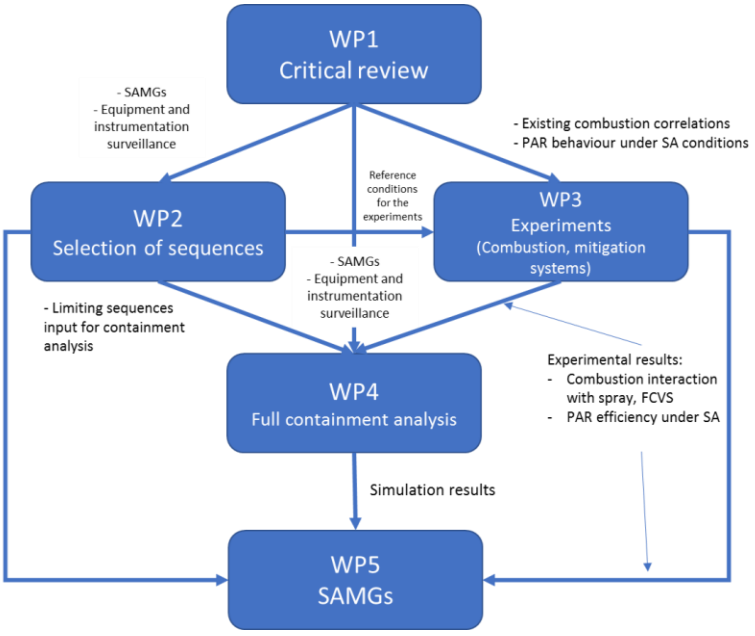

Figure 1. AMHYCO Project: Interaction of technical work packages.

Task 3.2 of WP3 has the specific objective to provide suitable numerical models describing the operation of passive auto-catalytic recombiners (PARs) in the late phase of a severe accident. These models have

been validated against both, existing experimental data from other programs and new targeted experiments performed in the framework of AMHYCO. PARs are the key hardware for mitigating the hydrogen risk in many European nuclear power plants. The operational behavior of PARs has been studied in various national and international projects in the past, starting with early test programs conducted primarily by manufacturers, such as Siemens/Germany (now Framatome) and AECL/Canada [2]. Although some of the aforementioned projects were already addressing the effect of ex-vessel phenomena, such as the presence of carbon monoxide and nuclear aerosols in the containment atmosphere, a first initiative to enhance PAR models towards the conditions in the late accident phase had been taken within the PAR model development exercise in the framework of the SAMHYCO-NET project [3]. In the framework of AMHYCO, the further development of the numerical models is based on a broader experimental database with a specific focus on severe accident sequences with high combustion risk.

2. STATE OF KNOWLEDGE AT THE START OF THE TASK

Previous research on the effect of carbon monoxide on PAR operation has revealed a complex interaction (see [4-6]), which includes regimes of unaffected/parallel or competing hydrogen-oxygen and carbon monoxide-oxygen reaction, as well as conditions where catalyst deactivation due to catalyst poisoning by carbon monoxide can be observed.

The experiments in AMHYCO Task 3.2 focus on the effect of carbon monoxide on the operation of PARs under oxygen-lean conditions. In the context of PAR operation, “oxygen-lean” means that there is not enough oxygen available to the catalysts to allow parallel/unaffected conversion of hydrogen and carbon monoxide. In extreme cases, lean conditions can lead to a so-called poisoning of the catalyst, which is caused by persistent adsorption of carbon monoxide on the catalyst surface, thus, preventing adsorption of oxygen in a sufficient amount. Specifically, the exact conditions to distinguish between the above-mentioned regimes, which are expected to depend on gas composition, temperature and pressure, need to be further clarified in detail.

The analyses of the results of the accident scenario simulations performed by WP2 [7] led to the following conclusions with regard to the experimental program:

- Almost through the entire ex-vessel phase, the atmosphere is oxygen-lean with regard to PAR operation. Consequently, the experimental program and model development needs to focus on such conditions.
- PAR models should be validated for atmospheres with temperatures between 40 °C and 140 °C, pressure up to 3.7 bar and steam at saturation.
- Hydrogen fractions are typically higher than or equal to carbon monoxide fractions.
- Potential for PAR ignition should be considered in the models with corresponding criteria, if no catalyst temperature is calculated.

3. EXPERIMENTAL PROGRAM

The experimental program in Task 3.2 involves three facilities located at Forschungszentrum Jülich GmbH (FZJ), Germany. In REKO-1, scoping tests with small catalyst sheets were performed to identify the relevant process parameters. Experimental data in full catalyst scale were obtained in REKO-3, to support project partners’ model validation. Finally, the effect of pressure on catalyst poisoning was studied in REKO-4.

3.1. Scoping Tests in the REKO-1 Facility

The REKO-1 facility is a modular flow reactor which is composed of tube elements according to the test specifications and requirements (Fig. 2). The facility allows the investigation of the recombination behavior of small catalyst specimen (here: 5 x 5 cm²) under well-defined forced flow boundary conditions.

Figure 2. REKO-1: Cylindrical flow reactor (left); catalyst temperature measurement (right) [8].

Scoping tests were performed to understand the impact of parameters such as gas and catalyst temperatures, gas composition, and flow velocity on the catalyst deactivation process for both platinum- and palladium-based catalysts. Due to the small size of the catalyst samples these experiments can be carried out very efficiently in a comparatively short time, thus allowing for a large number of measurements. At the same time, the results are rather qualitative in nature, since geometry effects and heat radiation losses are not representatively mapped.

Based on these scoping tests, the following conclusions are reached:

- Platinum- and palladium-based catalysts behave differently with regard to catalyst deactivation by carbon monoxide.
- The relevant gas species for catalyst poisoning are oxygen and carbon monoxide. The amount of hydrogen in the gas mixture seems to be of secondary importance.
- The effect of the gas temperature on the catalyst deactivation needs to be considered, especially for the palladium-based catalyst.
- Catalyst deactivation needs to be studied under humid conditions as different results may be obtained compared to dry conditions.
- The flow velocity of the gas mixture has only minor effect on catalyst deactivation.

3.2. Catalyst Section Tests in the REKO-3 Facility

Using the identical peripheral set-up as REKO-1, the REKO-3 facility (Fig. 3) is equipped with a vertical modular flow tube reactor with rectangular cross section, which is composed of different elements according to the test specifications and requirements. The setup allows investigation of the recombination behavior of catalyst sheets under well-defined and steady-state forced flow boundary conditions. For the present study, four parallel catalyst sheets are exposed to stationary flows of gaseous mixtures including hydrogen, oxygen, nitrogen, steam and carbon monoxide inside a vertical flow reactor.

The tests provide steady-state data on full-scale catalyst sheets suitable for model development and validation. The resulting database has been used by the Task 3.2 partners to further advance their numerical PAR models.

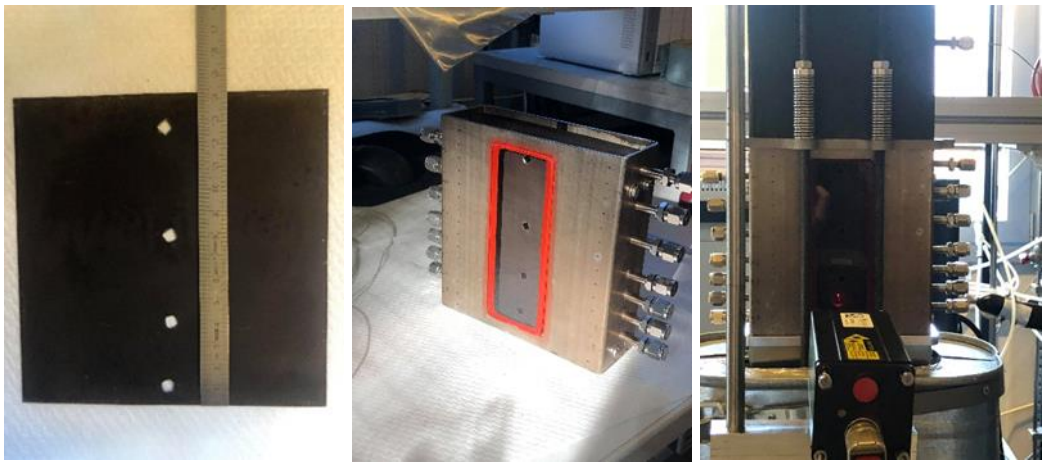

Figure 3. REKO-3: Optical measurement of the catalyst temperature [8].

Despite the complex surface processes, the experimental data allow to derive empirical criteria for catalyst poisoning with sufficient (if necessary conservative) precision, which are based on two easy to obtain parameters, i.e. the gas temperature and the oxygen concentration.

3.3. Natural Convection Tests in the REKO-4 Facility

In contrast to the previous forced-flow facilities REKO-1 and REKO-3, the REKO-4 facility (Fig. 4) simulates the geometry of an entire plate-carrier PAR, and thus operates under natural flow. The model PAR is installed inside a cylindrical steel pressure vessel, allowing to expose the PAR to a controlled atmosphere at ambient and elevated pressures.

A typical test sequence is shown in Fig. 5. The test R4-B04 starts with a pre-reduced oxygen concentration of 6.2 vol.% (black line: O2R 4.01) at a gas temperature of approx. 40 °C (green line: TR 4.16). Immediately after the hydrogen injection starts (blue line: hydrogen fraction KR-4-20), the recombination starts on the catalyst surface as indicated by the increase of the catalyst temperature (red line: TR 4-12) and the temperature inside the PAR chimney (orange line: TR 4-13). At the same time, the carbon monoxide injection starts (not represented in the graph). Due to the catalytic recombination of hydrogen and carbon-monoxide, the oxygen concentration decreases, also leading to a decrease of the catalyst temperature (as due to the oxygen-lean condition, the availability of oxygen is limiting the PAR performance).

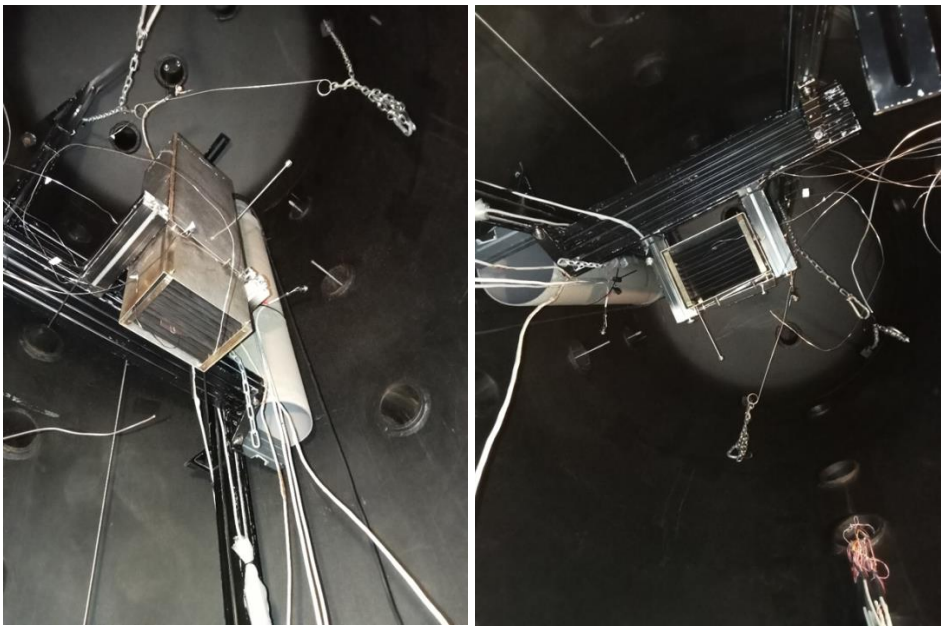

Figure 4. REKO-4: Mounting of the PAR inside the pressure vessel [8].

At approx. 3150 s (marked by the blue circle), the gradient of the catalyst temperature decrease changes rapidly, indicating an abrupt stop of the catalytic reaction. The oxygen fraction measured at the point of change of the temperature gradient is considered the poisoning concentration.

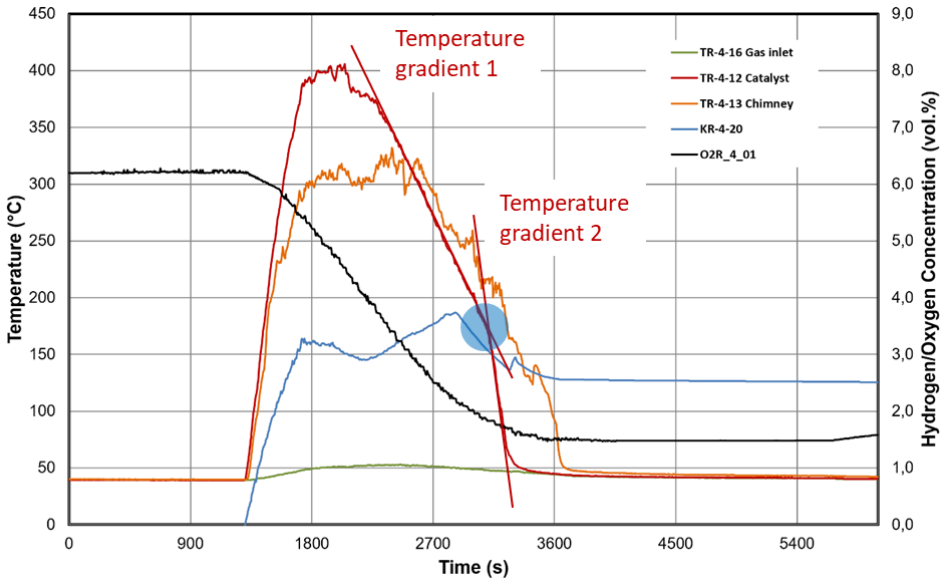

Figure 5. REKO-4 test R4-B04 as example for a typical test sequence [8].

The oxygen concentrations and the corresponding gas temperature where poisoning has been observed in the REKO-4 tests were added to the ranges of partial and full poisoning obtained from the REKO-3 experiments. For both catalysts (platinum and palladium), the results show no significant difference

between experiments performed at 1 bar and 2 bar absolute pressure. For the platinum-based catalyst, no temperature effect was observed between 20 °C and 80 °C, which corresponds with the previous findings of only minor impact of the gas temperature on the poisoning process. The obtained data for the palladium-based catalyst confirm quite accurately the significant temperature effect observed before.

3.4. Conclusions of the Test Program

Overall, the experimental results have shown a non-trivial relationship of the amount of carbon monoxide and the impact on the PAR performance. Due to the complex interaction of catalytic surface processes, mass transfer, and the catalyst and gas temperature, a precise interpretation of the experiments against the needs of manageable model complexity codes was difficult.

With due consideration of the required accuracy of full containment analysis with system codes, and considering as well the measurement uncertainties, the following empirical correlations are proposed to describe the effect of catalyst deactivation (poisoning) by carbon monoxide in the temperature range of 20 °C to 150 °C:

- For platinum-based catalysts, poisoning may occur if the oxygen fraction decreases below a value of 2.0 vol.% (conservative value: 2.5 vol.%). Poisoning occurs quite instantaneously. Partial poisoning has been observed only for a few cases.
- For palladium-based catalysts, poisoning occurs if the oxygen fraction decreases below a value which depends significantly on the gas temperature. Different approaches to derive an empirical function according to the data could be considered, for example
  $y_{O2,poison} = 2.82 - 0.016 \cdot T$ for T = 20 °C to 115 °C
  $y_{O2,poison} = 4.35 - 0.029 \cdot T$ for T = 115 °C to 150 °C
  Above 150 °C, no poisoning effects have been observed for the palladium-based catalyst.

The poisoning effects observed were independent of the pressure and didn't depend significantly on the hydrogen or carbon monoxide concentration. However, more sophisticated empirical models might consider even these effects.

Note that generic non-branded catalytic sheets were used in the experiments employing only pure catalytic metals. Catalytic materials employed in nuclear power plants may have a more complex composition which may reduce the sensitivity to carbon monoxide. Therefore, the results presented in this report can be seen as conservative enveloping.

4. NUMERICAL MODEL DEVELOPMENT

The experimental program of Task 3.2 allows further advancement and validation of numerical models simulating PAR operation as a function of the atmospheric boundary conditions. These models are:

- The empirical Framatome engineering correlation, implemented in most of the relevant codes used for accident simulation [9]
- PARUPM (UPM), a mechanistic model involving detailed surface chemistry [10]
- REKO-DIREKT (FZJ), a mechanistic model based on a mass transfer approach [11]
- SPARK (IRSN), a 3D model involving full surface and gas phase chemistry as well as multi-species CFD [12]

For model development and validation, two databases were developed, the REKO-3 database and the THAI database. Latter was generated on the basis of available data from the OECD/NEA-THAI projects [13]. The generation of a database of quasi-steady state measurement points derived from the transient

THAI experiments allows an overview of the PAR model performance for the entire experimental program rather than only comparing single transient experiments with model calculations. It is suited for both correlation model and detailed PAR models.

4.1. Framatome Correlation

The new generated THAI database has been applied to confirm existing limitations of the empirical Framatome engineering correlation in modelling PAR operation under oxygen-lean conditions. This was already identified for a single case in the framework of the SAMHYCO-NET project, the precursor to the AMHYCO project. Improvements of the correlation based on the laws of diffusion-controlled reaction kinetics have been implemented, which allow for a more consistent transition from oxygen-rich to oxygen-lean PAR operation.

Fig. 6 shows the performance of the improved engineering correlation in reproducing the PAR recombination rates observed in the THAI HR-12 experiment. The switch between oxygen-rich and oxygen-lean conditions as well as the recombination rate during oxygen starvation meets the experimental data better than before.

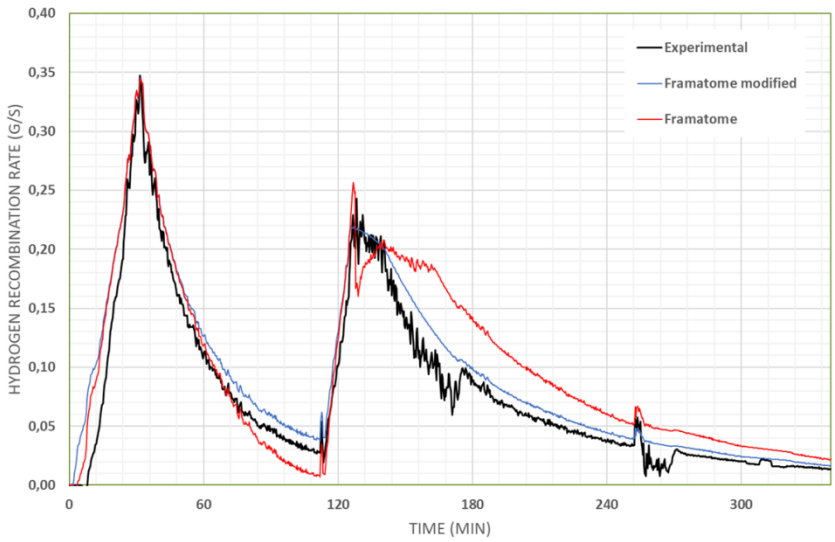

Figure 6. Original and improved Framatome correlation versus THAI HR-12 data.

The comparison of observed hydrogen recombination rates and predicted recombination rates over the entire THAI database (Fig. 7) demonstrates the successful improvement of the engineering correlation as well as the suitability of the database for model validation.

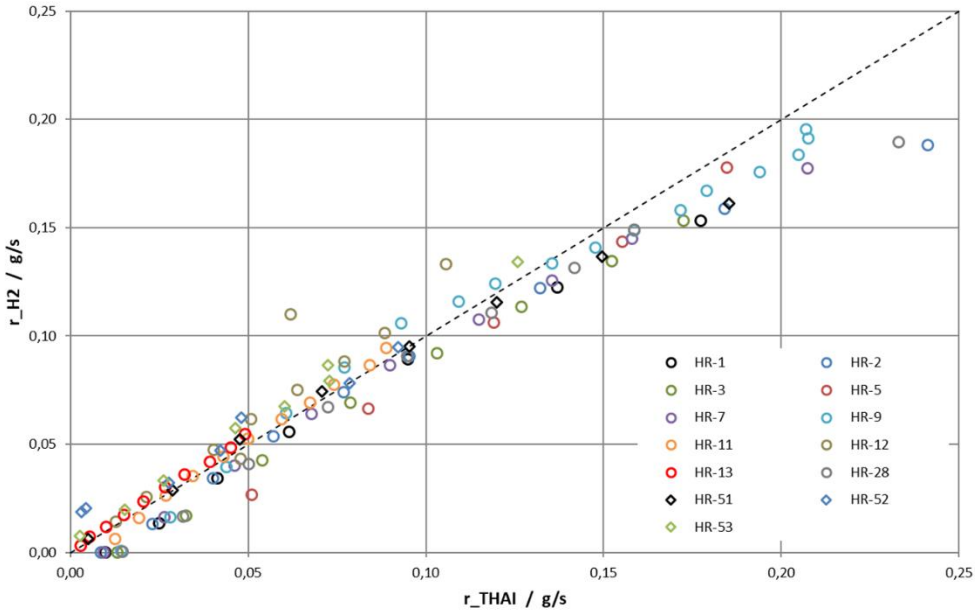

Figure 7. Comparison of the improved Framatome correlation rates with the THAI database.

The corresponding validation data for CO recombination rates, which involve new experiment from the OECD/NEA THEMIS project, cannot be shown in the present document due to contractual reasons. The new proposed engineering correlation is currently under internal review process.

4.2.2. PARUPM

Through the implementation of the linear system of differential equations that include the simplified Deutschmann mechanism, PARUPM is capable of simulating the relevant phenomena associated with the surface reactions of hydrogen and carbon monoxide on plane-parallel catalyst sheets. In the framework of Task 3.2, the PARUPM code has been further developed and validated. The most relevant enhancements include the implementation of a chimney model as well as a diffusion-based reaction model.

The transient model can approximate the PAR heating phase and its cooling down, as well as the transient changes in the boundary conditions. Fig. 8 shows the recombination rate obtained during the OECD-THAI experiment HR-1 as well as the recombination rates predicted by PARUPM, using the boundary conditions from the experiment. Apart from a moderate overprediction of the recombination rate in the first injection phase of the experiment, the only significant discrepancy between the model and the experimental recombination rates occurs at the end of the second injection phase, which is due to a PAR-induced ignition of the local vessel atmosphere causing high recombination rates during the combustion process. PARUPM doesn't contain an ignition module, thus this behavior is not analyzed by the code. Overall, the comparison with experimental data confirms that the code can simulate the behavior of a real passive autocatalytic recombiner under realistic conditions.

The validation results obtained demonstrate that the physico-chemical strategy used in the PARUPM model can simulate the behavior of the recombiner under a variety of conditions, from typical concentrations and settings to more extreme ones such as oxygen starvation, catalyst poisoning, or elevated inlet temperatures. Furthermore, the code can reproduce the behavior of a PAR in transient conditions, where pressure, temperature, and gas concentrations are constantly changing at the recombiner inlet.

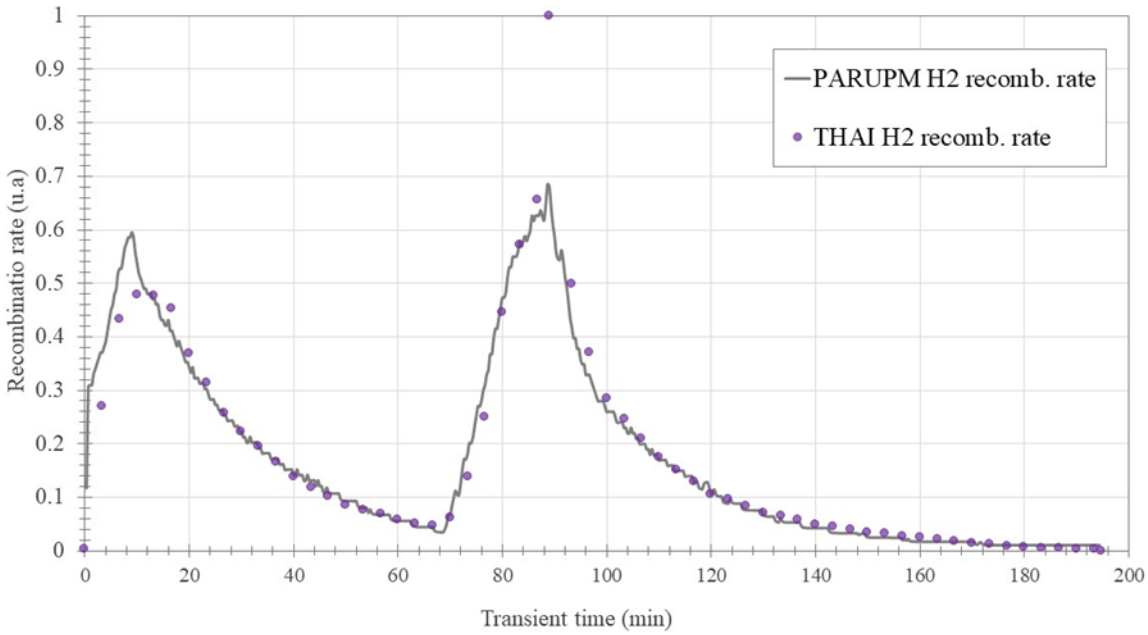

Figure 8. PARUPM: Recombination rate with PARUPM (grey line) and experimental recombination rates (purple dots) during the transient (HR-1).

More details of the PARUPM enhancement and validation have been published in [14, 15]. The PARUPM code is planned to be implemented in 3D thermo-hydraulic analysis codes, such as GOTHIC, as it was implemented in the past in lumped parameters codes like MELCOR.

4.2.3. SPARK

The further development of the SPARK code includes an update of the surface reaction mechanisms for platinum-based catalysts as well as an approach to implement a surface mechanism for palladium-based catalysts. The new Pt mechanism allows PAR simulation during deactivation conditions in the presence of carbon monoxide. The enhanced SPARK code was used for post-calculations of both REKO-1 and REKO-3 data to verify that the SPARK code is able to reproduce the experimental results.

Fig. 9 shows an example of a SPARK simulation of poisoning on a platinum catalyst for a gas mixture containing 4 vol.% hydrogen and 2 vol.% carbon monoxide. Fig. 9 (left) shows the hydrogen distribution inside the gas channel between two catalyst sheets for three different fractions of oxygen during the deactivation process. On the left, 2.6 vol.% oxygen represents the beginning of poisoning. For 2.5 vol.% oxygen, the active zone of the catalyst is moving towards the upper edge of the catalyst (middle). On the right, the catalyst is completely deactivated at 2.3 vol.% oxygen. The same effect can be observed for the carbon monoxide fraction between the catalyst sheets on the right side of Fig. 9.

The improvements and validation work performed in the framework of AMHYCO was published in [16].

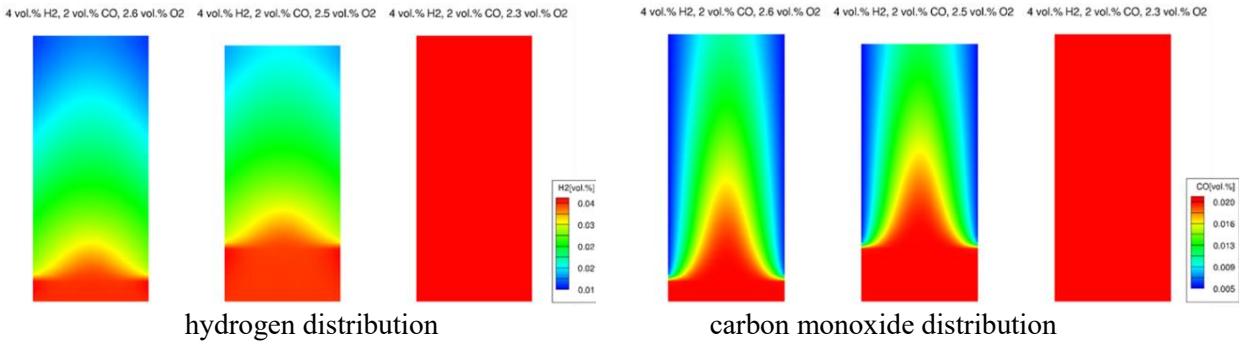

Figure 9. SPARK: Hydrogen (left) and carbon monoxide (right) distribution in the channel between two catalyst sheets for 2.6 vol.% O_2 (left), 2.5 vol.% O_2 (middle) and 2.3 vol.% O_2 (right)

4.2.4. REKO-DIREKT

Unlike in numerical codes with a detailed surface kinetics model, such as PARUPM and SPARK, the catalytic reaction inside the PAR is calculated in REKO-DIREKT with a mass transfer approach. The catalyst poisoning is represented with the empirical correlations described in Section 3.4. For this purpose, the selection of platinum- or palladium-based catalyst has been implemented in the user input.

Fig. 10 shows the predicted versus the experimentally observed hydrogen outlet concentrations and Fig. 11 the catalyst temperatures over the THAI database. The calculated hydrogen outlet concentrations agree with the experimental data very well over the entire range (Fig. 10). A specific feature of REKO-DIREKT is the calculation of the temperature profile along the catalyst sheets. A comparison of calculated and measured values is shown in Fig. 11. Again, the agreement is very good, although the calculated temperature values are systematically too high.

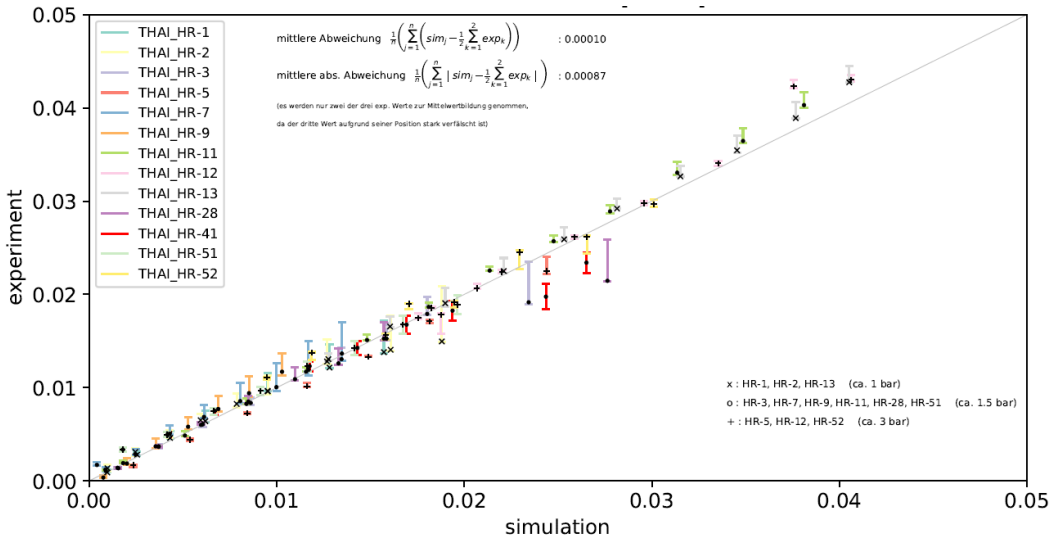

Figure 10. REKO-DIREKT: Calculated outlet hydrogen concentration compared with THAI data

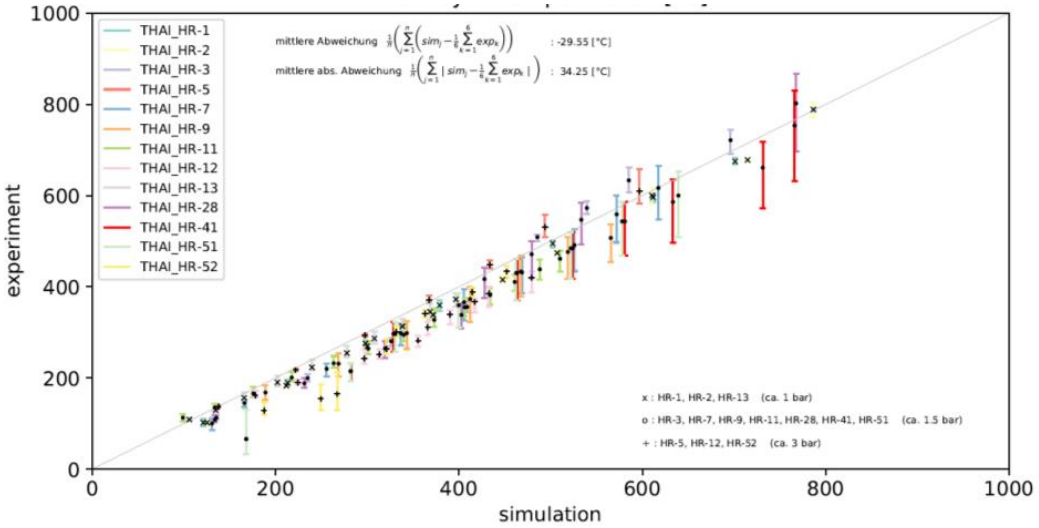

Figure 11. REKO-DIREKT: Calculated catalyst temperatures compared with THAI data

5. CONCLUSIONS

In the framework of the AMHYCO project, Task 3.2 aimed at providing realistic PAR simulations under the conditions of the late phase of a severe accident. The experimental program focused on a better understanding of the impact of carbon monoxide on the PAR performance to provide data for further developing the different numerical codes. Specifically, the effect of oxygen starvation is considered to be well understood, and the results allow to derive empirical criteria for catalyst poisoning with sufficient precision, which are based on easy to obtain parameters (gas inlet temperature and oxygen concentration) even for those PAR models that don't involve surface chemistry.

In addition to the REKO-3 database, which is especially suited for model development and validation for the PAR geometry independent mechanistic models, a second database of quasi-steady state measurement points was derived from the transient experiments performed in the framework of the OECD/NEA-THAI projects. This new database allows validation of the PAR model performance for the boundary conditions of the entire experimental program rather than only comparing single transient experiments with model calculations. It is suited for both correlation model and mechanistic PAR models.

Making use of both experimental databases, REKO and THAI, the PAR models involved (Framatome engineering correlation, PARUPM, REKO-DIREKT and SPARK) were significantly enhanced and prepared for further use in the simulation tools for accident scenarios in the further course of AMHYCO.

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