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EXPERIMENTAL INVESTIGATION OF PAR DEVICE FOR HYDROGEN RECOMBINATION IN REKO-4 FACILITY

Mahmut Cüneyt KAHRAMAN* and Paul-Martin STEFFEN**
*Department of Energy Systems Engineering, Yalova University,
77200 Yalova, Turkey, mcuneyt.kahraman@yalova.edu.tr, ORCID: 0000-0002-1535-3832
**Institute of Energy and Climate Research (IEK-6), Forschungszentrum Jülich GmbH,
52425 Jülich, Germany, paul-martin.steffen@rwth-aachen.de, ORCID: 0000-0001-7583-1800

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Abstract: The Fukushima accident has proved that passive safety systems in reactors are necessary to avoid hydrogen accumulation. One of these systems are PAR (passive autocatalytic recombiner) to remove the hydrogen gases inside the containment in case of an accident. This paper aims to investigate a PAR device with catalyst sheets to calculate the hydrogen consumption, determine the temperature distribution, and PAR performance in the test facility. Test series were carried out in the test facility REKO-4 under atmospheric pressure and natural flow conditions. For the test purposes, hydrogen between 2.0 and 6.0 vol.% was injected into the vessel with the volumetric flow, 1.9 m^3/h. The first rise in the temperature of the PAR inlet was found after 100 seconds of first hydrogen injection, this result demonstrated that hydrogen consumption started. The results from the test series show that the PAR device can remove hydrogen and the PAR performance can be determined from the test results.
Keywords: Nuclear safety, Light water reactor, Hydrogen recombination, PAR, Recombiner, Nuclear accident

REKO-4 TEST DÜZENEĞİNDE HİDROJEN REKOMBİNASYONU İÇİN PAR CİHAZININ DENEYSEL İNCELENMESİ

Özet: Fukuşima kazası, hidrojen birikimini önlemek için reaktörlerdeki pasif güvenlik sistemlerinin gerekli olduğunu kanıtladı. Bu sistemlerden biri olan PAR (pasif otokatalitik rekombiner), bir kaza durumunda koruma kabında oluşabilecek hidrojen gazlarının uzaklaştırılmasında kullanılır. Bu makalenin amacı, test düzeneğinde PAR cihazının performansını ve sıcaklık dağılımını belirlemek, hidrojen tüketimini hesaplamak için katalizör levhalı bir PAR cihazını araştırmaktır. Testler atmosferik basınç ve doğal akış şartları altında REKO-4 test düzeneğinde gerçekleştirildi. Test için, hacimce 2-6 % arasında hidrojen 1,9 m^3/h hacimsel debi ile kaba enjekte edildi. PAR girişinin sıcaklığındaki ilk artış, ilk hidrojen enjeksiyonundan 100 saniye sonra gerçekleşti, bu sonuç hidrojen tüketiminin başladığını göstermektedir. Testlerden elde edilen sonuçlar, PAR cihazının hidrojeni tüketebildiğini ve PAR performansının test sonuçlarından belirlenebileceğini göstermektedir.
Anahtar Kelimeler: Nükleer güvenlik, Hafif su reaktörü, Hidrojen rekombinasyonu, PAR, Rekombiner, Nükleer kaza

INTRODUCTION

In March 2011, an earthquake of magnitude 9.0 occurred off the eastern coast of Japan causing a 15-metre tsunami. This disaster resulted in a loss of coolant accident in the Fukushima Daiichi Nuclear Power Plant. Insufficient cooling of the core led to zircaloy oxidation and hydrogen generation by the interaction of overheated steam with zircaloy fuel cladding. Hydrogen leaking into the reactor buildings resulted in explosions in NPP units 1,3 and 4. The explosions damaged structures of units and radionuclides were released to the atmosphere contaminating land and sea (Simon *et al.*, 2012).

One of the lessons learned from the Fukushima accident is the necessity of installing passive safety systems for the control of hydrogen inside the containment. So-called passive auto-catalytic recombiners (PARs) have been back-fitted to European NPPs since the 1990s (Reinecke *et al.,* 2013) (Häfele 2012).

Experiments performed on PARs aim to analyse device behavior and performance. In order to process data, the computer code REKO-DIREKT was developed in cooperation of RWTH Aachen and Forschungszentrum Jülich (FZJ). Before this research, other researchers have worked on PAR performance with different conditions. First experiments were performed in the REKO-3 facility under well-defined forced-flow conditions that were focused on the processes inside the catalyst section of a PAR device. In a next step, REKO-4 has been developed to focus more on chimney flows, PAR design and its interaction with the surrounding conditions (Simon *et al.*, 2012).

This paper describes an investigation of hydrogen recombination in REKO-4 under natural flow conditions. On this project at IEK-6/FZJ, the research was performed to illustrate PAR reaction capability. Moreover, the catalyst temperatures at different heights of the catalyst sheets were measured by thermocouples to have a better understanding of the reaction process. During the experiments, measurements such as temperature, pressure, and hydrogen concentration were taken by using measurement devices such as thermocouples, pressure gauges, thermal conductivity gauges (katharometers). Different analyses were done, and final data was discussed and evaluated to have a better understanding of PARs working principle and hydrogen recombination.

Hydrogen recombination by PARs

Hydrogen recombination is one method of hydrogen consumption by a device in order to prevent hydrogen accumulation and explosion. The release rate and the total amount of hydrogen depend on the reactor type and the particular severe accident.

In case insufficient core cooling, zirconium as fuel cladding material reacts with steam at the temperature above 1200 °C and zirconium oxidation occurs. This adds extra thermal load due to the exothermic reaction. Additionally, hydrogen is produced as product of the reaction. The stoichiometric equation for oxidation of the zircaloy cladding with steam is:

$$Zr + 2H_2O \rightarrow \mathrm{Zr}O_2 + 2H_2 + Q \qquad (1)$$

where Q = $-586\ kJ\ mol^{-1}$ is the exothermic heat release per unit mole of zirconium oxidized. (Häfele 2012)

PARs are used to consume hydrogen. Hydrogen and oxygen react exothermally on the catalytic sheets inside a PAR device. This reaction generates steam and heat. The reaction heat creates a buoyancy-driven flow, which makes PAR devices completely passive, as PARs do not need external energy supply.

$$H_2 + \frac{1}{2}O_2 \rightarrow H_2O + Q \qquad (2)$$

where Q is heat.

In Figure 1 and Figure 2, recombination process and schematic size of PAR are shown shortly. In the leading part of PAR, catalyst sheets are located. The catalyst sheets are surfaces where recombination reaction occurs. They form a set of parallel vertical flow channels. Hydrogen inside the PAR is consumed with oxygen, and steam is produced as the reaction product. Since heat is generated during the reaction, the buoyancy-driven flow occurs. The buoyancy-driven flow is induced inside the chimney part, which ensures a continuous gaseous flow through the PAR (Reinecke *et al.*,2013). It is vital for the continuous hydrogen consumption in the recombination process. In order to assess the efficiency of PAR applications, studies of PARs modeling focus on;

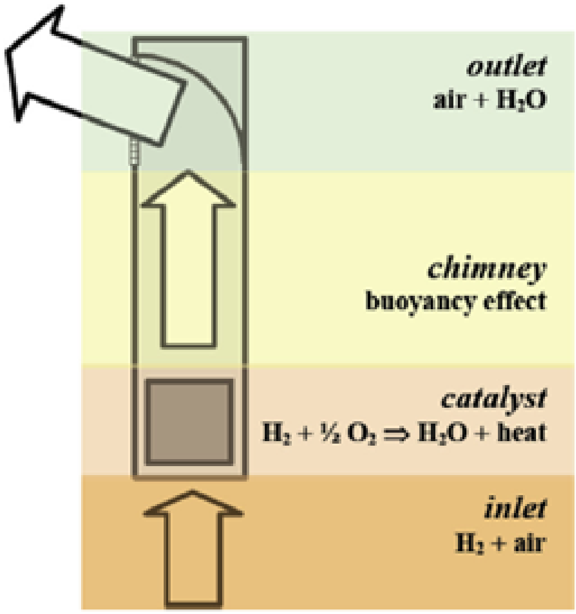

Figure 1 Recombination steps (Simon *et al.*, 2014)

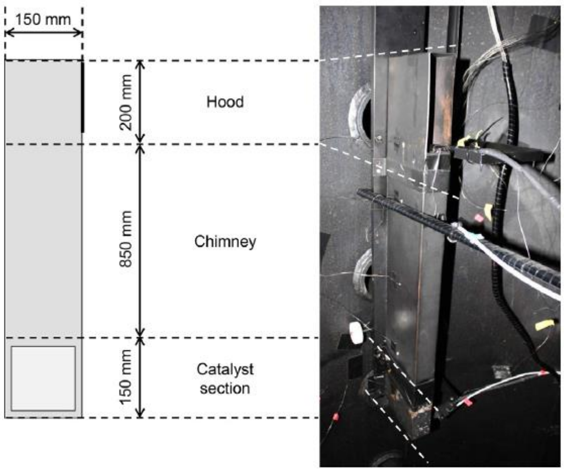

Figure 2 Schematic Size of PAR (Simon *et al.*, 2014)

- hydrogen consumption and buoyancy-driven flow
- preventing of catalyst poisoning
- possible ignitions at the hot parts as a result of exothermic reactions

The PAR (Figure 2), which has been used in the test series, consists of three parts: catalyst section, chimney section, and hood. It has a rectangular square section of approximately 5 x 15 cm² and a total height of 120 cm.

The catalyst section is equipped with four catalyst sheets (Figure 3), each with a thickness of approximately 1.5 mm (Bachellerie 2003). The catalyst sheets consist of stainless-steel plates and are coated with an aluminum oxide wash-coat and then with platinum.

DESCRIPTION OF THE TEST FACILITY

Test facility REKO-4

The test facility REKO-4 (Figure 4) is designed to investigate the operational behavior of PARs under natural flow conditions and consists of a pressure vessel with a free volume of approximately 5.3 m³. The vessel has an internal height of 3.7 m, an internal diameter of 1.4 m, and allows experiments with a maximum operating pressure of 2.0 bar (a). The design pressure of the vessel is 25 bar (a) at 280 °C to withstand possible hydrogen combustion loads. 32 flanges allow the equipment of the vessel with different measurement techniques and a manhole ensures good accessibility for the installation of the PAR and the inner instrumentation. Hydrogen is injected through a horizontal pipe with a diameter of 10 mm and the hydrogen injection rate is controlled by a mass flow controller (Bachellerie 2003).

Measurement devices

For the analysis of the behavior of the PAR and the interaction with the surrounding atmosphere inside the REKO-4 test facility, sensors for the measurement of the hydrogen concentration, temperature, humidity, and pressure are installed at different measurement positions.

Thermal conductivity gauges are used for the measurement of the hydrogen concentration. In total, 20 hydrogen sensors are distributed inside the REKO-4 vessel. Two (KR-07_H2 / KR-20_H2) of them are positioned at the PAR inlet and another one at the PAR outlet (KR-08_H2). Sensor KR-07_H2 (Figure 5) is located directly below the catalyst sheets and is strongly affected by the heat radiation of the catalyst sheets leading to measurement errors during the experiments. Therefore, sensor KR-20_H2 is laterally attached allowing a more accurate measurement. The remaining hydrogen sensors are attached to different mounting rails at the bottom, middle and top of the vessel.

The temperature measurement is another vital measurement technique used for the analysis of the PAR behavior. Therefore, thermocouples (TC) (Type K) are used to measure the temperature of the catalyst sheets, the PAR housing, and also the temperature of the surrounding atmosphere (Simon *et al.*,2012). To measure the temperature distribution along the catalyst sheets, drillings with a diameter of 0.6 mm inside the catalyst sheets are equipped with TCs (Figure 3 and Figure 6) (Bachellerie 2003). Further TCs are installed at the PAR inlet, PAR outlet, and inside the PAR chimney. To determine the surrounding conditions, approximately 40 TCs are positioned at the mounting rails and at the vessel wall.

Figure 3 Catalyst sheets


Figure 4 REKO-4 facility

Humidity sensors are located at the bottom and top of the vessel and register humidity changes due to the recombination reaction. Additionally, a manometer is installed within REKO-4 to measure the course of the pressure during the experiments.

Two different manometers are placed in REKO-4, these are analogue manometer and capacitive pressure sensors which measure pressure by detecting changes in electrical capacitance (Simon *et al.*,2012).

The position of the PAR, the hydrogen injection, the manometer, and the mounting rails for the TCs and hydrogen sensors are shown in Figure 7.

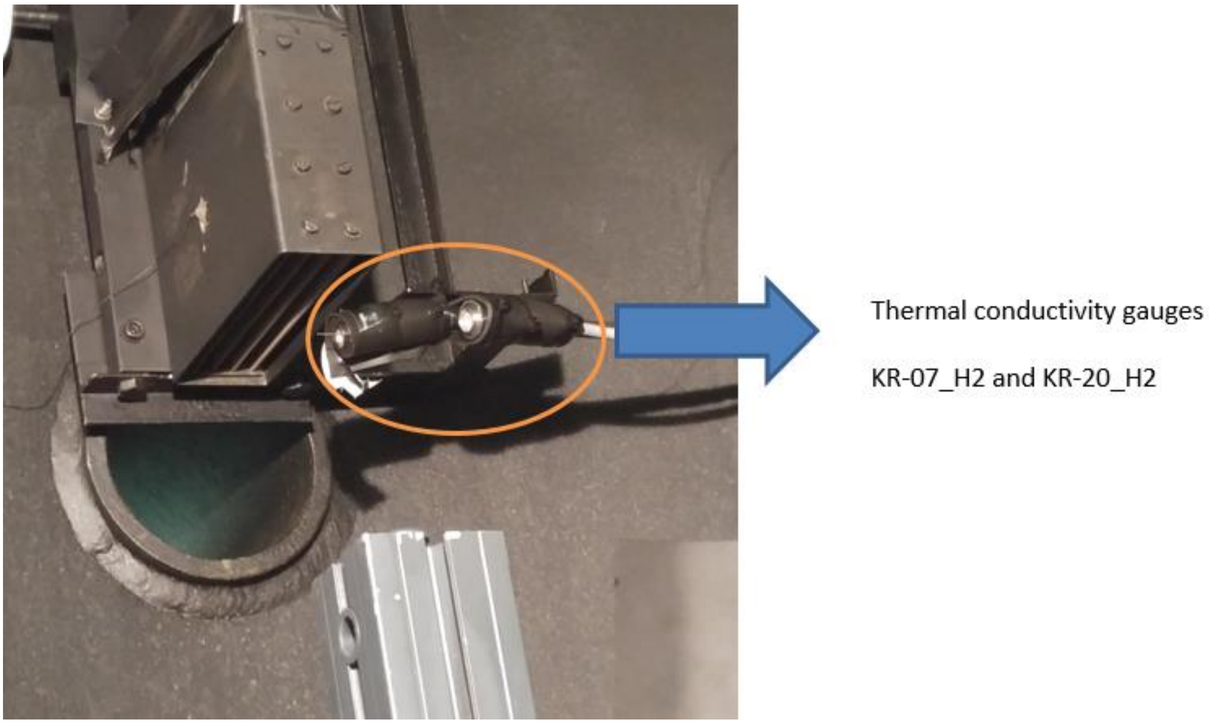

Figure 5 Hydrogen sensors at PAR inlet

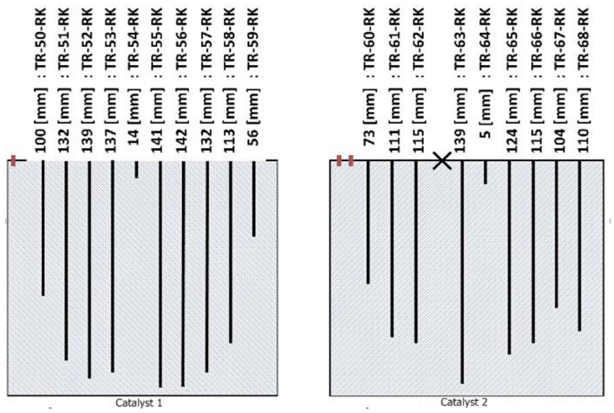

Figure 6 Thermocouples position inside catalyst sheet 1 and catalyst sheet 2

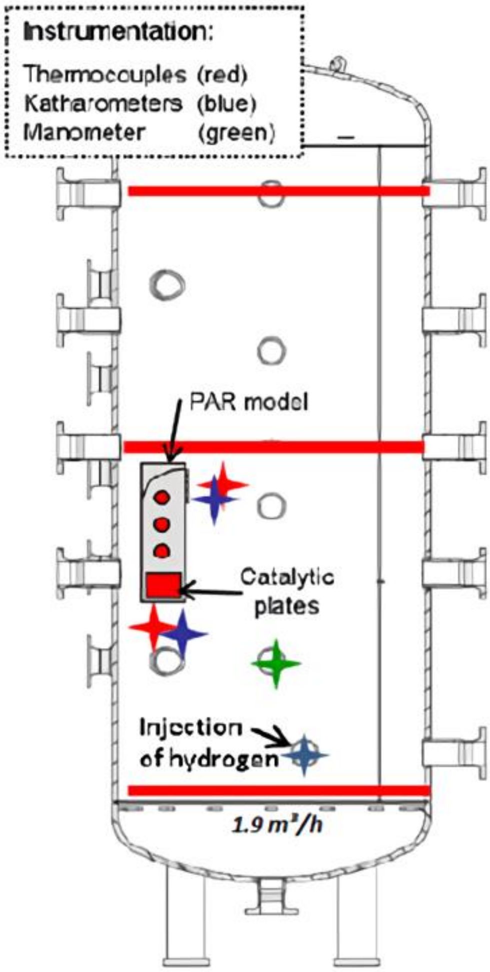

Figure 7 Locations of measurement devices in REKO-4 (Simon *et al.*, 2012)

REKO-4 TEST PROCEDURE

After the completion of the test setup the manhole is closed, sealed carefully, and a pressure test performed to ensure that there is no leakage. The experiments have been performed under natural flow conditions with an initial temperature of 28 °C, an initial pressure of 1 bar (a), and an initial absolute humidity of 1.42 g/m³. All the tests are performed under these conditions.

The experiments were started with the injection of hydrogen. The amount of hydrogen was chosen in accordance with the vessel pressure and the targeted hydrogen concentration. The hydrogen was injected with a rate of 1.9 m³/h.

During the experiments, the PAR inlet hydrogen concentration varied between 2.0 and 6.0 vol.% and further hydrogen was injected as soon as approximately

2.0 vol.% was reached. With the start of the hydrogen injection (t=0), the experimental data such as temperatures, hydrogen concentrations, and pressure were measured and recorded continuously by the measurement devices and the process control system, respectively. The first hydrogen injection phase took approximately 10 minutes. However, reaction at the catalyst sheets started already after 100 seconds, which was observed by the TCs inside the sheets (Figure 8). The reason for this temperature rise is the exothermic reaction of hydrogen with oxygen.

Several hydrogen injection phases followed to observe the operational behavior under quasi steady state conditions.

After the last hydrogen injection, the hydrogen concentration inside the vessel decreased within approximately 18 minutes from 5 to 1 vol.%, which is measured by a hydrogen sensor at the PAR inlet (Figure 9, deviating is t=0 defined as the moment when the last hydrogen injection is completed).

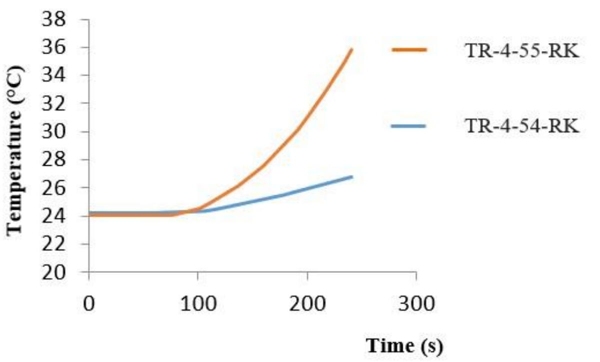

Figure 8 Catalyst sheet temperature during start-up phase (TR-4-54-RK is trailing edge of the catalyst; TR-4-55-RK is leading edge of catalyst)

The relative humidity inside the vessel increased as soon as the catalytic reaction started. After 500 seconds the relative humidity decreased due to the rise of the atmospheric temperature (Figure 10).

During the experiment, four hydrogen injections up to 6 vol.% of hydrogen were conducted with a total experiment duration of 5 hours. Figure 11 and Figure 12 show the changes of the temperature, hydrogen concentration, pressure, and humidity during the experiment. Numbers on the figures indicate number of hydrogen injections. For the pressure measurement, the values were read from PRSAH 4.01 sensor. Sensor TR_4_55_RK at the leading edge and sensor TR_4_54_RK at the trailing edge of catalyst sheet 1 were used to observe the temperature changes due to the exothermic reaction. Hydrogen sensor KR_4_20 at the PAR inlet and sensor KR_4_19 at the PAR outlet were employed to determine the recombination efficiency.

For the humidity measurement, HR_4_01 and HR_4_02 were used. As soon as the first hydrogen injection started, both hydrogen sensors show an increase of the hydrogen concentration inside the vessel. During the injection phase, hydrogen reached the catalyst and the TCs indicated a temperature increase. Moreover, a pressure and humidity rise occurred relating to the hydrogen concentration and reaction, respectively. After the injection was completed, the hydrogen concentration started to decrease due to the recombination reaction. The catalyst temperature decreased with the hydrogen concentration and lead to a reduced recombination rate.

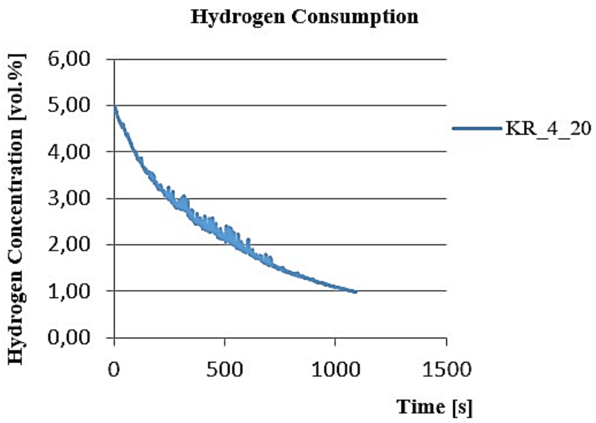

Figure 9 Hydrogen consumption after the last injection

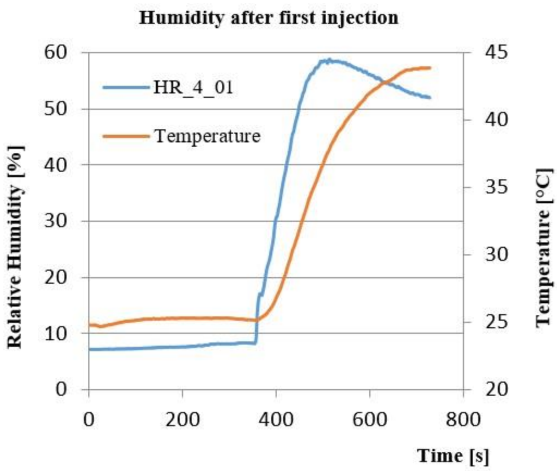

Figure 10 Humidity measurement and temperature change inside the vessel after start-up phase

EXPERIMENTAL RESULTS

The data from this experiment are used to evaluate PAR performance and catalysts working conditions. Furthermore, the analyses illustrate the operational behavior of PARs.

Hydrogen removal coefficient (λ)

PARs performance is a vital parameter of consuming hydrogen in the vessel. The hydrogen concentration is exponentially decreased with λ. λ gives hydrogen concentration decrease at a rate proportional to its current value, its unit is s^{-1}. The equation can be expressed as;

$$C = C_0 \times e^{-\lambda t} \tag{3}$$

C_0; $initial\ hydrogen\ concentraion$
C; $hydrogen\ concentraion\ after\ t\ time$
λ; $hydrogen\ removal\ coefficient$

Figure 9 shows hydrogen consumption after the last hydrogen injection and this equation is applied to the measurement data to find λ. For this test, λ is found to be 0.00148 s^{-1}. λ defines the performance of the PAR device and depends on temperature, pressure, and hydrogen concentration. It is an important parameter for the modelling of PARs.

Correlation between hydrogen concentration and catalysts temperature

The correlation of the hydrogen conversion and maximum catalyst temperatures provide clarification of the interactions of reaction kinetics, heat, and mass transfer, and the flow conditions inside the PAR. Since a maximum catalyst temperature was expected in the leading part of catalysts, “TR_4_55_RK” thermocouple data was analysed. For hydrogen conversion, PAR inlet hydrogen concentration was considered. Therefore, data was read from KR_4_20 katharometer.

Figure 13 clearly illustrates the correlation maximum catalysts temperature and hydrogen concentration at the inlet of PAR. With the increasing hydrogen concentration, the maximum temperature of catalysts is seen to have a logarithmic rise. Increasing temperature is the proof of exothermic recombination process.

Comparison of the measured catalyst temperature in dependence on the height of the catalysts

The temperature of the catalyst is measured with thermocouples that are inserted into the catalyst sheets at different heights. Thermocouples exact positions are illustrated in the Measurement section, Figure 6. It is expected to have variable temperatures with catalyst sheet heights. The reason of this expectation is arriving of rich hydrogen on the lower part (leading part) of catalyst sheets. Rich hydrogen with oxygen increases the temperature of the lower part of the catalyst sheet due to the exothermic reaction. The higher points are exposed to lower concentration since most of the hydrogen gases are already recombined in the lower part of catalyst sheets. Therefore, it is expected to observe higher temperature on the lower part.

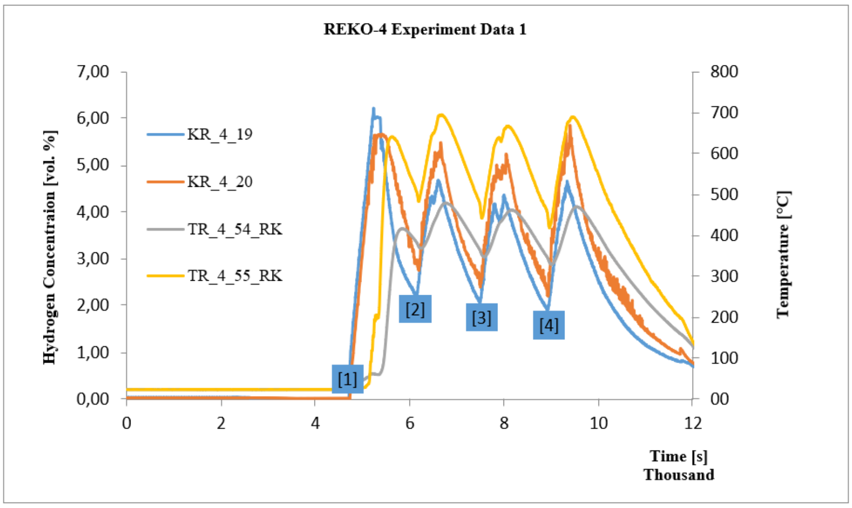

Figure 11 REKO-4 experimental data 1

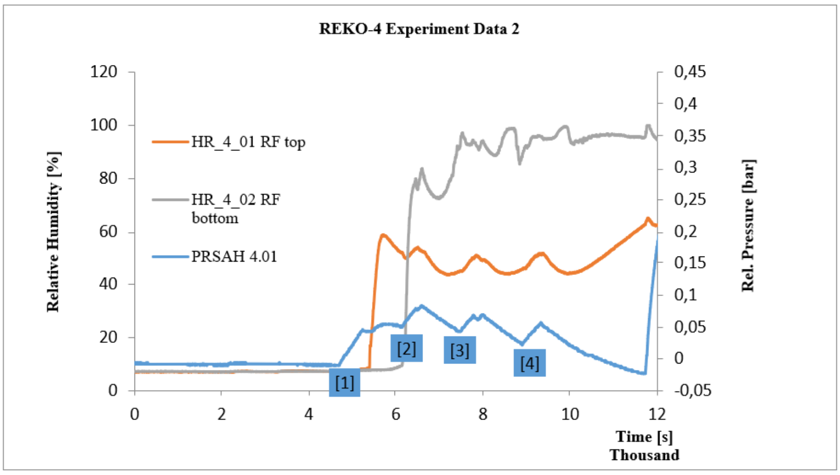

Figure 12 REKO-4 experimental data 2

Figure 14 shows briefly that temperatures decrease with the height of the catalyst sheet. ("0" reference point indicates the leading part of catalyst sheet in Figure 14.) In the experiment, different concentrations were measured such as 2, 4, 5.9 vol.%. In the 5.9 vol.% concentration measurement, the higher temperature difference occurs due to high concentration difference between the leading part and the trailing part of catalyst sheet.

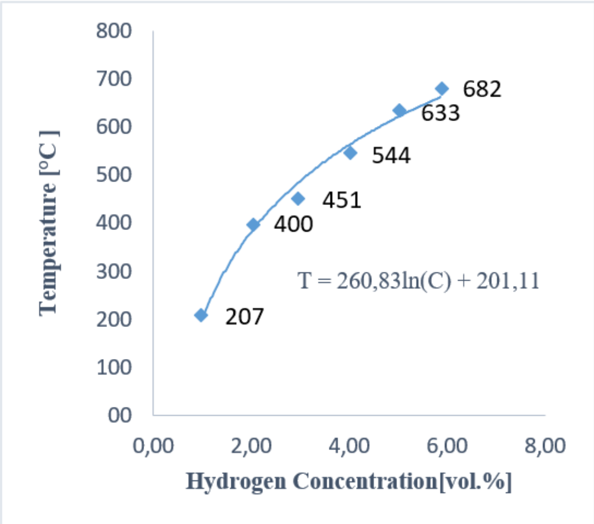

Figure 13 Correlation between PAR inlet concentration and maximum catalysts temperature

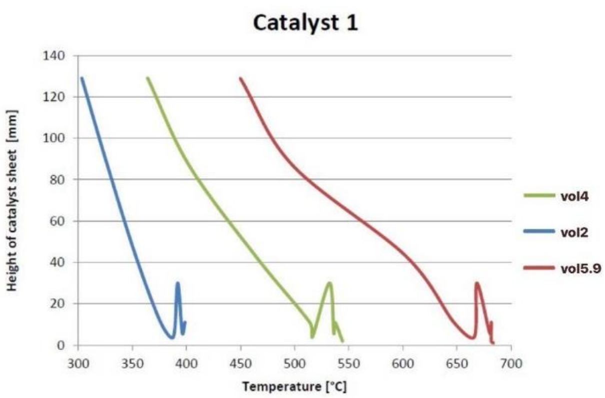

Figure 14 Catalyst 1, temperature over catalyst sheet height

CONCLUSION

Prior work has been performed in the REKO-3 facility, which was focused on the processes inside the catalyst section of the PAR. Those tests were performed under well-defined forced-flow condition.

In this experiment, the PAR in REKO-4 facility was tested. The main aims of these test series were to investigate detailed hydrogen recombination under natural convection conditions and to examine the PAR performance. Tests were performed under atmospheric pressure with hydrogen injection concentration between 2 and 6 vol.%.

The first temperature rise was found after 100 seconds of first hydrogen injection. The meaning of temperature rise is that hydrogen recombination started on the catalyst sheets after this time. However, it required more time, at least 200 seconds for start-up of PAR operation on the leading part of the PAR. After start-up of the PAR operation, a sharp decrease in the hydrogen concentration was observed and a reduction from 5 to 1 vol.% was achieved in approximately 18 minutes. This data provides that the PAR is a suitable device for removing hydrogen from a vessel under atmospheric conditions and conforms the purpose of installing PARs to prevent hydrogen accumulation. Additionally, this study indicates that the PAR is a completely passive device and can work without supplying any energy.

Moreover, another important correlation was seen between hydrogen concentration and catalyst sheet temperature after the catalyst sheet reached the working temperature. The experimental data showed that an increase of hydrogen concentration provides a rise in the catalyst temperature.

Finally, this study has revealed that the test facility, REKO-4, is suitable to perform hydrogen recombination tests with PARs under natural conditions. Test results and analyses support the idea that PAR devices can consume hydrogen gases released to the vessel and avoid hydrogen accumulation. As a summary, conclusion of this study with new boundary conditions,

- Hydrogen consumption proved that PAR device is ideal device for hydrogen recombination passively, several hydrogen concentrations were consumed successfully.
- REKO-4 is suitable test utility for detailed investigation of hydrogen recombination.
- PARs operational behavior (e.g. response time; first rise in temperature) is illustrated.
- PARs performance (e.g. hydrogen removal coefficient) is measured.

However, some limitations are observed. These test series were performed only under atmospheric pressure conditions. This study results should be compared with tests under different boundary conditions (e.g. different pressure) to verify the performance of the PAR. Secondly, the number of measurement devices should be increased in REKO-4 facility to get more detailed results. Using these analyses, future studies can be illustrated, and results can be used for modification of PARs.

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