PASSIVE AUTOCATALYTIC RECOMBINERS (PAR) INDUCED IGNITION AND THE RESULTING HYDROGEN DEFLAGRATION BEHAVIOUR IN LWR CONTAINMENTS

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ABSTRACT

The Fukushima Daiichi nuclear accident of 11 March 2011 indicated that in the event of a severe accident in Light Water Reactors (LWR), large amounts of hydrogen may be generated by core degradation and released into the containment. Even if several hydrogen mitigation measures such as, containment inerting, igniter, and Passive Autocatalytic Recombiners (PAR) are available, hydrogen deflagration at relatively low H_2 concentrations, e.g. below 10 or 12 % in the presence of an operating PAR or even by a random ignition source might not be completely avoided. Depending on locally available hydrogen concentration, turbulence and structural configurations within the containment, ignition initiated by a PAR can cause deflagration or under certain conditions even local detonations. The resulting pressure and temperature levels may pose a threat to the integrity of the containment building. Therefore, PAR inlet conditions at which ignition may occur are important to be determined experimentally in order to assess the possible combustion mode upstream of a PAR by using validated safety analysis tools for the reactor case.

In the present paper, PAR-inlet conditions leading to an ignition and the resulting hydrogen deflagration behaviour in a closed vessel atmosphere are discussed. Tests reported in this paper have been conducted in the frame of OECD/NEA THAI project (2007 - 2009). Performance of three different commercially available PAR designs (based on plate- and pellet-type catalysts) have been investigated in the THAI test facility (H = 9.2 m, D = 3.2 m, V = 60 m^3) under accidental conditions. Results indicate that a PAR exposed to hydrogen concentration higher than about 5.5 vol % can act as ignition source for the hydrogen-air-steam mixture present in the PAR environment and initiate a hydrogen deflagration. PAR induced ignition is directly correlated with the catalyst surface temperature which in turn depends on the H_2 concentration present at the PAR inlet. Following an ignition, flame propagation starts always at the upper end of the PAR in upward direction. The course and strength of a hydrogen deflagration initiated by PAR ignition depends on the available gas composition nearby the PAR outlet as well as gas distribution in the surrounding vessel atmosphere.

Keywords: PAR, ignition, hydrogen deflagration, severe accident, containment

1. INTRODUCTION

In case of a severe reactor accident hydrogen gas can be generated by metal-water reaction during core degradation and core-concrete interaction at high rates in the short term, and by water radiolysis at a low, continuous rate in the long term. Released into the air-filled containment, hydrogen can form flammable or even detonable mixtures, thus posing a threat to the integrity of the containment. After the occurrence of the TMI- 2 accident, different countermeasures are already being employed in several nuclear reactor types for hydrogen control and risk mitigation, e.g, dilution of combustible gases, inertization of the atmosphere, removal of the hydrogen by deliberate ignition or recombination [1]. However, in order to ensure that these mitigation systems will perform optimally during an early as well as late phase of an accident, their performance behaviour under a wide range of accidental conditions shall be known. The

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need to investigate and understand the hydrogen transport and distribution in the containment compartments as well as the effectiveness of mitigation measures under postulated severe accident conditions has also been affirmed by the the nuclear accident at the Fukushima-Daiichi plant [2]. For this purpose, both large scale experiments reasonably representative of dominant accident sequences and accident simulation tools validated against experimental data are necessary for the analysis of the containment response during accidents and the planning of accident management measures.

Passive Autocatalytic Recombiner (PAR) based hydrogen mitigation system is one of the established strategies to mitigate the possible hydrogen risk during design basis or severe accident conditions in a nuclear reactor containment. The name “Passive Autocatalytic Recombiner” describes the two essential features of a PAR: “Autocatalytic” because the catalytic process is self-starting and “passive” because it requires no active energy supply from outside. Depending on the reactor design and taking into consideration a wide spectrum of accident scenarios for hydrogen release, transport, and distribution within multi-compartment containment structure, typically 30 to 60 PARs are installed inside a LWR containment [3].

Generally, a commercial PAR unit is an open ended vertical channel equipped with catalytic material in the lower part. In case of an accident with hydrogen release, hydrogen enters a PAR unit already diluted and pre-mixed with air (-steam). If hydrogen-air(-steam) mixture comes in contact with the catalyst surface the hydrogen and the oxygen of the air become “recombined” to steam and the reaction energy leads to a temperature increase. The hot, hydrogen-poor gas ascends to the upper end of the box (chimney effect) and gives room to fresh, hydrogen-rich gas to be subsequently recombined by the catalyst. The action is self-starting and continues until the hydrogen (or oxygen) in the vicinity has been consumed. The majority of the commercial PAR vendors utilize platinum or palladium as catalyst material coated on a base material in the form of plates or pellets [3]. The natural convective flow-loop produced by PAR operation promotes mixing of the combustible gas mixture in the containment. As PAR does not need an active energy source, the availability of mitigation measure even during station black out scenario can be ensured.

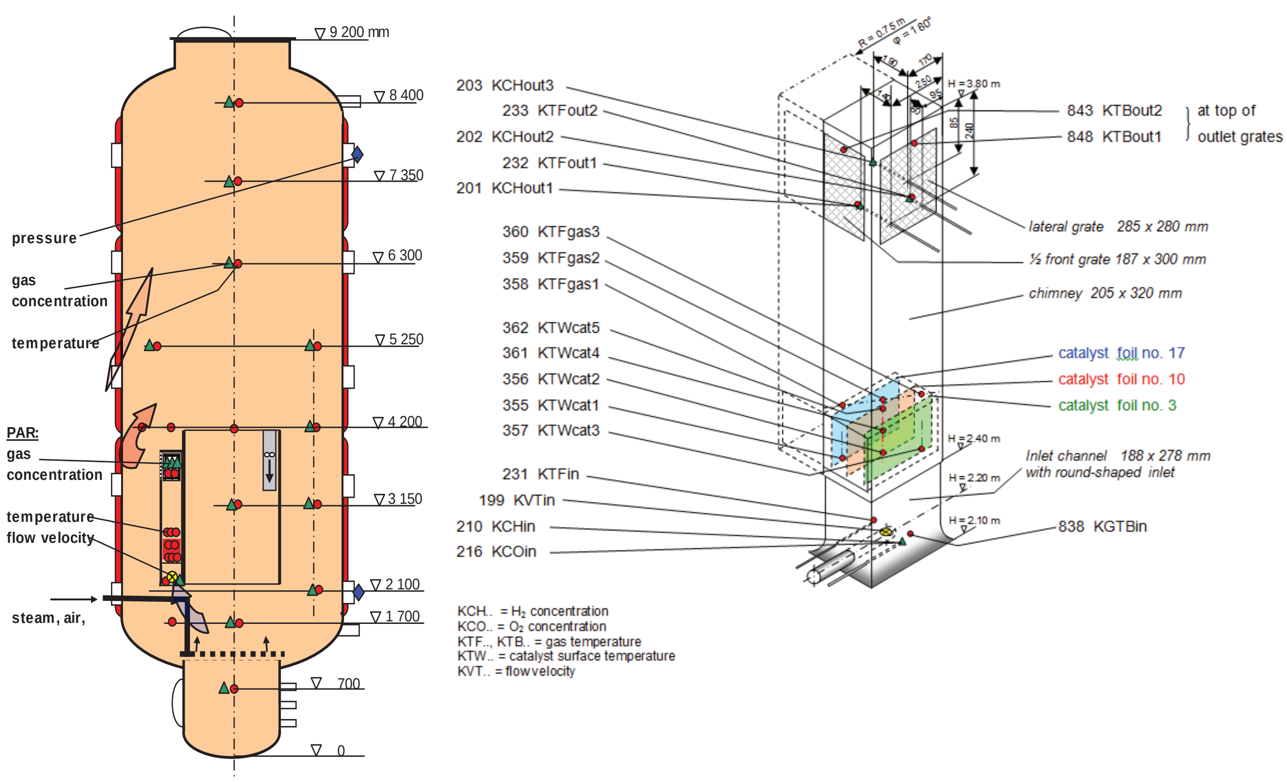
Data on the behaviour of PAR types under accident conditions were elaborated earlier by the vendors and also in some research projects carried out by Battelle Frankfurt in the 640 m^3 BMC test facility [4], tests in 90 m^3 Surtsey vessel at Sandia [5], and by CEA Cadarache in the 16 m^3 KALI vessel [6], and others. These experimental programs successfully demonstrated the potential of PAR based system for hydrogen mitigation by confirming their early start-up behaviour and long-term capability to recombine hydrogen in the present of an excessive amount of steam or even under presence of potential catalytic poisons. However, availability, consistency and completeness of the PAR performance data under a range of accident typical conditions are limited. A systematic PAR database required for the purpose of code validation and development purpose is also considered to be sparse [7]. Additionally, the performance of a PAR under accident-typical conditions, such as PAR operation in an oxygen lean atmosphere, influence of adverse flow conditions (e.g. wall effect, condensation induced counter- current flow), interaction with fission product, exposure to high hydrogen release rate and elevated hydrogen concentration is not well understood.

In the above-mentioned context, 32 Hydrogen Recombiner (HR) tests conducted in the frame of OECD/NEA THAI project [8] provide a valuable database both for hydrogen control concept demonstration and code validation purposes by investigating three different commercially available PAR units under severe accident representative conditions. The large THAI vessel allows PAR operation with unrestricted natural convection which includes interaction of PAR performance and vessel atmosphere distribution. The test series evaluates the influence of various parameters which may have adverse effects on PAR performance, such as: steam content, oxygen starvation, containment pressure, and ignition initiation by PAR, etc. Interaction of PAR operation and fission product in the vessel volume is an additional objective with particular focus on investigating metal iodide conversion to gaseous iodine by an operating PAR and possible PAR poisoning under adverse operating conditions.

The focus of the present paper is on PAR induced ignition behaviour. For accident scenarios with excessive hydrogen release rates, some of the PAR units in the containment can become overloaded and reach extremely high operation temperatures, which may cause an ignition of the steam-air-hydrogen atmosphere. The relevance to reactor safety is the destructive potential of deflagrations with the peak pressures and high temperatures involved. The PAR inlet conditions at which ignition may occur are important to be determined experimentally in order to assess the possible combustion mode upstream of a PAR by using validated safety analysis tools. Above PAR location, all the combustion modes are possible for the same accident scenario depending on the available gas composition, geometry, operating safety systems (e.g. spray, cooler, PAR, venting), and the prevailing thermal hydraulic conditions (e.g. pressure, temperature, turbulence). Starting as a slow hydrogen deflagration near the ignition point, the hydrogen flame can be strongly accelerated if favourable conditions are encountered along its pathway. Therefore, PAR induced ignition and the resulting H_2 deflagration behaviour in the surrounding atmosphere provide a basis to assess the severity of a hydrogen explosion which is related to the propagation speed of the deflagration.

2. TEST FACILITY AND INSTRUMENTATION

The tests have been conducted in the technical-scale THAI (Thermal-hydraulics, Hydrogen, Aerosols, and Iodine) containment test facility, a 60 m³ stainless steel vessel 9.2 m high and 3.2 m in diameter. The schematic diagram of the THAI test facility is shown in Fig. 1a.



(a) (b)

Fig. 1: a) THAI test configuration for the HR test series, b) PAR specific instrumentation

The THAI vessel is designed for a maximum overpressure of 1.4 MPa at 180°C and can withstand moderate hydrogen deflagrations. The facility is equipped with the necessary supply systems for steam, compressed air, nitrogen, oxygen, light gases (helium and hydrogen), aerosols and gaseous iodine, the latter also possible to be labelled with radioactive tracer I -123. The vessel walls can be heated and/or cooled to establish the desired thermal hydraulic conditions [8,9].

Fig.1 also shows the location of the test PAR unit in the THAI vessel. PAR specific instrumentation using AREVA PAR as an example is shown in Fig. 1b. The PAR unit was installed on the outer side of a hollow cylinder (1.4 m diameter and 2 m in height). PAR units based on plate-type catalysts (provided by AREVA GmbH, Germany and AECL now CNL, Canada), and pellet-type catalyst (provided by NIS Ingenieurgesellschaft mbH, Germany) have been operated in a comparable manner in the 60 m³ THAI test vessel.

For the tests, the capacities of the available plate-type PAR units have been scaled down to the size of the THAI facility by reducing the number of catalyst plates or foils to approximately 50 % of the original number, and by inserting a vertical partition wall to reduce the active PAR flow cross section accordingly, see Fig. 2. The total catalytic surface of the investigated PAR units was in the range of 1.44 m^2 – 1.89 m^2. The large size of the THAI test facility allows operation of medium-sized commercial PAR units with free, unrestricted natural convection which includes interaction of PAR performance and vessel atmosphere distribution. Hydrogen is released via the ring-shaped perforated feed line at elevation H = 1.26 m. Due to buoyancy, the released hydrogen ascends and distributes almost homogeneously within the vessel volume above. It also reaches the PAR inlet at its lateral position at elevation H = 2.1 m.

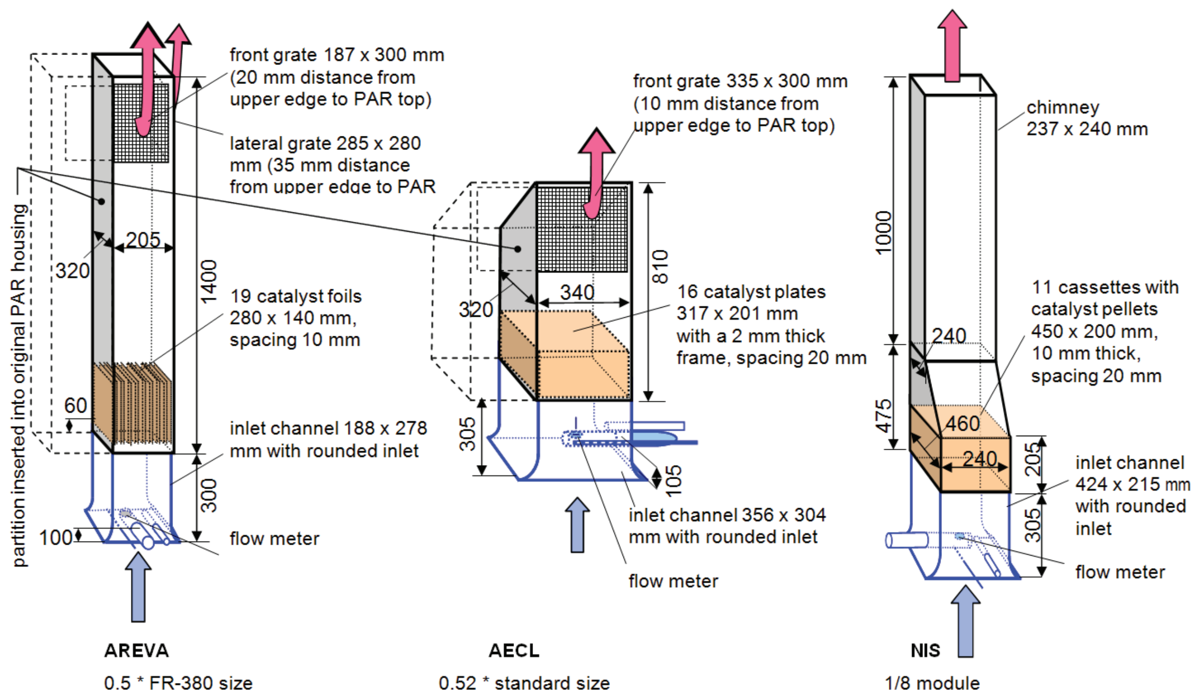


Fig. 2: PAR units tested in the THAI test facility.

A dense instrumentation for measuring local gas concentration and temperature in the vessel atmosphere is installed to evaluate PAR interaction with the surrounding gas atmosphere.

Each PAR unit has been equipped with instrumentation to measure inlet and outlet H_2 concentration and temperature, catalysts surface temperature and flow velocity at the PAR inlet (Fig. 1b). For adequate determination of their performance, the PAR units have been equipped with an additional instrumented inlet flow channel which allows local volume flow measurement at the PAR inlet and thus in turn makes it possible to directly calculate the H_2 recombination rate of the PAR unit. Comparison tests with and without flow channel indicate no negative influence on the PAR function.

Hydrogen gas concentration at the PAR and in the vessel atmosphere is measured with 15 sampling based thermal-conductivity sensors (accuracy ± 0.2 vol %). As thermal conductivity sensors only provide dry-gas concentration (H_2 concentration in air), steam content in the experiments is determined using other means, such as thermal hydraulic mass balance or directly by measurements with relative humidity sensors.

Two of the H_2 concentration measurement locations (one at the PAR inlet and one in the vessel atmosphere) are also equipped with electro-chemical principle based O_2 sensors (accuracy ± 0.5 vol %). Total vessel pressure is measured with strain-gauge-type pressure transducers backed up by the readings of a high-precision manometer (accuracy ±4 mbar).

To monitor the teamperature and pressure peaks due to hydrogen combustion, "fast" sheathed 0.5 mm diameter thermocouples (measuring range 0 - 1200 K, accuracy ± 2.5 K up to 333°C and ± 0.75% above 333°C) and pressure transducer with eigenfrequency of 35 kHz are used. Data acquisition with "fast" transducers is performed in 1 ms time steps.

3. TEST MATRIX

Table 1 provides the initial boundary conditions for the tests investigating PAR performance (H_2 recombination onset, recombination rate, and H_2 depletion efficiency) as a function of initial thermal-hydraulic conditions, oxygen-starvation condition, and PAR overload by high hydrogen concentration to investigate PAR induce ignition behaviour. Majority of the tests are conducted using the plate-type AREVA and AECL PARs. Only a limited number of tests at 1.5 bar pressure, 74 °C gas temperature, and 25 vol % steam are performed with the pellet-type NIS PAR. In this paper mainly results obtained with AREVA PAR are discussed.

Table 1: PAR test matrix

Test parameters \ Test objectives		PAR performance	Oxygen starvation	Ignition potential
p = 1.0 bar	25 °C, dry	x		x
	86 °C, 60 % steam	x	x	
p = 1.5 bar	25 °C, dry	x		x
	74 °C, 25 % steam	x		x, xx*
		x	x	
	90 °C, 47 % steam	x		x
		x	x	
	97 °C, 60 % steam	x		x
		x	x	
p = 2.2 bar	25 °C, dry	x		x, xx
	108 °C, 34 % steam	x	x	
p = 3.0 bar	25 °C, dry	x		
	117°C, 60 % steam	x	x	
		x		x

* multi-ignition tests

In case of "multi-ignition" tests, H_2 release has been continued after the first ignition in order to provoke additional ignition events. In addition to the 30 PAR performance tests, two experiments HR-31 and HR-32 were specifically designed to study the fission product interaction with an operating PAR. The test HR-31 refers to the conversion of metal iodide into gaseous iodine by PAR operation, which is of relevance for the radiological source term. Challenging experimental test conditions were necessary with PAR catalysts plate temperatures up to 900°C and aerosol concentration > 4 g/m^3 to obtain significant iodine conversion. The second experiment HR-32 was designed to investigate the potential poisoning effect by exposure of PAR initially covered with condensed steam to aerosols (hygroscopic and inert aerosol mixture), I_2 (iodine radio-tracer I-123). Results of HR-31 and HR-32 tests are reported in [9].

4. TEST PROCEDURE

A typical test procedure is shown in Fig. 3 by using test results of a plate-type PAR. Majority of the PAR performance tests consisted of four test phases with ascending and descending H_2 concentrations following the two consecutive H_2 injections in phase-1 and phase-3. Descending H_2 concentration is also the prevailing mode in the course of reactor accidents, periods of ascending H_2 concentration are comparatively short and therefore less important for the overall PAR behaviour.

In the test phase-1, H_2 is released at a moderate rate (~ 0.15 - 0.2 g/s) into the test vessel. Immediately after onset of PAR operation, H_2 is switched to higher injection rate (~ 0.30 - 0.45 g/s) resulting in further increase of H_2 concentration and H_2 recombination rate. Hydrogen injection is interrupted as soon as a level of approximately 5.5 vol % H_2 at the PAR inlet (i.e. below the expected PAR ignition level) has been reached. In the test phase-2, measurements of decreasing H_2 concentrations and other relevant parameters, such as H_2 recombination rate, H_2 depletion efficiency are used to determine the PAR performance. Prior to starting the second test phase, O_2 in vessel atmosphere is replenished if necessary for a test with "ignition" or further reduced by injecting nitrogen for a test with "oxygen starvation". For the tests specific to "ignition", O_2 surplus ratio is maintained well above 2.0 before starting the second H_2 release. Oxygen surplus ratio is defined as $\Phi = 2 * C_{O2} / C_{H2}$, where C_{O2} and C_{H2} are oxygen and hydrogen concentrations at the PAR inlet, respectively.

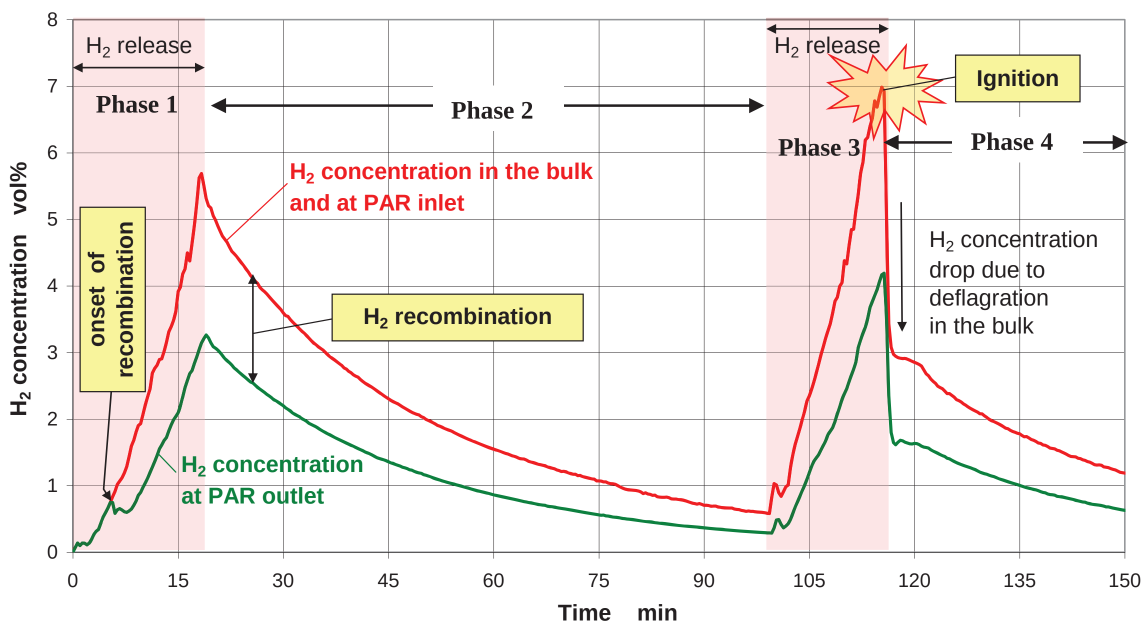


Fig. 3: Example of the test procedure for the PAR "ignition" specific tests

In the test phase-3, H_2 injection is again resumed at higher injection rate. For the investigation of PAR ignition, H_2 concentration is increased until the operating PAR becomes so heavily loaded that ignition occurs. Immediately following an ignition, H_2 release has been terminated. Test phase-4 starts after ending the H_2 release. Ignition leads to a sudden H_2 concentration drop due to the initiated deflagration. In case of an incomplete combustion, PAR continues to recombine the remaining H_2 concentration in the vessel atmosphere. Test ends when PAR comes to a standstill or if H_2 concentrations in the vessel atmosphere have been reduced below 0.3 vol %.

5. RESULTS AND DISCUSSION

PAR performance

Test results indicate that both plate-type and pellet-type PARs are self-starting even at relatively cold and wet conditions, as soon as a specific threshold value of the local hydrogen concentration is exceeded. The first indication of hydrogen recombination onset is a moderate increase in catalyst temperature. For the complete PAR performance test series, test results indicate that H_2 concentration required for the onset of hydrogen recombination by PARs varies from 0.2 vol% to 4.4 vol% depending on temperature, pressure, and steam content. Dry atmosphere, elevated pressure and temperature promoted early recombination onset. Steam-saturated conditions resulted in delayed onset. Once being heated up the PARs remain operating until a lower concentration threshold of approximately 0.3 vol% H_2 has been reached.

After onset of H_2 recombination and the resulting heat development at the catalyst, PARs are self-feeding due to the buoyancy effect inside the chimney. The resulting hydrogen recombination rate is mainly dependent on PAR size and type, hydrogen concentration available at the PAR inlet and the vessel pressure. Test results shown in Fig. 4 indicate that hydrogen recombination rate by the PAR increases nearly proportional to the hydrogen concentration and to the operation pressure. The effect of increasing steam content combined with an increasing temperature in THAI tests was determined to be very small on the measured hydrogen recombination rate.

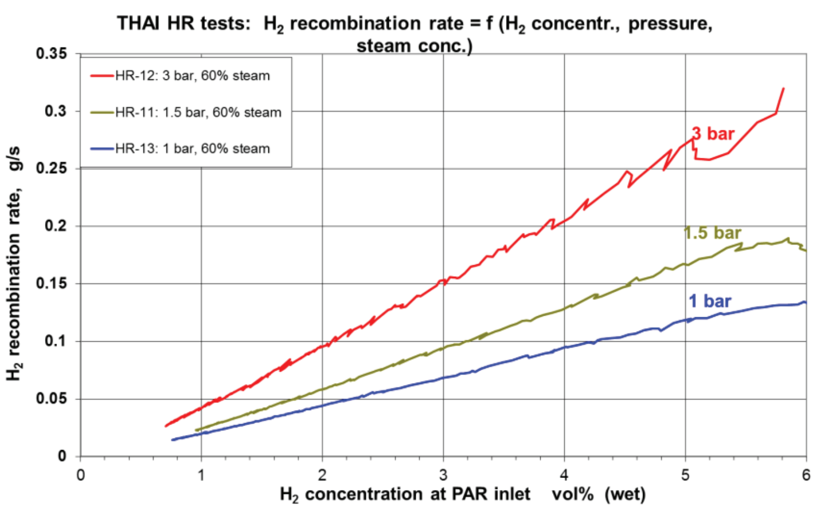


Fig. 4: PAR recombination rate as a function of H_2 concentrations and pressures

Hydrogen recombination in a PAR is incomplete and this can be quantified by hydrogen depletion efficiency γ (in %) calculated from the measured H_2 concentrations at the PAR inlet and outlet, $\gamma = (C_{H2in} - C_{H2out}) / C_{H2in} \cdot 100$. Hydrogen depletion efficiency at a given H_2 concentration is also PAR design specific as it mainly depends on the gas residence time in the PAR catalyst zone and on the diffusion length from the vertically flowing gas to the catalyst surface (and probably also on catalyst material). As depicted in Fig. 5, at low H_2 concentrations (< 0.5 vol %), depletion efficiency increases with decreasing H_2 concentration. This is caused by decreasing flow velocity and increasing residence time of the gas in the catalyst zone. At higher H_2 concentrations (>2 vol %), depletion efficiency is independent from the H_2 concentration. H_2 depletion efficiency decreases with an increase in pressure.

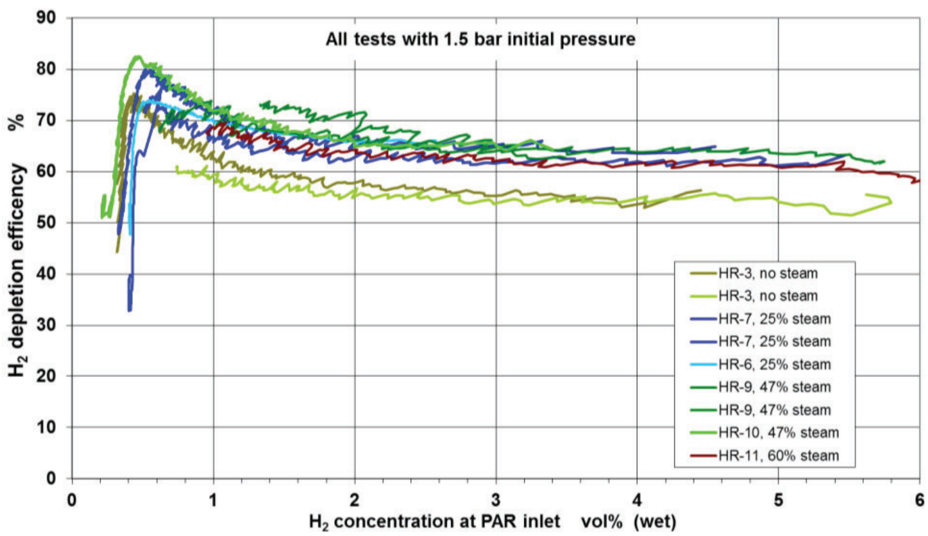


Fig. 5: H_2 depletion efficiency as a function of H_2 concentrations and steam content

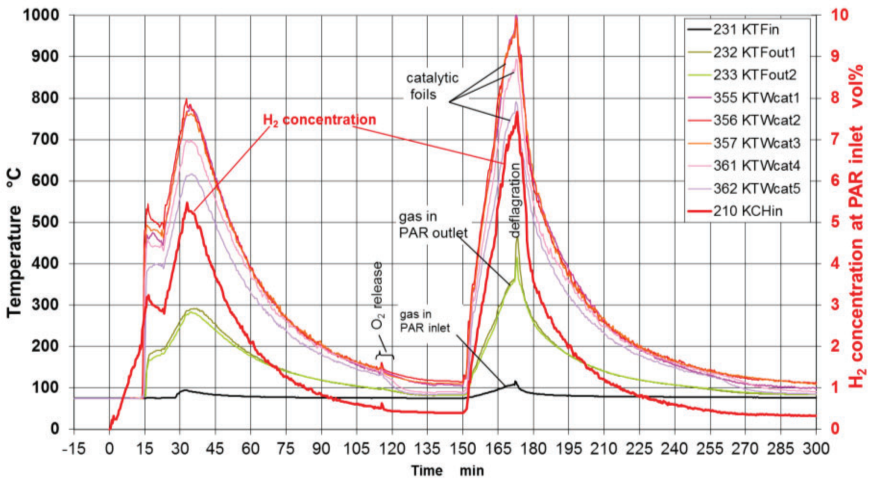
For the three investigated PAR designs, the hydrogen depletion efficiency was determined to be varying between 40 - 70 % in an atmosphere containing sufficient oxygen surplus. A minimum oxygen surplus ratio Φ between 2 and 3 (depending on the PAR design) is necessary to ensure unimpaired PAR performance independently from steam content. The minimum value of oxygen surplus ratio is significantly higher than the stoichiometric ratio (Φ = 1). As soon as Φ falls below 2, the hydrogen recombination rate and catalyst temperature decrease drastically. At oxygen surplus ratio Φ = 1, the PAR capacity falls below 50 % of the design capacity.

PAR induced ignition

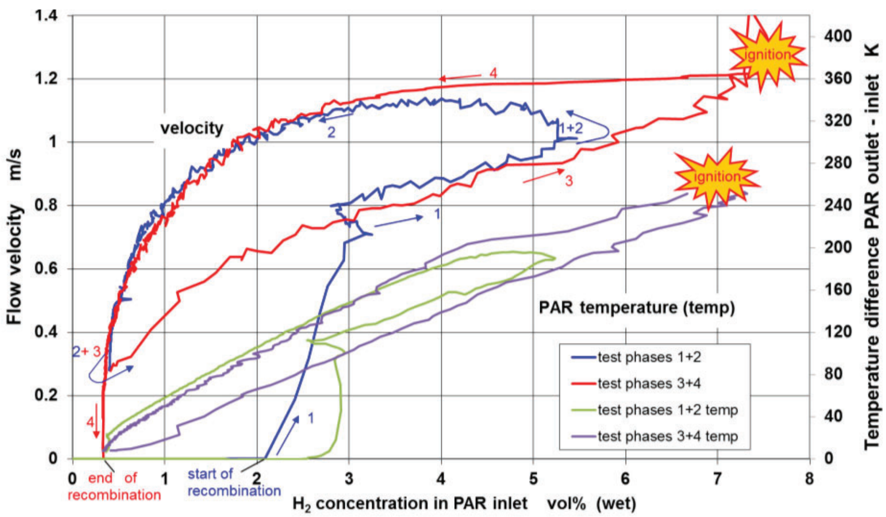
THAI test results indicate that a PAR exposed to a high hydrogen concentration acts as an ignition source for the hydrogen present in the PAR environment (respectively in the test vessel volume) and can initiate a hydrogen deflagration.

Majority of the PAR ignition tests followed the test procedure consisting of four test phases as defined in the section “test procedure”. A selected number of tests were also conducted with one H_2 release consisting of phase-1 and phase-2 and ignition occurred at the end of phase-1.

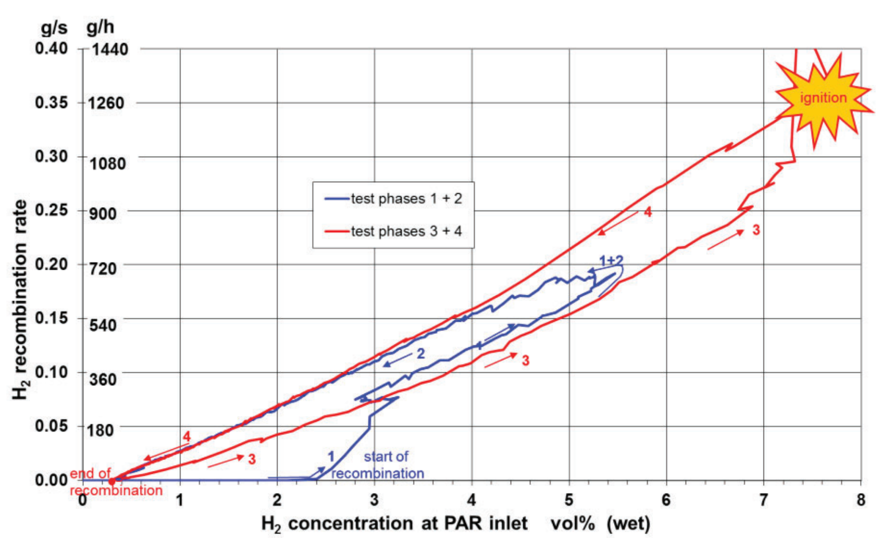
In the following, test HR-7 is exemplarily discussed to demonstrate the PAR induced ignition behaviour. This test is conducted following the standard test procedure with four test phases. After establishing the initial test conditions (1.5 bar pressure, 74 °C gas temperature, and 25 vol % steam), from t = 0 min onwards, hydrogen is released at a moderate rate of 0.19 g/s. Due to buoyancy, the released hydrogen ascends and distributes almost homogeneously within the vessel volume and also reaches the PAR inlet. Onset of H_2 recombination occurred 14 min after starting the H_2 injection and at PAR inlet H_2 concentration of 2.4 vol %. Onset of H_2 recombination by PAR is indicated by temperature rise at the catalyst foils, by H_2 concentration decrease at the PAR outlet (Fig.6a), and by the start of flow meter operation in the PAR inlet channel (Fig. 6b).



a) PAR catalysts temperature and inlet H_2 concentration



b) Flow velocity at PAR inlet and PAR inlet-outlet temperature difference



c) H_2 recombination rate

Fig. 6: HR-7 test: PAR performance results

The measured data in Fig. 6b also indicate that H_2 concentration of about 2.4 vol.% at PAR inlet results in a small PAR inlet-outlet temperature difference which is able to create measurable convective flow (lower limit for turbine flow meter is 0.25 m/s) through the PAR. After onset of H_2 recombination, the catalyst temperatures increase almost linearly with H_2 concentration. As soon as the catalyst temperature has approached to 800 °C and the H_2 concentration at the PAR inlet to 5.5 vol%, H_2 release has been interrupted to prevent early ignition. Then the ongoing H_2 recombination by the PAR results in a gradual decrease of H_2 mass in the test vessel. This long-lasting period of hydrogen decay in phase-2 yields a large amount of data for a reliable determination of the H_2 recombination rate as a function of H_2 concentration (Fig. 6c).

Test phase-3 starts with release of H_2 at a mass flow rate of 0.38 g/s. As far as PAR performance is concerned, H_2 concentration rise shows the same effects as observed in test phase-1. The end of the H_2 release in this phase is given by a hydrogen deflagration in the upper vessel zone, resulting into sudden drop in H_2 concentrations. Due to deflagration, vessel pressure rise from 1.512 bar to 1.825 bar. Afterwards, the residual hydrogen is gradually recombined by the PAR. Prior to ignition, concentrations at PAR inlet were 7.35 vol% H_2, 24 vol% steam and 13.2 vol% O_2 and maximum catalyst temperature was 975 °C. The H_2 recombination data determined during second H_2 depletion period (with an extended H_2 concentration range) are in good agreement with those of the first H_2 depletion period.

In addition to the THAI PAR ignition tests following the above-mentioned test procedure, two PAR tests HR-29 and HR-30 (initial steam content of 60 vol %,) have been conducted to study the effect of steam condensation on the resulting H_2 deflagration behaviour. In case of excessive steam release under steam-inerted, not-flammable conditions, high local H_2 concentrations can be formed. If subsequently steam concentration rapidly drops, e.g. due to spray operation, ignition by a PAR may occur at a high H_2 concentration level and result in a severe deflagration. In these two THAI tests, the THAI vessel is operated with continuous wall cooling. At first, steam-inerted conditions have been achieved by continuous steam and hydrogen release. Additionally, oxygen has been fed to compensate the oxygen consumed by recombination. Then, steam release has been stopped to rapidly decrease steam content and reach flammable conditions and obtain ignition by PAR. Test results indicate that under investigated test conditions, oxygen starvation at the PAR ($\Phi < 2.2$) avoids high catalyst temperature at excessively high H_2 concentrations and also impedes ignition near the ignition limit of the mixture. In experiments HR-29 and HR-30, ignition could only be achieved by artificially increasing O_2 concentration above 21 % of the $N_2 + Ar + O_2$ (=”air”) portion of the vessel atmosphere.

Based on THAI HR test series results, it could be concluded that ignition is directly correlated with the PAR catalyst surface temperature which in turn depends on the H_2 concentration present at the PAR inlet. The prerequisite for the PAR induced ignition vary with PAR design. Nevertheless, from the THAI experiments, a narrow range of the measured parameters at the time of ignition could be identified.

In the THAI PAR tests, ignition occurred in a rather quiescent atmosphere, containing O_2 surplus > 2- 3 (depending on the PAR design), and as soon as the conditions given below in the Table 2 were fulfilled:

Table 2: PAR-inlet conditions for the occurrence of an ignition

Gas atmosphere	**Measured catalyst surface temperature (maximum)**	**Measured H_2 concentration (minimum) at the PAR inlet**
Dry condition (without steam or superheated steam)	890 – 920 °C	5.5 - 7.5 vol % H_2 with 0 vol % steam (depending on the PAR design)
Wet conditions (condensing steam)	960 – 1005 °C	8 - 9 vol % H_2 with 45 vol % steam

6. HYDROGEN DEFLAGRATION BEHAVIOR FOLLOWING AN IGNITION BY PAR

Hydrogen concentration at the PAR inlet describes the load of the PAR in the moment of ignition but not necessarily the atmosphere above the PAR into which the hydrogen deflagration propagates. Hydrogen concentrations and flow conditions upstream and downstream of a PAR may differ significantly depending on an accident scenario, which in-turn may have an influence on PAR induced ignition behaviour. Furthermore, the course and strength of a hydrogen deflagration initiated by PAR ignition depends on several factors, e.g. gas distribution in the vessel above PAR outlet, atmospheric flow conditions, H_2 and inert gas concentration, geometry of the vessel internals.

In THAI tests, the released hydrogen at H = 1.26 m distributes by buoyancy within the vessel volume and a well-mixed vessel atmosphere develops. After onset of hydrogen recombination, PAR releases hot gas into the vessel dome and a temperature stratification of the vessel height gradually develops, which – due to continuous H_2 release into the lower vessel zone – finally leads to inverse hydrogen stratification. The inverse hydrogen stratification has two consequences: on the one hand the PAR takes its inflow at H = 2.1 m from the lower vessel zone with elevated H_2 concentration so that its recombination rate is higher compared to well-mixed H_2 distribution in the vessel. On the other hand in case of PAR ignition, deflagration develops into the hydrogen-lean vessel dome resulting in a pressure rise which is smaller than in case of well-mixed H_2 distribution. The latter effect is one reason for the relatively small deflagration pressures obtained in the THAI HR tests. Table 3 summarizes the measured deflagration pressures (P_0 = absolute vessel pressure prior to ignition, ΔP = pressure rise during deflagration) and the H_2 concentrations at PAR inlet and in the vessel dome immediately prior to ignition.

The effect of gas distribution on hydrogen deflagration induced pressure rise can be demonstrated by making a comparison between HR-6 and HR-7 tests. In HR-7 test, the measured deflagration pressure rise and the PAR inlet H_2 concentration immediately prior to ignition were 0.313 bar and 7.35 vol %, respectively. In HR-7 test, deflagration remained confined within the vessel dome. In another THAI test HR-6 with same initial test conditions as established for HR-7 test but consisted of the first two test phases only, ignition already occurred at the end of test phase-1. Due to the shorter PAR operation time, atmosphere stratification was less marked. The significantly higher H_2 concentration (7.1 vol % in HR-6 as compared to 5.7 vol % in HR-7) in the vessel dome in HR-6 resulted in a stronger deflagration than HR-7 and covered the entire vessel volume. In HR-6 test, the measured deflagration pressure rise and the PAR inlet H_2 concentration immediately prior to ignition were 1.28 bar and 7.5 vol %, respectively.

Similarly, in THAI "multi-ignition test" HR-4 the first two ignitions occurred rather early with an H_2 concentration in the dome area at the lower ignition limit (4 vol % H_2 for H_2-air mixtures at ambient temperature). Consequently, the deflagration was very smooth and the pressure rise very low. In contrast, the third ignition in HR-4 occurred at or near the PAR inlet. The flame proceeded first into the vessel bottom zone, and from there upwards into the inner cylinder and annulus zones, and finally into the dome zone. Due to the combustion in the bottom zone with relatively high local H_2 concentration (6.6 vol % H_2) and flame propagation in the entire vessel volume, the pressure increase following the third ignition was higher than after the previous burns.

The atmosphere stratification developing in the THAI vessel during PAR operation significantly affects the strength of combustion after ignition by the PAR. Therefore, the HR results on PAR-induced deflagration certainly help to improve understanding of those effects. However, pressure and temperature peaks produced by PAR induced deflagration are specific to the size of the THAI test facility. For modelling and assessment of deflagrations in homogeneously mixed or in pre-established stratified atmospheres, data are available from hydrogen deflagration tests series [8].

Table 3: PAR ignition data (selected HR tests performed in a quiescent atmosphere)

Test No./initial conditions		Pressure P_0 prior to ignition	H_2 concentration at PAR inlet	H_2 concentration in vessel dome	Pressure rise ΔP due to deflagration
HR-1 (1 bar, 25 °C, dry)		1.09 bar	6.55 vol%	4.7 vol%	0.18 bar
HR-3 (1.5 bar, 25 °C, dry)		1.51 bar	6.6 vol %	4.2 vol %	0.136 bar
HR-4 (2.2 bar, 25 °C, dry)	1st ignition	2.46 bar	5.95 vol %	4.0 vol %	0.065 bar
	2nd ignition	2.52 bar	6.25 bar	4.0 vol %	0.03 bar
	3rd ignition	2.56 bar	6.6 vol %	4.0 vol %	0.351 bar
HR-6 (1.5 bar, 74 °C, 25 % steam)		1.58 bar	7.5 vol %	7.1 vol %	1.28 bar
HR-7 (1.5 bar, 74 °C, 25 % steam)		1.512 bar	7.35 vol %	5.7 vol %	0.313 bar

7. POTENTIAL IMPACT TO REACTOR ACCIDENT SCENARIO

The required number of PARs (or active catalyst surface) and their locations inside containment are estimated based on most severe accident scenario specific to a particular reactor design. Global detonation or fast hydrogen deflagration which may occur if global hydrogen concentration exceeds by 13 vol % H_2 (dry mixture) is prevented by the fact that PARs intend to ignite already at 9 vol % H_2 concentration (wet mixture) the latest. The ignition limit for dry gas mixture is further reduced down to 7.5 vol % H_2 as demonstrated by the THAI tests. The THAI test data also indicate that for the PAR induced convective flows, hydrogen flame propagation occurs always in the upward direction commencing at PAR outlet.

Inlet hydrogen concentrations at which PAR tends to ignite are measured to be lower than the minimum H_2 concentration required for downward burn (8.7 vol % H_2 in dry mixture and 12 vol % H_2 in wet mixture) [8]. The THAI PAR ignition test data did not indicate any flame acceleration under investigated test conditions.

In the worst case scenario, the potential of flame acceleration or detonation due to PAR induced ignition will need to satisfy a number of conditions, e.g. size of vessel, geometry of vessel internals, gas composition available in the neighbourhood of a PAR, etc. The test conditions in THAI were limited to exclude any flame acceleration or detonation. Flame acceleration can occur due to turbulence, geometry, presence of obstacles, and wall roughness. The multi-compartment H_2 deflagration experiments in 640 m^3 BMC facility [10] demonstrated that flame acceleration is also possible at H_2 concentration as low as 9-10 vol % depending on initial pressure, steam concentration, and geometry of compartments. The flame acceleration (without any symptoms of detonation) occurred due to jet ignition effect produced by prevalence of specific distribution of hydrogen and geometry of the vent in the connected compartments. The presence of an additional component in the hydrogen-air gas mixture may further influence the detonability range. In a Committee on the Safety of Nuclear Installations report [11], it is reported that an addition of CO to hydrogen-air mixtures increases the detonation sensitivity for a particular hydrogen concentration. At room temperature, a hydrogen-air mixture containing 10 vol % H_2 will not detonate, but an addition of 5 vol % CO will convert this hydrogen-air mixture into a detonable mixture. Therefore, it is of utmost importance to consider potential gas components that can be present in the gas while analyzing hydrogen combustion behavior and the associated safety measures.

CONCLUSIONS

In the present paper, experimental finding related to PAR induced ignition behaviour and the resulting H_2 deflagration behaviour as a function of thermal-hydraulic conditions and gas-distribution are discussed. Experiments provide a valuable database both for hydrogen control concept demonstration and code validation purposes by investigating three different commercially available PAR units subjected to identical severe accident representative conditions.

The large THAI vessel allows PAR operation with unrestricted natural convection which includes interaction of PAR performance and vessel atmosphere distribution. Test results indicate that ignition occurs as soon as critical levels of hydrogen concentration at the PAR inlet or catalyst temperature, respectively, have been reached. The hydrogen concentration threshold (mainly for the plate-type PARs) at which the catalytic recombiner could become an ignition source has been determined between 5.5 vol% to 9 vol% depending on the recombiner design and presence of steam.

The test data indicate that for the PAR induced convective flows, hydrogen flame propagation occurs always in the upward direction commencing at PAR outlet. Inlet hydrogen concentrations at which PAR tends to ignite are lower than the minimum H_2 concentration required for downward burn. The course and strength of a hydrogen deflagration initiated by PAR ignition depends on the available gas composition nearby the PAR outlet as well as gas distribution in the vessel. Under investigated test conditions in the THAI vessel, negative hydrogen concentration gradients develops, which prevent flame propagation.

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