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AMHYCO engineering correlation to describe the conversion of flammable gases in Framatome passive autocatalytic recombiners

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ABSTRACT

As lessons learned from the Three Mile Island 2 and Fukushima Daiichi accidents, the mitigation of severe accidents in nuclear power plants by dedicated systems and back-fittings as well as operational procedures became internationally accepted standards. Concerning the risk of in-containment hydrogen combustion, many plants worldwide installed passive autocatalytic recombiners (PARs). To quantify the impact of these PARs on the containment atmosphere during a core damage accident, their performance is usually described by simplified engineering correlations which can be implemented in nuclear containment simulation codes (e.g., GOTHIC, MELCOR, ASTEC, COCOSYS). This paper presents a state-of-the-art upgrade of the engineering correlation for PARs with plate-type catalysts, which was developed in the EU AMHYCO project. The aim of rephrasing the existing correlation was to extend the range of validity of the PAR correlation to gas mixtures encountered in the late accident phase, i.e., to atmospheres with carbon monoxide content and low oxygen concentrations. Further phenomena reconsidered are recombiner start-up and termination of operation, poisoning by carbon monoxide and recombiner-induced ignition. The work presented here is based on the generic plate-type PAR correlation developed within the EU AMHYCO project Work Package 3. In the following, this generic correlation is adapted to the specific PAR technology of the nuclear vendor Framatome and validated on experiments performed in the frame of the OECD/NEA-THAI programs.

1. Introduction

The environmental impact of a severe accident in a nuclear power plant depends predominantly on the integrity of the containment, acting as last technical-airtight barrier. However, the containments of Generation II nuclear power plants are not designed to withstand the loads of a severe accident. Already the Wash1400 report (NRC, 1975) considers the release of hydrogen by core oxidation, and the possibility of a subsequent combustion event challenging the containment integrity. However, at that time, the international consensus of nuclear regulators was that the probability for a core melt event was sufficiently small, not justifying any mitigating actions.

On March 28th 1979, a partial core melt occurred in the pressurized water reactor (PWR) Three Mile Island reactor unit 2. The core oxidation is estimated to have generated ~450 kg hydrogen (Henrie and Postma, 1983), which was released via the pressurizer and the pressurizer relief tank into the containment. About 10 h after start of the accident, the hydrogen/air mixture ignited within the containment. In the specific accident progression in Three Mile Island with only a partial core melt, the availability of containment heat removal keeping the overall containment pressure low, and the absence of a deflagration-to-detonation transition, the hydrogen combustion did not threaten the containment integrity. Nevertheless, in less favourable accident sequences, a combustion may well have the potential to cause a containment failure (NEA, 2014).

The occurrence in Three Mile Island initiated a large-scale international research program about the risk of combustible gases in nuclear power plants (NEA, 1999; NRC, 1983) and led to a strengthening of regulatory requirements. In October 1978, the U.S. NRC issued rule 10CFR50.44, requesting the installation of a combustible gas control system in all plants, capable to mitigate a 5 % core oxidation at design base accident conditions (NRC, 1978, 2007). Nowadays, Generation II nuclear power plants have by design an operational combustible gas control system. However, these systems (relying on electric power and thermal recombination) usually have neither the capacity to consume the volume of hydrogen released by a core melt accident, nor are they qualified against the harsh conditions occurring during a severe

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Nomenclature

c	[-] model parameter
D	[m^2/s] diffusion coefficient
E	[J] energy
k_1, k_2	[g/(s bar)], [g/s] PAR-type specific model parameters
M	[g/mol] molar mass
$\dot{m}$	[g/s] mass conversion rate
P	[W] heat release rate
p	[bar] absolute pressure
Q	[J/kg] combustion energy per unit of mass
T	[°C], [K] temperature
t	[s] time
x	[vol.%] gas concentration

Greek letters

X	[-] scaling factor
γ	[vol.%] weighted gas concentration
η	[vol.%] volumetric gas concentration

Subscripts

cat	catalyst
CO	carbon monoxide
eff	effective
H_2	hydrogen
in	inlet
low	low gas concentration
max	maximum
min	minimum
O_2	oxygen
poison	catalyst poisoning
start	PAR start-up

accident.

With time, the U.S. NRC further elevated its requirements. In 1981, boiling water reactor (BWR) containments employing a pressure suppression system (Mark I and Mark II) had to inert the containments with nitrogen (NRC, 2003). In 1985, containments vulnerable against hydrogen combustion events (BWR Mark III containments and PWR ice condenser containments) had to install hydrogen control systems capable to mitigate a core oxidation of 75 % (NRC, 2007; NRC, 2003). For all plants licensed after 2003, 10CFR50.44 requests the capability to withstand the oxidation of 100 % of the core (NRC, 2007; NRC, 2003) – however, older PWR with large dry containments are not required to have such systems.

Western Europe initially followed the U.S. NRC regulations, i.e., the inertisation of BWR containments and the installation of an active combustible gas control system for design base accidents. Nevertheless, after the Chernobyl accident in 1986, some European regulators further elevated their requirements concerning hydrogen mitigation. For example, in Germany, it became de-facto mandatory in 1997 to install passive autocatalytic recombiners (PARs) in all PWR (RSK, 1997; Sonnenkalb and Mertins, 2011), independent of their age. The PAR system had to have the capacity to cope with the hydrogen released in a severe accident on a best-estimate basis. This included the in-vessel core oxidation phase as well as a possible release of combustible gases by the molten corium-concrete interaction (MCCI) after failure of the reactor pressure vessel (RPV). For the 1300 MW$_e$ German PWR, the reference PAR system design had a recombination capacity of 200 kg hydrogen per hour at a concentration of 4 vol% and 3 bar-abs (RSK, 2015).

In the 1990s and 2000s PAR-based hydrogen mitigation systems were subsequently introduced into all PWRs with large dry containments in Belgium, France, Netherlands, Russia, Sweden, South Africa, China, and South Korea.

On March 11th 2011, the Fukushima Daiichi nuclear accident occurred. After this accident, additional countries started deviating from U.S. NRC rulings and requested a severe accident-dedicated hydrogen mitigation system for their Generation II PWRs. These countries included Spain (CSN, 2012), Switzerland (ENSI, 2015), and the United Kingdom (ONR, 2017). Even plants already equipped with active igniters experienced an increasing regulatory pressure to further strengthen their hydrogen mitigation capabilities, independent of electric power, e.g., the Spanish Mark III BWR Cofrentes (CSN, 2012).

Nowadays, a severe-accident dedicated hydrogen mitigation system based on passive autocatalytic recombiners represents the state-of-the-art hydrogen safety precaution in PWR and non-inertised BWR (NEA, 2014). In this paper, an improved engineering correlation for Framatome PARs is presented, in the following labelled as ‘AMHYCO Correlation for Framatome PAR’. This correlation shall be easy to integrate into containment simulation codes, the correlation shall be numerically stable, and shall not elongate the computation time of a containment simulation. Therefore, a simplified approximative solution is preferred over a complex more physically accurate description which is offered by mechanistic codes such as REKO-DIREKT (Reinecke et al., 2016), SPARK (Meynet et al., 2014), or PARUPM (Domínguez-Bugarín et al., 2024).

The AMHYCO correlation presented in this paper is adapted specifically for the Framatome PAR design. The nuclear supplier Framatome openly cooperates with national and international research organizations for decades to test and confirm the reliability and effectiveness of its PAR design and catalyst technology. Therefore, a broad experimental database is available to validate the correlation presented in this paper to high nuclear quality standards. The authors do not make any claim about the performance and operation of other PAR designs or catalyst materials from other vendors.

This work focuses on the performance of PARs under boundary conditions typical for the conditions in a light water reactor containment during a severe accident. In the early phase of a severe accident the fuel cladding oxidizes, releasing hydrogen from the reactor coolant system into a reasonably oxygen-rich possibly steam-interted containment atmosphere. In the late accident phase, after RPV failure, the oxygen-levels inside the containment are reduced, while a molten corium-concrete interaction may release large amounts of hydrogen and carbon monoxide (CO). Latter is known to act as catalyst poisoning by persistently adsorbing on the catalyst’s active sites. The aim of rephrasing the original AREVA PAR correlation within the AMHYCO project was to extend the range of validity of the PAR correlation especially to gas mixtures with high carbon monoxide content and low oxygen concentrations. Further, new previously not considered phenomena like PAR poisoning and PAR-induced ignition are incorporated. The deduced PAR correlation may not be used for atypical PAR applications, for example in comparatively colder waste storage facilities, without additional verification and validation efforts.

The subsequent chapters discuss the formulation and the physical interpretation of the engineering correlation. Additional information concerning the implementation of the PAR correlation into a containment code can be found in the Deliverable D5.1 of the EU AMHYCO (‘Towards an enhanced accident management of the hydrogen/carbon monoxide combustion risk’) project (AMHYCO, 2025).

2. Hydrogen mitigation by PARs

After the accident in Chernobyl in 1986, Framatome, as successor of

Siemens Kraftwerk Union / AREVA, invented the PAR technology (FRA, 1996) and developed it into a nuclear qualified safety system. In the following Chapter 2.1, the physical PAR component is present, and Chapter 2.2 discusses the overall operation principle of a Framatome PAR.

2.1. Passive autocatalytic recombiner component

In a Framatome PAR, numerous thin stainless-steel sheets (FRA, 2024a) are coated with a porous layer of catalytically active noble metals. The coated steel sheets are arranged vertically in a drawer at the bottom end of a larger metal housing, see Fig. 1. This drawer ensures the quick access to the catalytic sheets, and the planar geometry of the sheets allows for convenient visual inspection or collecting samples for periodic testing. The PAR housing walls protect the catalytic sheets from any mechanical damage, and the top cover protects the catalyst from direct exposure to spraying of water and aerosol deposition. Due to the gas inlet at the bottom and a lateral gas outlet at the top of the housing, a chimney effect inside the housing promotes natural draft through the PAR.

Framatome PARs have performed reliably in international test programs like in the THAI facility (THAI, 2010; THAI, 2023) or in the PHEBUS experiments (Eckardt and Losch, 2010). Due to the proven high reliability of Framatome PAR in their extensive qualification and test program (FRA, 2012), the availability of Framatome PARs during a nuclear incident or accident is broadly accepted by regulatory institutions. More than 150 nuclear power plants worldwide installed Framatome PAR (FRA, 2024a).

Commercial Framatome PARs are available in different sizes and capacities, depending on the available installation space and the locally required recombination capacity. Table 1 lists properties of the most common PAR sizes (FRA, 2024a; Kim et al., 2019) as well as known values for the prototypical Framatome-like PAR used in the THAI experimental facility (Freitag et al., 2022). The PAR-size describing constants k_1 and k_2 will be used later in Chapter 3.

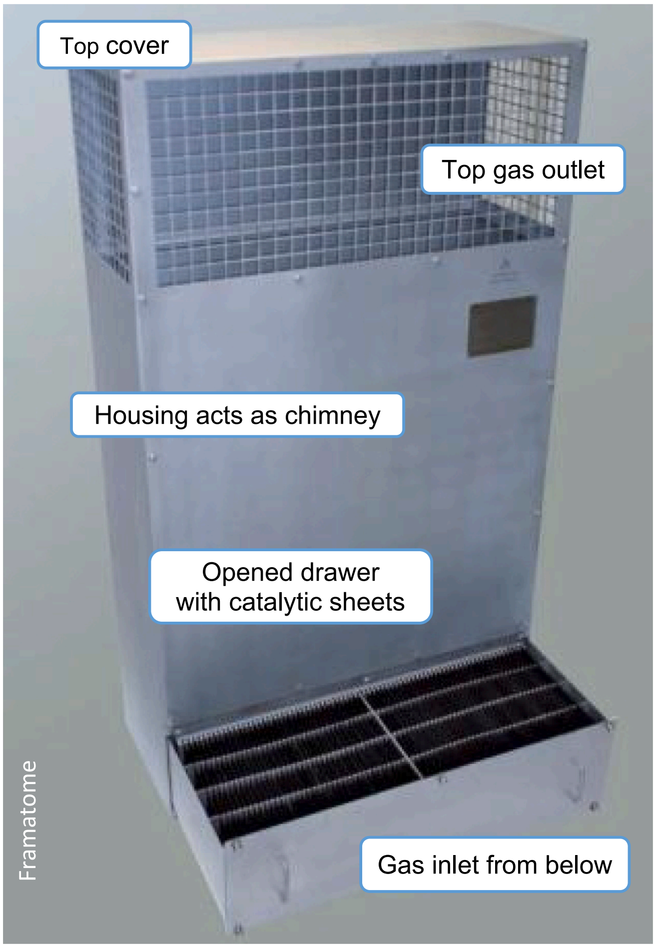

Fig. 1. Photo of a Framatome passive autocatalytic recombiner.

2.2. Overview of the PAR operation

PARs are considered to operate autocatalytic. The recombination of combustible gases is an exothermic process, releasing heat. This leads to a positive feedback loop as the chemical recombination on the catalytic surfaces is accelerated by higher temperatures. On an initially cold catalytic sheet exposed to low concentrations of combustible gases, only a rather low recombination rate is achieved (considered zero for numerical purposes). The PAR is considered to be in its inactive state. With increasing concentrations of combustible gases, the chemical heat release starts to overcome the heat losses of the PAR, and the PAR catalytic sheets suddenly start heating up (so-called light-off). At this point, also the rate of gas recombination suddenly strongly increases, reaching the specified recombination rates as listed in Table 1. Then, the PAR is considered to be in its active state. To facilitate the light-off of this autocatalytic process, the Framatome PAR design minimizes the thermal capacity of the catalyst carrier (by thin metal sheets), minimizes the heat losses of the sheets (by arranging them in parallel stacks), and prevents the ingression of water droplets from the top into the PAR, which could cool down the sheets.

In general terms, the behaviour of a PAR can be illustrated by the catalyst temperature as a function of an effective combustible gas concentration, see Fig. 2. The definition of the effective gas concentration is stated in Chapter 3.3. Initially, the catalyst is cold, i.e., it has a temperature equal to the environmental temperature. At a certain effective gas concentration, the PAR switches from its inactive state to the autocatalytic recombination active state, see Chapter 3.6. After the light-off of the catalyst, the PAR operates the better the higher the effective gas concentration becomes (compare the discussion in Chapter 3.3). At very high concentrations, the PAR and especially the PAR exhaust gases may become so hot, that they may ignite the surrounding atmosphere (see Chapter 3.7). With time, the combustible gases and/or oxygen get consumed in the containment, and the effective gas concentration decreases. With that decrease, also the PAR catalyst temperature will decrease. At a certain point in time, the PAR will switch from its autocatalytic active state back into its inactive state (see Chapter 3.4) due to a lack of combustible gases or oxygen. Alternatively, in presence of a catalyst poison like carbon monoxide, the PAR may cool-down even at a high effective gas concentration due to catalyst poisoning (see Chapter 3.5).

Table 1
Standard Framatome PAR sizes and the Framatome-like PAR in the THAI facility, their specified hydrogen depletion rates, and the constants describing the PAR operation.

PAR type	Size (width × depth)[mm × mm]	Sheets	k_1[g/(s·bar)]	k_2[g/s]	H_2 depletion @ 1.0 bar [kg/d]	
					at 3 vol% H_2	at 6 vol% H_2
FR90/1-1500T	1550 × 326	150	0.137	0.167	79	157
FR90/1-750T	800 × 326	75	0.061	0.074	35	70
FR90/1-380T	430 × 326	38	0.031	0.037	18	35
PAR in THAI	205 × 320	19	0.0137	0.016	–	–

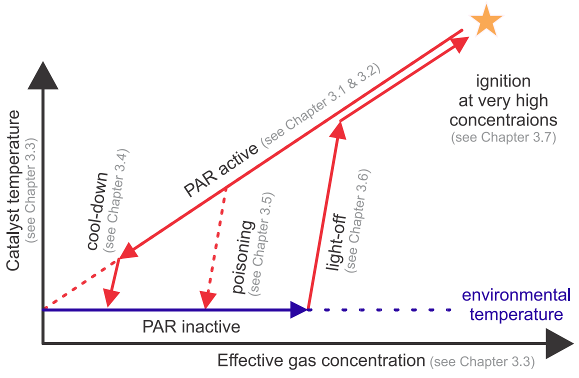

Fig. 2. Illustration of the operation of a PAR.

3. AMHYCO correlation for Framatome PARs

With progress made in the SAMHYCO_NET project (Reinecke et al., 2022) and the following AMHYCO project (Reinecke et al., 2023), a new engineering correlation approach was developed by the Research Centre Jülich, incorporating the experimental insights of the operation of recombiners with plate-type catalysts in recent years. This chapter presents the adaption of this new correlation approach specifically to Framatome PARs. The resulting correlation, referred to as'AMHYCO correlation for Framatome PAR‘, shall enhance the description of the Framatome PAR performance especially in gas mixtures containing hydrogen as well as carbon monoxide which may originate from molten corium-concrete interaction.

3.1. Hydrogen recombination rate

The original AREVA correlation to describe the in-house PARs considers a linear dependency of the recombination rate on pressure (Reinecke et al., 2022). Maintaining this overall functional form of the correlation but including the insights into PAR operation gained in AMHYCO WP3.2 (Reinecke et al., 2023), the hydrogen recombination rate (in units of grams per second) is reformulated as

$$\dot{m}_{H2}\left[\frac{g}{s}\right] = (k_1 \bullet p + k_2) \bullet \eta_{H2} \bullet c_{low} \bullet c_{poison} \bullet c_{start} \tag{1}$$

$$\eta_{H2} = \begin{cases} \nu_{H2} & \text{if } (0.84\nu_{O2} \geq \nu_{H2} + 0.414\nu_{CO}) \\ 0.84\nu_{O2} - \min(0.414\nu_{CO}, 0.84\nu_{O2}) & \text{if } (0.84\nu_{O2} < \nu_{H2} + 0.414\nu_{CO}) \end{cases}. \tag{2}$$

with the PAR-type depending constants k_1 and k_2 (see Table 1), the absolute pressure p, the weighted hydrogen gas concentration η_{H2}, the PAR shut-down parameter c_{low} at low gas concentrations (see Chapter 3.4), the PAR poisoning parameter c_{poison} in the presence of carbon monoxide (see Chapter 3.5), and the PAR start-up parameter c_{start} (see Chapter 3.6), all depending on the volumetric gas concentrations (with the unit vol.%) of oxygen ν_{O2}, hydrogen ν_{H2}, and carbon monoxide ν_{CO}. Note, in this paper, gas concentrations always refer to the physical concentration (i.e. wet gas concentrations) in the local atmosphere, including steam, not the often experimentally measured dry gas concentration. Further, the correlation does depend only implicitly on the presence of steam via the definition of the wet gas concentrations and the ambient pressure.

The mathematical expressions in Eqs. (1) and (2) are based on two assumptions.

The first assumption is that the gas diffusion from the bulk gas to the catalytic sheet surfaces limit the overall PAR recombination rate. The numerical constants in the conditions in Eq. (2) are determined by the gas diffusivities D_x and the stoichiometric rations of oxygen and carbon monoxide with respect to hydrogen (Reinecke et al., 2023), i.e., $0.84 \approx 2 \bullet (D_{O2}/D_{H2})^{2/3}$ and $0.414 \approx (D_{CO}/D_{O2})^{2/3}/(D_{H2}/D_{O2})^{2/3}$. As long as the (stoichiometric-weighted) diffusion rate of oxygen to the catalytic surface is larger than the combined diffusion rate of hydrogen and carbon monoxide, i.e., $(0.84\nu_{O2} \geq \nu_{H2} + 0.414\nu_{CO})$, the availability of hydrogen limits the recombination rate for hydrogen. Thus, the hydrogen recombination rate becomes proportional to ν_{H2}.

The second assumption is that if there is not sufficient oxygen available to oxidize the diffusion inflow of hydrogen and carbon monoxide onto the catalytic sheets simultaneously $(0.84\nu_{O2} \leq \nu_{H2} + 0.414\nu_{CO})$, then carbon monoxide is oxidized first, and hydrogen is subsequently oxidized only if some oxygen is left over (Klauck et al., 2021). With that assumption, the hydrogen recombination rate becomes proportional to the oxygen diffusion rate minus the carbon monoxide diffusion rate.

In the remaining paper, the notation of oxygen-starvation‘ refers to the condition $(0.84\nu_{O2} < \nu_{H2} + 0.414\nu_{CO})$ as this is the relevant condition for PAR operation. The opposite of oxygen-starvation is labelled as'oxygen-surplus’.

In the limiting case of oxygen-surplus and no carbon monoxide concentration, i.e., in conditions typically encountered in the containment of a nuclear power plant during the early phase of a severe accident, Eq. (1) becomes equivalent to the original AREVA correlation (Reinecke et al., 2010). Latter correlation was successfully validated in numerous Framatome-internal as well as national and international tests.

The assumption in Eq. (2) that carbon monoxide is oxidized always first is only approximately true for a pure platinum catalyst. A palladium catalyst, for example, seems to favour hydrogen over carbon monoxide especially in a low-oxygen environment (Klauck et al., 2021). Commercial Framatome PARs (FRA, 2024a) installed in nuclear power plants employ different noble metals and various admixture elements, to make the catalyst as resistant as possible against the stopping of one of the catalytic reaction paths. Therefore, the assumption of oxidizing carbon monoxide always first may be less valid for Framatome PAR. Nevertheless, a comparison to available experimental data obtained with real Framatome catalytic sheets show that the constructed correlation is still in good agreement (FRA, 2024b).

3.2. Carbon monoxide recombination rate

The carbon monoxide recombination rate (in units of grams per second) can be deduced in analogy to Eqs. (1) and (2), depending on the absolute pressure p, the PAR-type specific constants k_1 and k_2 (see Table 1), the PAR shut-down parameter c_{low} (see Chapter 3.3), the poisoning parameter c_{poison} (see Chapter 3.5), the PAR start-up parameter c_{start} (see Chapter 3.6), and the weighted carbon monoxide concentration η_{CO}:

$$\dot{m}_{CO}\left[\frac{g}{s}\right] = \frac{M_{CO}}{M_{H2}} \bullet (k_1 \bullet p + k_2) \bullet \eta_{CO} \bullet c_{low} \bullet c_{poison} \bullet c_{start} \tag{3}$$

$$\eta_{CO} = \begin{cases} 0.414\,\nu_{CO} & \text{if } (0.84\,\nu_{O2} \geq \nu_{H2} + 0.414\,\nu_{CO}) \\ \min(0.414\,\nu_{CO}, 0.84\,\nu_{O2}) & \text{if } (0.84\,\nu_{O2} < \nu_{H2} + 0.414\,\nu_{CO}) \end{cases}. \tag{4}$$

The molecular mass fraction M_{CO} / M_{H2} can be well-approximated to 14. As it is assumed that carbon monoxide is always oxidized first, its recombination rate depends on the concentration of carbon monoxide if oxygen is in surplus or is limited by the availability of oxygen in case of starvation condition.

3.3. Effective gas concentration and maximum catalyst temperature

In this chapter, the PAR correlation in the previous chapters is used to make a statement about the temperature of the catalyst inside an active Framatome PAR. While this represents a slight detour, the insights gained thereby make the subsequent treatment of PAR shutdown, poisoning, and PAR-induced ignition more transparent and bridges the gap to the PAR operating interpretation as shown in Fig. 2.

In presence of combustible gases and oxygen, the PAR rapidly switches from its inactive state (with low chemical activity, considered zero in containment simulations) into its active state (offering the specified hydrogen and carbon monoxide recombination rate). In stationary condition, the chemical heat release rate P in the PAR is proportional to the gas mass conversion rates $\dot{m}_{H2}$ and $\dot{m}_{CO}$ times the respective combustion energies Q_{H2} and Q_{CO} per unit of mass

$$P = Q_{H2} \bullet \dot{m}_{H2} + Q_{CO} \bullet \dot{m}_{CO}. \tag{5}$$

With the simplifying assumptions that the molar heat of combustion of hydrogen ($Q_{H2} \bullet M_{H2}$ 240 kJ/mol) and carbon monoxide ($Q_{CO} \bullet M_{CO}$ 280 kJ/mol) is similar, based on Eqs. (2) and (4), the heat release power P becomes proportional to a function of gas concentrations, which is referred to as 'effective gas concentration' x_{eff} from here on forward

$$P \propto x_{eff} = \min(\nu_{H2} + 0.414\,\nu_{CO}, 0.84\,\nu_{O2}). \tag{6}$$

Due to the chemical power release, the catalytic sheets heat up until their heat loss to the environment equalizes the chemical power release. If we assume, that heat radiation losses can be neglected due to the geometrical arrangement of the catalytic sheets, see Fig. 1, the heat loss of the catalytic sheets should be about linear in temperature (at least at low temperatures), and thus, the catalyst temperature should depend linearly on the effective gas concentration $x_{eff} = \min(\nu_{H2} + 0.414\,\nu_{CO}, 0.84\,\nu_{O2})$.

Such a linear dependency of the catalyst temperature on gas concentrations was confirmed by the THAI experiments as well as observed in small-scale experiments in the REKO facility in the frame of the AMHYCO project (Reinecke et al., 2023), see Fig. 3. At given hydrogen concentrations (1 vol%, 2 vol%, 4 vol%), the oxygen concentration is varied. In oxygen-starvation, when oxygen limits the PAR recombination rate, the increase in oxygen leads to a nearly linear increase of the catalyst temperature. In oxygen-surplus, hydrogen limits the PAR operation. Thus, the catalyst temperature becomes independent of the oxygen concentration.

Based on THAI experiments using original Framatome PAR sheets (FRA, 2024b), the following correlation can be deduced, linking the maximum catalytic sheet temperature $T_{cat,max}$ to the PAR gas inlet temperature T_{in} and the effective combustible gas concentration x_{eff}:

$$T_{cat,max} = T_{in} + \frac{(160 \pm 20)\,\mathrm{K}}{1\ \mathrm{vol\%}} \bullet x_{eff} \tag{7}$$

As the catalyst temperature depends on the PAR catalyst activity and the PAR design, Eq. (7) is valid only for Framatome PAR. Note that Eq. (7) refers to the local peak temperature observed within the PAR in the THAI experiments, not the average catalyst temperature. This correlation was validated on numerous THAI experiments. As example, in Fig. 4 (left), the measured maximal catalyst temperature (T_{max}, blue), the minimal catalyst temperature (T_{min}, red) and the gas temperature flowing into the PAR (T_{in}, turquoise), as measured in the experiment HR-53, is shown as function of the effective gas concentration x_{eff} over the entire duration of the experiment, containing two injection and depletion cycles.

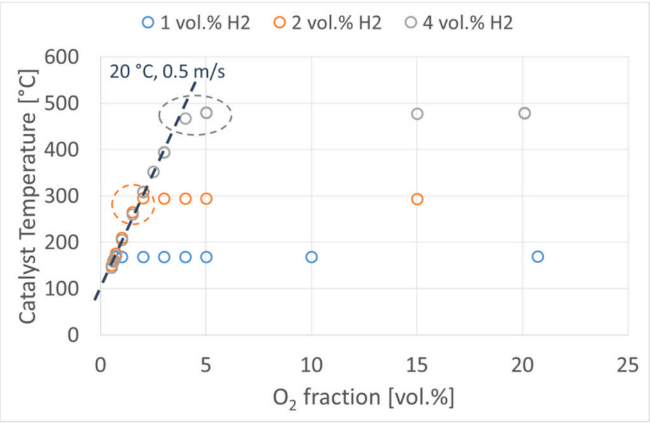

Fig. 3. Measured catalyst temperature for different mixtures of hydrogen, oxygen and nitrogen in the REKO-3 facility.

As long as the PAR catalytic sheet temperature fulfils the correlation in Eq. (7), the PAR is considered to be in its active state, compare Chapter 2.2. A significant deviation from the correlation in Eq. (7) allows for a clear identification of the termination of PAR operation at low gas concentrations (see Chapter 3.4), a delayed PAR start-up (see Chapter 3.6), or poisoning of the PAR by, e.g., carbon monoxide (see Chapter 3.5). As example, in Fig. 4 (right) the minimum catalyst temperature T_{min} suddenly departs from its somewhat linear dependency on x_{eff} at about 380 min, approaching ambient temperature. This can be attributed to a poisoning of the catalytic surface by carbon monoxide. As the maximum catalyst temperatures T_{max} still follow the correlation in Eq. (7), it can be concluded that the catalyst deactivation is only local/partial, while other surface regions of the PAR still remain active.

3.4. *Termination of PAR operation at low gas concentrations*

When a PAR is in its autocatalytic active state, it is hard to return to its inactive state as long as even traces of combustible gases are present. However, at very low effective gas concentrations, two phenomena must be considered. First, an elevated PAR temperature may not directly translate to a noticeable hydrogen recombination. And second, the lower the chemical recombination and thus the driven natural draught through the PAR, the more fragile the system becomes against external perturbations. It may be possible that the upward draft of the gases through the PAR housing may not be able to overcome an overall downward air movement within the PAR installation room or an overall turbulent gas movement in the atmosphere.

For the reasons stated above, a lower cut-off of the recombination rates is described by the multiplicative shut-down parameter c_{low} in Eqs. (1) and (3). This parameter shall have a functional form to be equal to one at reasonable high gas concentrations, become zero at the shut-down limits, and remain zero below these limits. Further, it shall depend only on the condition which limits the PAR operation, i.e., either the inflow of oxygen or the inflow of combustible gases. These criteria are realized by the phenomenological term

$$c_{low} = \max\left(\sqrt{\tanh\left[x_{eff} - 0.3\ \mathrm{vol.\%}\right]}\ , 0\right). \tag{8}$$

As shut-down limit, an effective gas concentration of 0.3 vol% is chosen, which reflects the lower limit observed in relevant experiments in the less favourable counter-current flow situation (FRA, 2024b). This choice is somewhat conservative as only the PAR in the outer regions of a containment will experience a downward convective gas flow, acting against a convection through a PAR. The PAR close to the centre axis of a nuclear containment will likely experience an upward draft, which would support the convective gas flow through the PAR housing even at a low effective gas concentration. Note that the exact value of the lower cut-off is likely only of minor importance to full containment simulations as at such low gas concentrations, the PAR recombination rates become very small anyway.

The $\tanh[\ldots]^{0.5}$ function in Eq. (8) ensures that at high concentrations the term becomes equal one, where the PAR operation is diffusion-limited, and that the term continuously decreases to zero at reaching the shut-down gas concentrations. Thereafter, the max-function ensures that the term remains zero for an effective gas concentration below the shut-

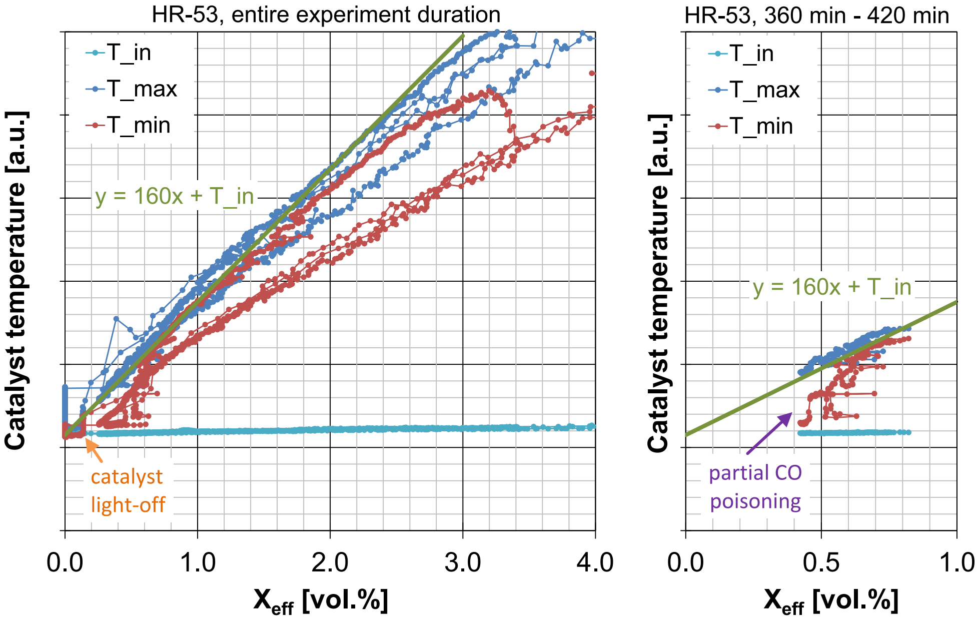

Fig. 4. Measured catalyst temperature in the THAI HR-53 experiment as function of the effective gas concentration.

down limit. Eq. (8) is based on functional considerations only. There is no physical justification for the usage of the specific mathematical functions. The suitability of the proposed phenomenological term for the shut-down parameter c_{low} is based solely on the comparison to experiments.

3.5. Poisoning by carbon monoxide

Carbon monoxide does adhere stronger to the catalytic surface than hydrogen. Thus, in certain situations the active centres of the catalytic surface can be occupied by carbon monoxide molecules, i.e., the PAR gets poisoned and does not offer its specified gas recombination capacity anymore.

This PAR poisoning by carbon monoxide was evaluated in the THAI experimental series as well as in REKO experiments in the frame of the AMHYCO project Work Package (WP) 3 (Reinecke et al., 2023). Based on the REKO experiments, it is proposed that the start of PAR poisoning can be attributed to the catalyst temperature falling below a certain threshold value, see Fig. 5. When the catalyst temperature drops below that ‘poisoning temperature’, the carbon monoxide molecules start accumulating on the PAR surface, blocking it. In the REKO facility it was determined that the poisoning temperature predominantly is a function of the carbon monoxide concentration and impacted by the environmental temperature for a generic palladium catalyst while being rather independent of the environmental temperature for a generic platinum catalyst. Palladium is more resistant to poisoning than platinum, thus, the poisoning temperatures for palladium are overall lower than for platinum, as can be seen in Fig. 5.

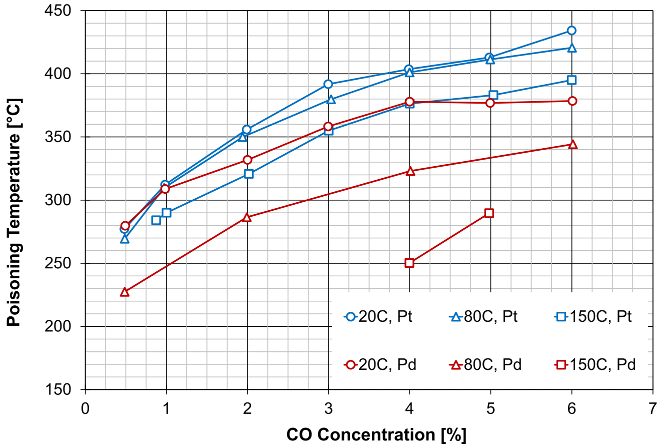

Fig. 5. Poisoning temperatures for Pt- and Pd-based catalysts (REKO-1 experiments).

Also in the recent THAI experiments, the impact of carbon monoxide was evaluated using real Framatome catalytic sheets. These THAI experiments show an overall similar poisoning behaviour as for the generic palladium catalyst in the REKO-1 facility. Based on the available experimental database (FRA, 2024b; Reinecke et al., 2023), the following conclusions can currently be made concerning the poisoning behaviour of Framatome PARs:

- For a poisoning of the PAR, the atmosphere must contain a non-negligible content of carbon monoxide. For the current state, a lower cut-off carbon monoxide concentration of 0.1 vol% is proposed.
- When exposing a Framatome PAR to hydrogen and carbon monoxide, the poisoning is a gradual process, first affecting the catalyst only locally (compare Fig. 4). As the poisoning mechanism depends on the absorption/desorption behaviour of carbon monoxide, the effect is reversible, and the catalyst can recover from its poisoned state in case the inlet gas composition leads to higher catalyst temperatures.
- Based on an evaluation of the THAI experiments, it seems that poisoning of the PAR occurs predominantly in oxygen-starvation condition $(0.84\,\nu_{\mathrm{O2}} < \nu_{\mathrm{H2}} + 0.414\,\nu_{\mathrm{CO}})$ when the catalyst temperature drops to $\sim 260\,^{\circ}\mathrm{C}$ (FRA, 2024b). Latter temperature criterion must be seen as a simplified zeroth-order approximation. The REKO experiments using generic catalysts indicate that the poisoning temperature is a function of gas concentration, environmental temperature, and affected by the presence of humidity (Reinecke et al., 2023). The description of such dependencies of the poisoning temperature on other physical parameters are, however, beyond the scope of this work and still subject of on-going research.
- Poisoning of the PAR in oxygen-surplus condition $(0.84\,\nu_{\mathrm{O2}} \geq \nu_{\mathrm{H2}} + 0.414\,\nu_{\mathrm{CO}})$ seems much harder to realize, requiring catalyst temperatures as low as about $140\,^{\circ}\mathrm{C}$ (FRA, 2024b) in the THAI experiments. For that zeroth-order approximative temperature criterion, the same limitations apply as for the temperature criterion for the oxygen-

starvation condition. In accident simulations with ambient temperatures of ~100 °C, a catalyst temperature of 140 °C would allow only such a low effective gas concentration that in most cases the shutdown criterion (compare Chapter 3.4) would act before the poisoning criterion. Thus, it may be debateable if there is a need for a poisoning criterion in oxygen-surplus at all.

Based on these observations and using Eq. (7), the following phenomenological poisoning parameter c_{poison} can be constructed:

$$c_{\text{poison}} = \begin{cases} 1.0 \text{ if} (\nu_{\text{CO}} < 0.1\%) \\ 1.0 \text{ if} x_{\text{eff}} > \dfrac{1\%}{160\,\text{K}} \bullet (140^{\circ}\text{C} - T_{\text{in}}) \text{ and} (0.84\nu_{\text{O2}} \geq \nu_{\text{H2}} + 0.414\nu_{\text{CO}}) \\ 1.0 \text{ if} x_{\text{eff}} > \dfrac{1\%}{160\,\text{K}} \bullet (260^{\circ}\text{C} - T_{\text{in}}) \text{ and} (0.84\nu_{\text{O2}} < \nu_{\text{H2}} + 0.414\nu_{\text{CO}}) \\ 0.0 \text{ otherwise} \end{cases} \tag{9}$$

The logical criteria in Eq. (9) are assumed to be evaluated from top to bottom until the first conditional “yes” is reached. Note that Eq. (9) does not compare the current catalyst temperature to a critical temperature, but the temperature the catalyst would have after light-off.

Overall, the situations where the poisoning of a Framatome PAR could occur can be considered as not relevant concerning nuclear safety. Poisoning occurs only at very low oxygen levels <3.0 vol% (Reinecke et al., 2024). However, when the concentration of oxygen or of the combustible gases within the containment of a nuclear power plant drops to such low values, there is no combustion risk anyway. If for scenario-related circumstances the oxygen concentration would locally rise again, the Framatome PARs would recover from their poisoned state, as shown in experiments.

The determination of the poisoning temperature in the THAI experiments and the REKO facility gave slightly different results. The THAI experiments on one hand use a prototypical PAR with original catalytic sheets and the experiments are rather transient, resulting in a significant scattering of the observed poisoning temperatures. On the other hand, the REKO facility operates in forced-flow steady state condition and uses generic catalyst samples. This may explain the difference of observed poisoning temperatures of ~260 °C in THAI to ~300 °C in REKO, see Fig. 5.

The poisoning temperatures identified are subject of significant uncertainties. Beside the differences encountered in THAI and REKO, it must also be pointed out that the poisoning temperature in oxygen-surplus environment was evaluated only in one single THAI experiment. However, these uncertainties in the poisoning temperature do not translate into large uncertainties of the PAR behaviour in an actual plant simulation. Increasing the poisoning temperature from 260 °C to a more conservative value like e.g. 340 °C would only lead to an increase of the effective concentration x_{eff} by 0.5 vol% at the onset of poisoning, compare Eq. (7).

The independent variables determining PAR poisoning are the respective gas concentrations and the environmental temperature. The catalyst temperature, as used in the poisoning criterion in Eq. (9) depends on the aforementioned independent variables. For catalyst arrangements where the catalyst surface temperature is not easily predictable, EU AMHYCO Deliverable D3.2 (Reinecke et al., 2023) presents an alternative poisoning correlation, depending only on the independent variables.

3.6. PAR start-up behaviour

PAR operation is autocatalytic, meaning that the exothermic recombination of the combustible gases heats up the catalytic sheets, which in return accelerates the recombination, leading to an even more rapid chemical energy release, compare Chapter 2.2.

To accelerate heat-up, the heat capacity of the catalytic sheets and their heat losses are minimized in the Framatome PAR. Nevertheless, the heat capacity of the catalytic sheets is finite. Their heat capacity may even be significantly increased by condensed water vapor adsorbed on the sheet surface. Furthermore, other foreign materials like soot or dirt may initially partially cover the catalytic surface. This may elongate the time delay until the PAR switches from its inactive to its active state.

The prediction of this start-up delay is the main factor of uncertainty to predict the behaviour of PARs. In the original AREVA PAR correlation the start-up delay was incorporated by the condition that the hydrogen concentration must exceed 2 vol% once. This criterion, however, has several disadvantages like e.g. giving the wrong impression that PARs would not start below 2 vol%. Therefore, in the following a new start-up criterion is constructed.

The light-off of catalysts was experimentally examined at the Research Centre Jülich (Allelein et al., 2018) by measuring the start-up delay of generic catalyst samples after exposition to a defined humidity. It was observed that the start-up delay time is linear dependent on the mass of condensate adsorbed on the catalytic surface, and about inverse proportional to the concentration of combustible gases. Only 4 vol% and 1.3 vol% hydrogen concentrations were measured, thus this statement should be regarded as approximate.

These results can be interpreted such that before the PAR can heat-up and reach its autocatalytic active state, the condensate adsorbed on the catalyst surface must be vaporized by the heat released from the slow catalytic recombination of gases on the cold catalyst surface when the PAR is still considered in its inactive state.

The evaporation of the surface condensate requires a certain amount of energy. Based on Eq. (6), the heat power released on the catalytic surface in a PAR should be about proportional to the effective gas concentration $x_{\text{eff}} = \min(\nu_{\text{H2}} + 0.414\,\nu_{\text{CO}}, 0.84\,\nu_{\text{O2}})$. Thus, a measure of the energy $E(t)$ released on the catalytic sheet up to time t is the time integration of the effective gas concentration:

$$E(t) \propto \int_0^t x_{\text{eff}}(t') \bullet \text{d}t'. \tag{10}$$

When assuming a relative humidity of 75 %, a mass gain of 4 mg of the catalyst samples was observed (Allelein et al., 2018). With this condensate load, the sample started with a delay time of the order of 5 min in an atmosphere containing 4 vol% hydrogen. Thus, as rough estimate, it takes 5 min × 4 vol% ≈ 20 vol% min for the catalyst sample to start.

While not important for containment simulations, for other applications where a PAR may be exposed to very low gas concentrations for a very long time, the criterion in Eq. (10) may benefit from a lower effective gas concentration cut-off.

Based on these considerations and observations, the following phenomenological PAR start-up parameter is proposed:

$$c_{\text{start}} = \begin{cases} 1 \text{ if } \displaystyle\int_0^t x_{\text{eff}}(t') \bullet \text{d}t' > 20\,\text{vol\%} \bullet \text{min} \\ 0 \text{ otherwise} \end{cases} \tag{11}$$

This criterion can be intuitively interpreted that a PAR starts operating in its autocatalytic active state after being exposed for 10 min to a mixture of 2 vol% hydrogen in air (or 5 min in a mixture of 4 vol% hydrogen in air, or 20 min in a mixture of 1 vol% hydrogen in air). The suitability of this approach was tested by comparison to the THAI experiments (FRA, 2024b).

In PAR experiments, e.g., in the THAI facility, a significant spread of PAR start-up delay times was observed. However, once a PAR reached the active state, there was no further start-up delay for the rest of the experimental run, even if the PAR became inactive in the meantime. This is interpreted as that the catalyst clears itself of any surface contaminants during activation and that the remaining heat prevents condensation or other deposits on the surface due to the thermophoresis effect for a considerable time. In Eq. (11), the time-integrated effective gas concentration is monotonously increasing. This ensures that a PAR, which reached its start-up criterion once in a simulation, will not show any start-up delay any more for the rest of the simulation, which is in line with the experimentally observed PAR behaviour.

As observed in experiments, when locally a spot on a catalytic sheet starts heating up, it will "burn" through the entire sheet and following the entire sheet stack assembly, bringing the entire PAR into its autocatalytic active state in a short period of time. Therefore, it can be expected that the PAR start-up becomes more and more reliable the larger the PAR assembly size becomes. Therefore, the proposed PAR start-up criterion of 20 vol% × min, deduced from small-scale catalyst samples (Allelein et al., 2018), is likely conservative. The experimental data from THAI (FRA, 2024b) point to a start-up criterion of 10 vol% × min. And even this criterion likely is conservative as the PAR in the THAI facility has the size of 1/10th of a PAR type FR90/1-1500T, compare Table 1.

Note that this argumentation about delayed PAR startup only refers to the impact of humidity, which is considered to be the most likely contributor to the delay. Further effects, e.g., of cable fire products, which could be released during accident initiating events, are not considered.

3.7. Par-induced ignition

The higher the effective gas concentration x_{eff}, see Eq. (6), in the containment atmosphere, the higher the PAR recombination rates, see Eq. (1) to Eq. (4), the hotter the PAR and the exhaust gases become. At a sufficiently high effective gas concentration, the PAR will ignite the surrounding atmosphere. The property of a PAR to ignite a combustible atmosphere does not represent a nuclear safety risk for the following reasons:

- Hydrogen has such a low ignition energy, that numerous possible sources of ignition exist in a nuclear containment. Therefore, when a combustible gas cloud could form inside the containment, the consequences of a potential ignition must be conservatively evaluated anyway in a safety assessment report, independent of the presence of PARs or other known sources of ignition.
- It is not the intended purpose of PARs to mitigate large gas clouds within a nuclear containment with such a high concentration of combustible gases that the cloud supports flame acceleration or even deflagration-to-detonation-transition. When the containment design promotes the formation of such gas mixtures in a time scale shorter than PARs can consume that hydrogen, e.g., in ice condenser containments, then PARs alone are not the most suitable system for hydrogen mitigation in the first place. For that reason, e.g., BWR containments with pressure suppression systems are mostly nitrogen inertised to exclude hydrogen combustion by displacing oxygen. The purpose of a PAR system is to prevent the formation of such high-concentration gas mixtures in slowly evolving systems like, e.g., large dry containments of a PWR (without early spray activation), confined spaces with radiolytic gas generation like waste storage tanks, or small hydrogen release flows, e.g., in battery rooms or due to radiolysis.
- Framatome PAR induce ignition at effective gas concentrations well below values where a deflagration-to-detonation must be expected. Thus, in a slowly evolving containment atmosphere, a PAR-induced ignition should lead to a laminar combustion, without leading to highly dynamic challenges to containment integrity. Such a laminar combustion, which does not threaten the containment integrity, can be seen as beneficial as it accelerates the consumption of the fuel or oxygen. Only in strongly transient situations, a cloud within a containment, which may get ignited by a PAR, may locally satisfy flame acceleration criteria. Such selected situations are to be examined in the licensing process of a combustible gas control system (Dimmelmeier et al., 2012).
- In none of the numerous experiments, where PAR-induced ignition occurred, did the PAR suffer blast damage or became inoperable thereafter. Based on that observation, a failure of the PAR system due to PAR-induced ignition of the containment atmosphere is not seen as a significant safety risk.

Experimentally, there is a certain spread of effective gas concentrations at the time of ignition. The PAR-induced ignition of the containment atmosphere was observed in the KALI-H2 and H2PAR tests at 6–7 vol% hydrogen (Mimouni et al., 2011; Reinecke et al., 2010). In the THAI experiments, PAR-induced ignition mostly occurred at effective gas concentrations of about (7 ± 1) vol.% (FRA, 2024b). It is proposed that the PAR-induced ignition likely depends on the chemical energy release of the PAR, and thus depends on the effective gas concentration x_{eff} as introduced in Eq. (6), compare Chapter 3.3.

The treatment of PAR-induced ignition in a containment simulation depends on the purpose of the simulation.

- If the simulation shall be a best-estimate approximation to reality, then the ignition criterion can be defined as $x_{\mathrm{eff}} > 7\,\mathrm{vol.\%}$, i.e., at the 50 % percentile of ignition as observed in experiments. Note that for numerical reasons, the logical larger connection ">" is preferred over an equal sign.
- For design simulation purposes, it may not be the goal to simulate reality as close as possible, but to find enveloping accident progressions. In case the simulation shall be conservative with respect to thermal loads onto containment structure, then the total hydrogen mass which burns during a defined period of time shall be maximized. If the simulation shall be conservative with respect to quasi-static combustion pressure loads, then the hydrogen combustion mass consumed in a single ignition event shall get maximized when the containment is already at elevated pressure. To find respective conservative peak values, the PAR ignition criterion can be varied within the bounding interval of $5.5\,\mathrm{vol.\%} > x_{\mathrm{eff}} > 8.5\,\mathrm{vol.\%}$. That concentration interval can also be used in an uncertainty analysis concerning PAR-induced ignition.
- If the hydrogen risk shall be assessed independent of a specific ignition event, then an ignition should not be considered in the containment simulation at all. Instead, after the simulation is run without combustion, the hydrogen risk can be evaluated by post-processing at every single point in time. This way, the conservative bounding worst ignition time and location can be identified. At this time then ignition can be artificially triggered and the resulting dynamic combustion processes (flame acceleration, pressure waves, …) can be evaluated by, e.g., means of computational fluid dynamics. The goal of this rather artificial approach is not a realistic description of the containment atmosphere, but a conservative bounding exclusion of a risk of containment failure by in-containment hydrogen combustion. Such an approach is for example used in the computational validation of the combustible gas control system for new-built EPR$^{\mathrm{TM}}$ reactors (Dimmelmeier et al., 2012). However, if no combustion is allowed in the containment simulation, then it must be ensured that the PAR recombination correlation does not leave its validated parameter regime. Therefore, the recombination rates predicted by the AMHYCO correlation (Eq. (1) and Eq. (3)) can be scaled down by the factor $\gamma = \min[1,\ (7\,\mathrm{vol.\%}/x_{\mathrm{eff}})]$. Thus, the effective gas concentration governing the behaviour of the PAR is artificially limited to values below 7 vol.%.

When performing containment simulations with considering PAR, it is good practice to specify how PAR-induced ignition is handled.

4. Validation against THAI experiments

The PAR correlation presented in this paper was deduced from the insights gained by small-scale experiments employing generic catalytic sheets (Reinecke et al., 2023; Reinecke et al., 2024), and subsequently validated by Framatome against the THAI experiments employing original full-scale Framatome PAR catalytic sheets. The evaluated experiments included HR-1 to HR-5 (dry atmosphere), HR-6 to HR-13 (steam atmosphere), HR27 and HR-28 (dry atmosphere), HR-29 and HR-30 (steam atmosphere), HR-31 (CsI-Interaction), HR-32 (iodine poisoning test), HR-33 and HR-34 (low oxygen), HR-35 (very low oxygen), HR-36 and HR-37 (PAR-induced ignition), HR-43 to HR-50 (counter-current gas flow), HR-51 to HR-53 (carbon monoxide poisoning), and HR-54 to HR-57 (carbon monoxide and hydrogen exposure). As some of these test data, especially the ones involving carbon monoxide, are not publicly available, the validation is documented in a non-public report (FRA, 2024b). The validation includes the comparison of the measured hydrogen and carbon monoxide recombination rates to the ones predicted by the correlation (compare Chapter 3.1 and Chapter 3.2), the dependency of the peak catalyst temperature on the effective gas concentration (compare Chapter 3.3), the PAR start-up delay (compare Chapter 3.6), as well as the PAR-induced ignition (compare Chapter 3.7). Overall, the deviation between predicted gas consumption rates and experimentally observed rates mostly falls within a ±20 % confidence interval.

Even though no full insight into all validation cases can be given here, to present at least a convenient overview about the increased accuracy of the new AMHYCO correlation in comparison to the preceding AREVA correlation, FZJ developed a steady-state validation database, see Fig. 6. In these plots, the experimentally observed hydrogen recombination rates at selected points in time in various THAI experiments are compared to the prediction of the respective correlations. The experiments in oxygen-surplus conditions are marked with open symbols, the experiments performed under oxygen-starvation are shown with filled symbols. Especially for the experiments under oxygen-starvation, the original AREVA correlation (upper plot in Fig. 6) showed a significant uncertainty. This uncertainty was significantly reduced by the new AMYHCO correlation (lower plot in Fig. 6). The rates for carbon monoxide recombination (not included here) show a comparable trend.

An example of the validation against specific experimental data of the HR-53 test from the German national THAI VI program (Freitag

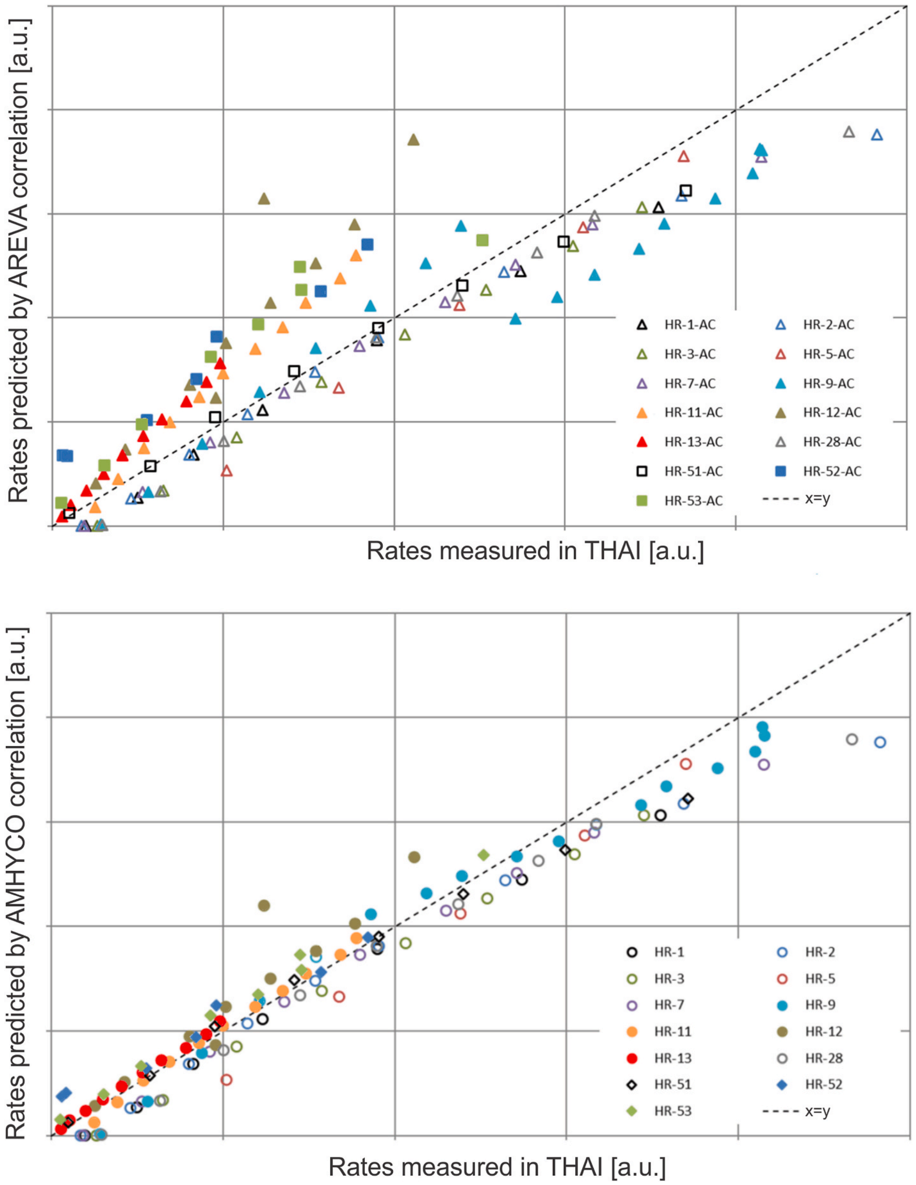

Fig. 6. Comparison of hydrogen recombination rates obtained from various THAI experiments to the predictions of the older AREVA correlation (upper plot) and the new AMHYCO correlation (lower plot). The open symbols mark experiments in oxygen-surplus, the filled symbols mark experiments with oxygen-starvation.

et al., 2020) is given in Fig. 7. In the HR-53 test, a prototypical PAR with original Framatome catalytic sheets is exposed to a defined atmosphere containing air, hydrogen and carbon monoxide. Based on concentration measurements below and above the PAR as well as a measurement of the gas flow rate through the PAR, the PAR recombination rates for hydrogen and carbon monoxide are experimentally determined, depending on the gas composition at the PAR entry opening. In the following Fig. 7, these measured recombination rates are compared to the predictions of the AMHYCO correlation.

The orange and green markers show the experimentally measured recombination rate for hydrogen and carbon monoxide, respectively. The red continuous line indicates the new engineering correlation for hydrogen as described in this paper. The red dashed line indicates the AREVA engineering correlation previously in use. In analogy, the blue lines indicate the engineering correlations for carbon monoxide.

In the beginning of test HR-53, a small amount of carbon monoxide is injected for ~5 min into the THAI vessel, which contains an oxygen-rich atmosphere. After ~8 min, the temperature of the PAR catalytic plates starts rising by ~20 K due to carbon monoxide recombination. Likely the high ambient gas temperature in the presence of excess oxygen limited poisoning in the initial stage. Nevertheless, the PAR recombination rate under this low carbon monoxide concentration is too low to be noticeable in Fig. 7. At ~20 min, hydrogen and carbon monoxide gets injected with a high flow rate into the THAI vessel. As the PAR is already active, it directly started recombining the combustible gases. Due to the conservatively chosen start-up criterion in the PAR correlation, see Chapter 3.6, the prediction for the PAR operation is initially delayed for about 10 min to 33 min. After the start-up of the PAR in the correlation, the predicted recombination rates follow the experimentally measured ones with high accuracy.

During the first hydrogen injection phase, the PAR operates in an oxygen-rich environment. In this regime, the new AMHYCO correlation is functionally identically to the old correlation. In the second injection phase, the PAR starts operating under oxygen-starvation. Here, the new AMHYCO correlation reproduces the experimental findings for hydrogen and carbon monoxide with a significantly higher accuracy than the old correlation.

At the end of the HR-53 experiment, at ~380 min, the PAR starts to become affected by carbon monoxide poisoning. The onset of the poisoning is so weak, that it is not noticeable in the measured recombination rates in Fig. 7, but only in the minimum catalyst surface temperatures, compare T_{min} in Fig. 4 (right). The onset of the poisoning phenomenon is also predicted by the new AMHYCO correlation at ~340 min. With satisfying the criterion in Eq. (9), the factor c_{poison} and thus the PAR recombination rates are conservatively set equal zero. The original AREVA correlation does not include a poisoning criterion.

5. Summary and conclusions

This manuscript establishes a new engineering correlation to predict the recombination rates for combustible gases by Framatome passive autocatalytic recombiners (PAR). This new correlation represents a continuation and generalization of the original in-house AREVA correlation based on latest findings obtained within the SAMHYCO-NET and AMHYCO projects. The new correlation, referred to as “AMHYCO correlation for Framatome PAR” shall enhance the description of PAR performance especially in the late “ex-vessel” stage of a severe accident where a molten corium-concrete interaction may release large amounts of hydrogen and carbon monoxide. In addition, criteria for PAR poisoning by carbon monoxide, PAR-induced ignition, and a new treatment for a delayed start-up of the PAR were implemented.

The new AMHYCO correlation has a mathematical complexity similar to the preceding AREVA correlation, ensuring that it can easily be incorporated into containment simulation codes, without significantly affecting their calculation times or calculation stability.

The main priority of constructing the correlation framework was to employ a minimum number of physically justifiable fit parameters to recreate the results of all considered experiments simultaneously. The following fits were used: the PAR-size describing constants k_1 and k_2 in Eqs. (1) and (3), a decreasing function and a lower gas cut-off concentration in Eq. (8), the PAR startup criterion in Eq. (11), the correlation of gas concentration to maximum catalyst temperature in Eq. (7), and two poisoning temperatures, one for oxygen surplus and one for oxygen starvation, in Eq. (9). Concerning selecting suitable numerical values for these fit parameters, the most accurate representation of a single experiment had a lower priority than the general applicability to all experiments.

The new AMHYCO correlation for Framatome PAR was validated against data from various existing and new experiments, especially experiments performed in the THAI facility. Furthermore, as the new AMHYCO correlation corresponds to the original AREVA correlation for hydrogen in an oxygen-rich atmosphere (in the absence of carbon monoxide), former experiments in that parameter regime (e.g., KALI-H2 and H2PAR tests) are part of the validation basis.

Overall, the AMHYCO correlation shows an improved agreement with the experimental observations compared to the original AREVA correlation in oxygen-starvation conditions (present in the late stage of a nuclear accident). The deviation between predicted gas consumption

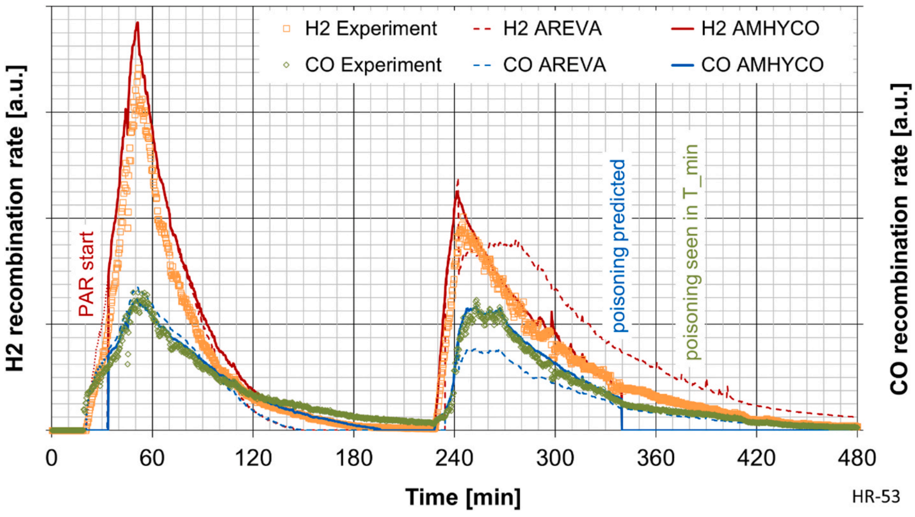

Fig. 7. Validation against THAI HR-53 experimental data.

rates and experimentally observed rates mostly falls within a ±20 % confidence interval.

Additional considerations concerning the numerical implementation of the proposed PAR correlation can be found in the appendix to the AMHYCO Deliverable D5.1 (AMHYCO 2025).

Note that all statements within this manuscript are to be considered valid only for PARs from the nuclear supplier Framatome. Experimental evaluation of different catalysts, e.g., in Phebus, showed that catalytic materials of different origin are not interchangeable. Also, the geometric design of the PAR, affecting gas flow resistance and catalyst heat loss, affect the PAR performance. Therefore, no statements concerning reliability or performance can be made for PAR designs of other nuclear vendors, which did not actively participate in this research program.

CRediT authorship contribution statement

Matthias Braun: Writing – original draft, Validation, Conceptualization. **Ernst-Arndt Reinecke:** Supervision, Resources, Investigation, Data curation.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: The author Matthias Braun declares that he is employed by Framatome GmbH, the world-market leader for passive autocatalytic recombiners. This fact may be considered as potential competing interest.

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Data availability

The data that has been used is confidential.

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