

REKO-Fire: new facility to investigate cable fire impact on Passive Autocatalytic Recombiners

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1 Introduction

Fires in nuclear power plants (NPPs) represent a significant risk to nuclear safety [1]. Cable fires may occur not only at any time in the course of a severe accident but may also be the initiating event of a severe accident sequence. During combustion, high amounts of aerosols combined with specific combustion gases (e.g. CO, CO₂) can be released. The effect of cable fire products inside the containment and their interaction with the catalyst surfaces of passive autocatalytic recombiners (PARs) is of significant relevance for safety analyses in order to assess the hydrogen mitigation efficiency under these conditions. PARs are passive devices that aim to recombine hydrogen through an exothermic catalytic reaction and consist of a vertical channel equipped with catalyst sheets in the lower part. The catalysts can be platinum or palladium-based. The reaction in a recombiner starts spontaneously as soon as the hydrogen concentration begins to increase in the reactor building atmosphere [2].

Cable fire products (CFPs) are physically a mixture of solid, liquid and gaseous components. Released particles are smallest drops of condensed, organic substances and also carbon-rich agglomerates of hundreds up to many thousands of nearly spherical primary particles [3]. These particles can easily distribute inside the containment and reach the catalyst surfaces of recombiners.

Several investigations have been carried out by the PARs manufactures and independent test centers as part of the qualification program for commercial recombiners used in NPP. During these qualification programs, the application of massive oil and cable fire products, no irregularities have been found in the operating behavior of recombiners [4]. However, fire products have played a role in two real cases. Studies have shown that organic residues of diesel fire led to functional failures during recurrent tests [5] and that volatile organic compounds delays the start-up for hydrogen removal by poisoning or blocking of the catalytic surface [6]. Also, after a plastic fire in the Swedish NPP Ringhals 2, severe contamination of the catalyst sheets with soot and significant corrosion damage has been observed [7].

The products generated in cable fires depend on the combustion conditions, which are defined by the availability of oxygen (level of ventilation) and temperature. In real fires, these parameters may vary locally and over time. The conditions are classified as well-ventilated combustion, where the fire is small compared to the room and the fuel is controlled; under-ventilated combustion, where the fire is large in relation to the room and the ventilation is limited; and flameless combustion that is divided into self-sustaining fire, oxidative pyrolysis and anaerobic pyrolysis, as shown in table 1.

Table 1: Classification of fire conditions based on ISO TS 19706, cited in [8].

Fire type or phase		Temperature [°C]		Environment O ₂ [%]	Exhaust gas	
		Fire	Hot layer		O ₂ [%]	CO ₂ /CO ratio
1 Flameless combustion						
a	Self-sustaining smoldering fire	450-600	RT	21	>20	1-5
b	oxidative pyrolysis	300-600	<50	21	>20	1-5
c	(Anaerobic) Pyrolysis	300-600	<50	0	0	<5
2 Well-ventilated combustion (small fire in relation to the room, fuel controlled)		>700	RT-500	>15	5-21	>20
3 Under ventilated combustion (large fire in relation to the room, ventilation controlled)						
a	Small impaired fire in enclosed spaces	>700	RT-500	<15	0-12	2-20
b	Fading fires in large or open spaces	>700	500-1000	<15	0-12	2-20

The amount of oxygen and fuel in a closed space is limited; consequently, the consumption of oxygen and fuel has an impact on the course of the fire. There are different phases during a fire in closed rooms. After ignition, flammable material is heated and the fire spreads. The room temperature is increased during the propagation phase and then the rise of the temperature is replaced by strong acceleration of fire propagation, leading to a full fire. The full fire continues while there are still combustion conditions. When oxygen or fuel is consumed, the fire becomes extinct and the temperature decreases.

In order to better quantify the impact of cable fire products and to assess the relevance for severe accident scenarios, a cable fire product generator has been established at Forschungszentrum Jülich.

2 Methodology

In order to further investigate the impact of cable fire products on catalysts used in PARs, the REKO-Fire facility, shown in Figure 1 has been established at Forschungszentrum Jülich. The facility design aims to combine a flow tube reactor for catalyst investigation with a steady-state tube furnace for constant generation of CFPs at different combustion conditions. Thus, the REKO-fire allows to simultaneously expose the catalyst to CFPs and hydrogen, enabling to quantify the influence of volatile components on the catalysts' start-up behavior.

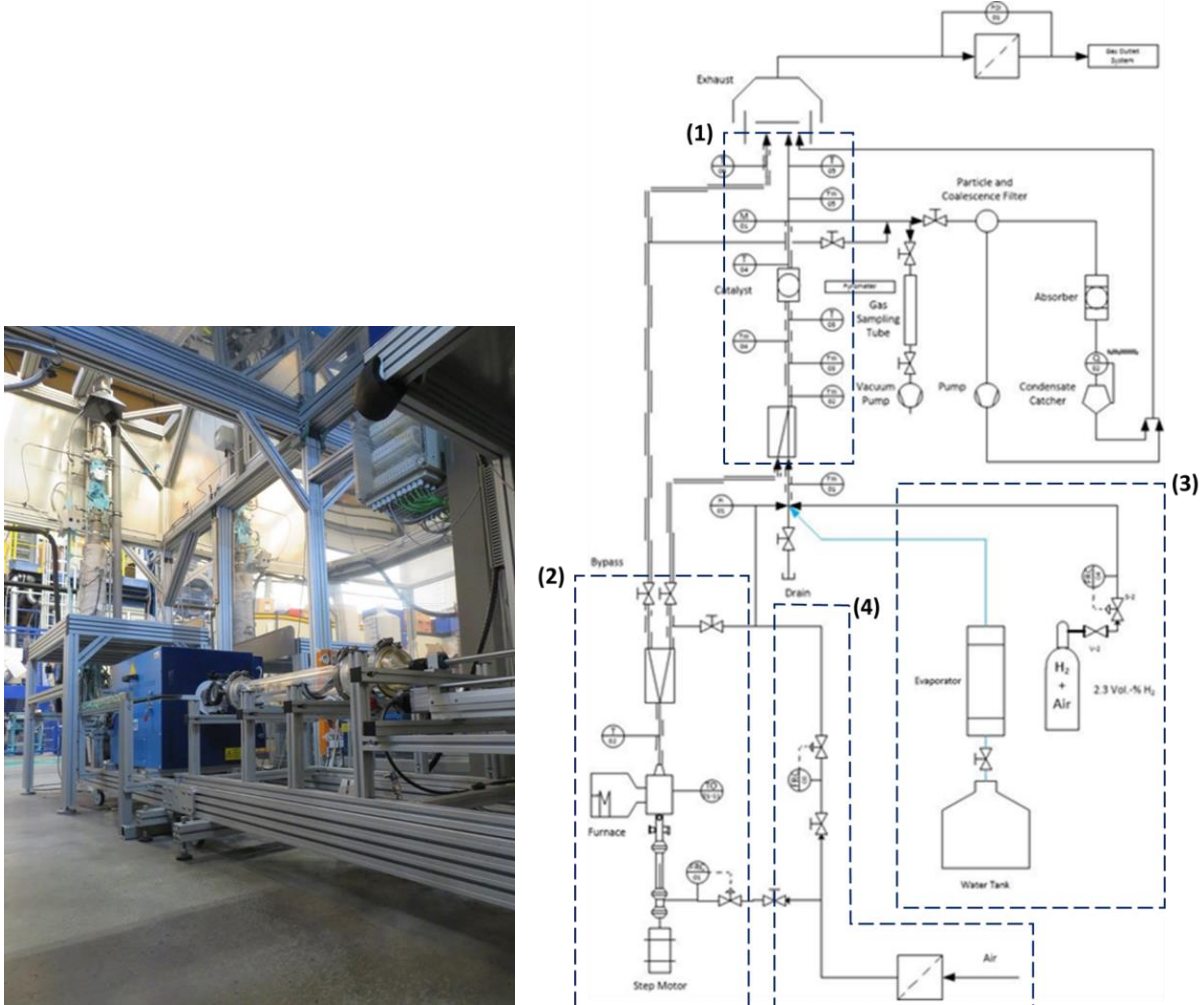


Figure 1: REKO-fire facility (left) and flow chart of facility (right).

The REKO-Fire facility is able to investigate the impact of cable fire products on the hydrogen reaction on catalyst samples. As shown in the flow chart in Figure 1, the facility consists of vertical cylindrical reaction tube (1), where the catalyst is placed, connected with 3 feed lines: cable fire product generator (2), hydrogen/air mixture and steam generator (3), and dilution air (4). The outlet of the reaction tube is connected to the off-gas treatment system. The gas temperature is measured by means of thermocouples (TCs) installed at the inlet and the outlet of the reaction channel. Several heating elements allow to heat the tube in order to reduce both heat losses and the deposition of fire products on the inner tube wall.

The concept of the cable fire product generation system is based on the "steady state tube furnace" described in ISO/TC 19700. It provides temporally constant and reproducible cable fire products from representative cable material avoiding "artificial" by-products, such as those produced by ignition using a gas burner. In addition to the composition of the cable material, the prevailing fire conditions are decisive for the extent and properties of the resulting products.

The cables used in the experiments are of the type FRNC-BX 4 x 2.5 mm², a representative power cable for German Convoi-type NPPs, manufactured by company Prysmian Kabel und Systeme GmbH. The catalyst samples used in this test series are thin sheets with a size of 5 x 5 cm². The supporting material is a thin sheet of stainless steel with a thickness of 50 µm. The ceramic washcoat consists of 4 mg/cm² γ-Al₂O₃. The amount of catalytical active material (platinum or palladium) is 1 mg/cm².

This work focuses on the impact of cable fire products generated in well-ventilated combustion, under-ventilated combustion and oxidative pyrolysis on palladium based catalyst. Experiments using platinum based catalyst have been previously performed. For each catalyst, two tests have been performed.

The test sequence starts with the continuously combustion of the cable which is led into the bypass. After steady-state conditions are achieved, the CPFs are led through the reaction channel, followed by hydrogen injection. Temperature measurement at the top of the catalyst indicates if the conversion reaction occurs: the temperature will increase when hydrogen is recombined since it is an exothermic catalytic reaction.

The gas composition during bypass operation was used to analyze the combustion regimes. For well-ventilated conditions, we observe the large portion of oxygen and a high CO_2/CO ratio. In the under-ventilated case, the off-gas contains almost no oxygen and a lower CO_2/CO ratio. In the pyrolysis case, almost no CO_2/CO is produced and high oxygen concentration.

3 Results

Experimental results of the tests with the palladium catalyst are given in figures 2, 3 and 4 for well-ventilated combustion, under-ventilated combustion and oxidative pyrolysis. The history plots show the gas temperature above the catalyst foil as an indicator of catalyst behavior (red curve, values of the primary ordinate axis), the hydrogen inlet concentration (blue line, secondary ordinate axis) and the CO_2 and CO outlet concentrations (purple and green lines, secondary ordinate axis) over time.

Figure 2 shows experiment performed with a palladium catalyst under well-ventilated combustion conditions. During H_2 injection, the secondary air flow was reduced by the same amount in order not to further dilute the combustion gas (CO and CO_2 volumetric fractions in this diagram). After the catalyst foil is exposed to the combustion products, three hydrogen injection phases are performed from $\sim 17,400$ s, $\sim 18,900$ s and $\sim 20,350$ s. The hydrogen concentration is subsequently diminished with each new injection phase from 1.0 vol.% to 0.5 vol.% and 0.25 vol.%. In each of these cases, catalyst start-up occurs immediately after start of the injection as indicated by the increase of the measured temperature. The end of the cable combustion is indicated by the drop of the CO_2 concentration starting at $\sim 20,500$ s during the third hydrogen injection.

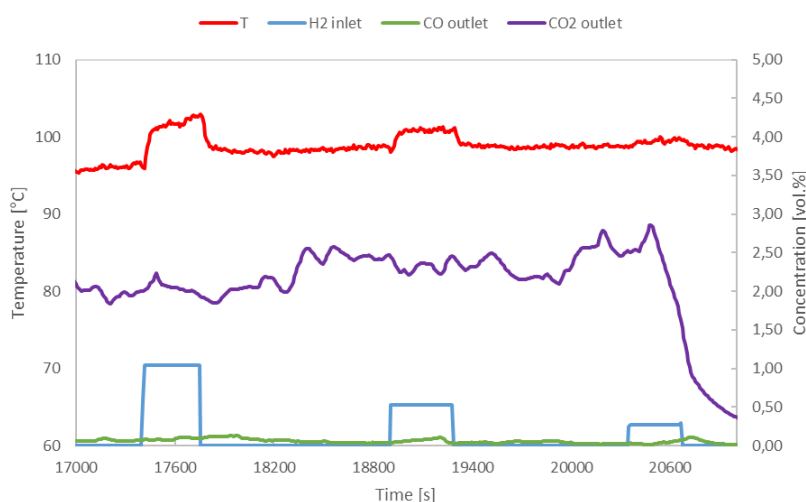


Figure 2: Pd catalyst behavior during well-ventilated combustion

Figure 3 shows the experiments performed in an under-ventilated combustion conditions. These conditions are obtained with a reduced primary air flow. The CO_2/CO ratio is significantly lower compared to well-ventilated combustion. After the catalyst foil is exposed to the combustion products,

two hydrogen injection phases are performed from ~14,400 s and ~15,250 s at 1 vol.%. In both cases, catalyst start-up occurs immediately after start of the hydrogen injection as indicated by the temperature measurement. However, when a third hydrogen injection is performed at a reduced hydrogen concentration of 0.5 vol.% (~16,200s), the catalyst remains inactive.

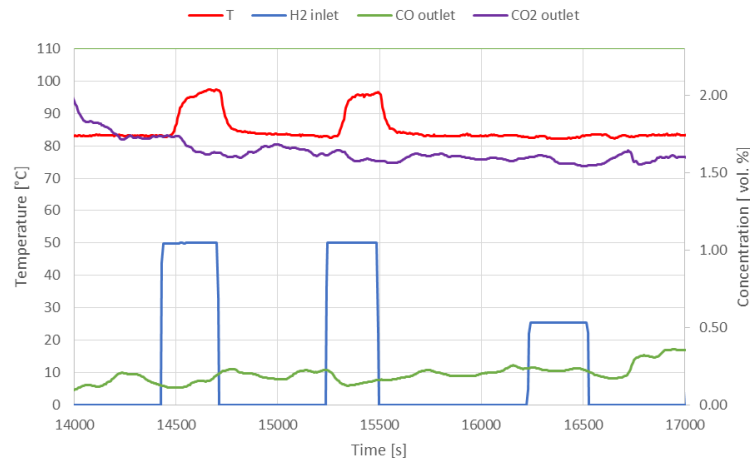


Figure 3: Pd catalyst behavior during under-ventilated combustion.

Experimental result of test with oxidative pyrolysis conditions are given in Figure 4. The conditions inside the furnace yield low concentrations of CO and CO₂, but significant amounts of organic substances. Start-up delay is observed in the first injection phase (~12,600 – 13,800 s) when the inlet hydrogen concentration needs to be increased to 2 vol.% to activate the catalyst. In a second injection series (~17,400 – 19,000 s), no reaction is observed at all. However, in-between injections of 1 vol.% hydrogen exhibit immediate or at least slightly delayed start-up of the catalyst.

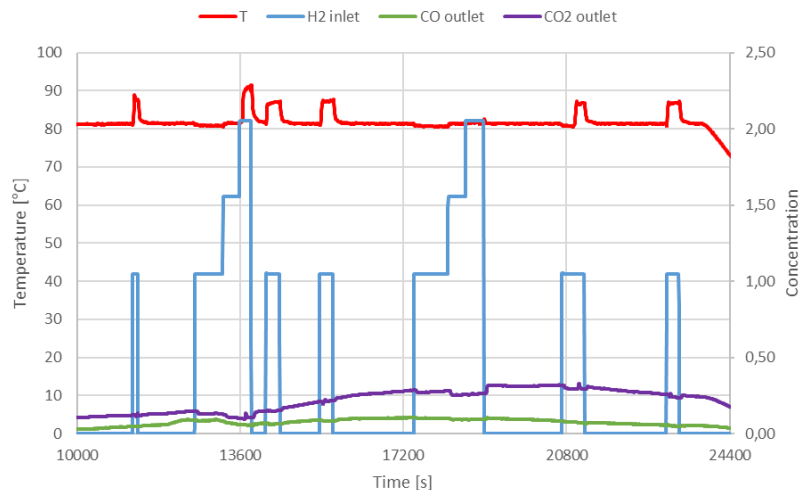


Figure 4: Pd catalyst behavior during oxidative pyrolysis.

4 Conclusions

Cable fires in a nuclear power plant can lead to the release of enormous amounts of aerosols alongside specific combustion gases (e.g. CO, CO₂). In the present investigation, the effect of CFPs obtained from FRNC cables on the catalyst behaviour of Pd catalysts for hydrogen recombination has been studied.

For well-vented cable fire, neither gaseous (CO₂ in relatively large amounts and very little CO) nor particulate (mainly soot) CFPs seem to affect the onset of the catalytic H₂ conversion for Pd-based catalyst.

In under-ventilated fire conditions, the Pd-based catalyst is only impacted by the CFPs at hydrogen concentration as low as 0.5 vol.%. Higher CO levels when compared with well-ventilated cable fires are suspected to be the reason for the observed deactivations.

The products generated from oxidative pyrolysis seems to cause deactivation and start-up delay due to the particulate deposition on the surface of the catalyst. Also experimental data indicates that the deactivation is more severe if the catalyst is exposed to the CFPs during a longer time

Therefore, it can be concluded from this work that carbon monoxide and particulate depositions from cable pyrolysis seem to be the main mechanisms for catalyst deactivation under the studied conditions. Since cable fires could be the initiating event, the catalysts might be affected by corresponding CFPs before the actual severe accident occurs. As a consequence, hydrogen mitigation by PARs might be significantly reduced. Based on the present study, future work will focus on under-ventilated fires and oxidative pyrolysis. New tests will involve different plant-typical cable types, e.g. PVC cables.

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