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Catalytic, hybrid lean combustion for gas turbines

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ABSTRACT

In the CATHLEAN project (an EU Framework 5 project performed between 2003 and 2006) a consortium of researchers investigated the development of an advanced, ultra-low NO*x*, hybrid burner for heavy-duty gas turbines that combines catalytic and lean-premix combustion components. Catalytic elements were developed that preheat the incoming lean fuel/air mixture up to temperature of ca. 1100 K at the required high (20–30 m/s) operating velocities and inlet temperatures (670–690 K). High-pressure aging of a highly active Pd-based catalyst at 10 bars indicated that the catalyst activity decreases significantly in the first 200 h of operation, and that an optimal catalyst design should contain different regions (in the axial direction) to allow optimization of catalyst activity and material stability, e.g. higher activity at the flow entrance, greater thermal stability at the reactor outlet where temperatures are high. Full-scale burner investigations demonstrated the feasibility of the CATHLEAN hybrid catalytic burner concept, and provided an indication of performance enhancement due to catalytic conversion. Both NO*x* emissions and lean stability limit were improved (25% reduction in NO*x* emissions and reduction of lean blow-out limit of ca. 200 K).

1. Introduction

Extensive effort has been placed on reducing thermal NO*x* emissions from gas turbines over the past decades. The market introduction of lean-premixed combustion was accompanied by a drastic reduction of these emissions. Today, the leading producers of modern, heavy-duty gas turbines (running on natural gas) achieve NO*x* emissions of 25 ppm (15% O_2) and below. The CATHLEAN ("*Cat*alytic *H*ybrid *Lean* Burner") project was motivated by anticipated reductions in NO*x* emission requirements for gas turbine power plants located in or near urban areas [1], and the potential that catalytic combustion designs such as [2] could impact the lowest achievable emissions rate (LAER) standards imposed upon new gas turbine-based power plants.

"Traditional" catalytically stabilized combustion (first demonstrated in 1974 [3]) is achieved within a honeycomb reactor in which the fuel/air mixture, or a portion thereof, is converted heterogeneously over a catalyst. Detailed descriptions of the fundamentals of catalytic combustion can be found in the literature [4–6]. A traditional catalytic combustion system is more or less completely dependent on proper catalyst function for operation. After a long development and multiple demonstrations of feasibility [7,8] this technology has never achieved a commercial breakthrough for large, heavy-duty gas turbines. Recently the technology was sold to Kawasaki who are offering the technology commercially to achieve <3 ppmv NO*x* for a 1.5 MW gas turbine GPB15X.

Traditional catalytic combustion is mainly seen as an alternative vehicle for NO*x* emissions abatement. Unlike tail-end systems such as SCR, catalytic combustion actively reduces the formation of pollutants, rather than cleaning up the flue gases. In contrast to the tail-end cleanup systems, catalytic combustion also has the potential to reduce pressure pulsations [9], and – by improving the lean combustion stability and reducing temperature non-uniformities in the exhaust gas – to reduce turbine cooling requirements, thereby increasing component lifetime [10]. Thus, for a similar cost as SCR [11], catalytic combustion provides additional benefits.

NO*x* improvements are expected in catalytically stabilized burners for the following reasons:

- Catalysts reduce the concentration of hydrocarbon radicals, whose presence is needed for the formation of prompt (or Fenimore) NO*x* [12] at temperatures well below the final flame temperature. Increasing the fraction of catalytic fuel conversion decreases the hydrocarbon radical pool and reduces the formation of prompt NO*x* [13,14].
- Zeldovich NO*x* formation will play an increasingly important role as the firing temperatures of large gas turbines are elevated (up to around 1500 °C, corresponding to adiabatic flame temperatures of nearly 1600 °C, or 1873 K) for reasons of cycle efficiency. Chemical kinetic computations indicate that the H_2O and CO_2 produced

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Table 1
List of partners participating in the Cathlean EU FP5 project.

Organization name	Symbol	Country
1. ALSTOM Power Ltd.	APUK	UK
2. Cranfield University		UK
3. ALSTOM Power Switzerland (APCH)	APCH	CH
4. Institut Français du Pétrole	IFP	F
5. Gaz de France	GdF	F
6. awtec AG	awtec	CH
7. Deutsches Zentrum für Luft und Raumfahrt	DLR	D
8. Ruhr Universität Bochum	RUB	D
9. Politecnico di Milano	Polimi	I
10. KTH Royal Institute of Technology, Stockholm	KTH	S

by fuel conversion within the catalyst have a strong mitigating effect upon thermal NO*x* production in the downstream homogeneous flame. Experiments conducted by Krill and Kesselring [15] confirmed this trend, for pressures and temperatures up to 10 bar and 1700 °C, respectively.
- Experiments and numerical studies indicate that NO*x* emissions from catalytically stabilized flames are significantly less sensitive to temporal fuel/air unmixedness than in the case of homogeneous combustion [16,17].

Dynamic stability should also be promoted by catalytic combustion, for the following reasons [9]:

- Heterogeneous conversion tends to decouple the chemical heat release from fluid dynamic instabilities because less thermal energy is released in regions of high turbulence intensity within a homogeneous flame front.
- Longitudinal pressure oscillations should be mitigated due to reduced coupling between pressure fluctuations within the homogeneous flame front and the fuel supply, located upstream of the catalyst support, and also because of the thermal inertia of the catalytic reactor.

It should be noted, however, that such instabilities are also a strong function of combustor geometry, and do not depend upon the burner alone.

Current designs for gas turbine catalytic combustors tend not to be fully compatible with the requirements of machine manufacturers and operators (an overview of relevant design and performance considerations is available in Ref. [18]). Typically, extensive combustor and casing modifications are necessary to accommodate additional components such as a pre-burner, and reliability and operability issues are not fully addressed. Little evidence exists to suggest that traditional catalytic combustion designs are directly applicable to larger machines characterized by higher firing temperatures and greater operating pressures. These concepts are also 100% dependent upon the proper functioning of the catalyst components, which are prone to overheating, activity loss and other forms of long-term degradation.

An advanced, ultra-low NO*x* burner containing catalytic elements operating at lean conditions was proposed and investigated in the current project. The hybrid design combines catalytic technology with novel aerodynamic components and new operating concepts, for introduction in current and future gas turbines in a lower risk manner. The idea is to have a burner design whose performance is enhanced by the presence of catalytic elements, without being completely dependent upon their functioning.

A 10-member EU consortium (whose members are listed in Table 1) was created within the context of a 3-year EU Framework 5 program, in order to research, design and develop a prototype hybrid burner.

Table 2
Overall boundary conditions for the retrofittable, hybrid burner.

Parameter	Boundary condition
Burner dimensions	Cylindrical envelope: ≈450 mm long, 200 mm diameter
Inlet velocity	20–30 m/s @ 400–450 °C
Pressure	15–25 bar
Total pressure loss	<3% of engine operating pressure
Total heterogeneous fuel conversion	>40% of total fuel flow

The present paper describes results of this project and an overall judgment of the performance and feasibility of the CATHLEAN burner.

2. The CATHLEAN burner

The proposed burner concept [19] was designed in order to be retrofittable to existing gas turbine combustors and operating conditions, significantly reducing the risks associated with its commercial introduction. These retrofit considerations resulted in stringent overall boundary conditions for the advanced, hybrid burner. Typical conditions derived from a gas turbine of the 100–150 MW class are listed in Table 2.

Fig. 1 depicts a schematic of the hybrid burner and its constituent components. The goal of retrofittability imposed tight operating constraints and required the development of a number of novel components, including a novel mixer for fuel/air. The burner was primarily designed for natural gas operation (permitting liquid fuel operation in a homogeneous combustion mode) and this is the focus of this paper.

The following component and operational requirements needed to be addressed and were the subject of research within the CATHLEAN program:

1. Sufficient catalytic ignition and conversion must be obtained at the short length and high linear velocities (i.e. large space velocities ≈ 10^7 h^{-1}) within the catalytic reactor. Novel catalytic formulations and synthesis and deposition methods were found to promote (a) low light-off temperatures (at approximately 670 K) and (b) long-term stability at high temperatures (up to 1200 K). Within the concept the catalyst activity must be sufficient to preclude the use of a pre-burner for ignition at gas turbine, full-load, conditions.
2. The burner must have means for aerodynamic flame stabilization, which must be strong enough to firmly anchor homogeneous combustion in the event of large-scale deactivation of the catalytic reactor, or when operating at part load conditions with associated low inlet temperatures. A novel swirl generator was

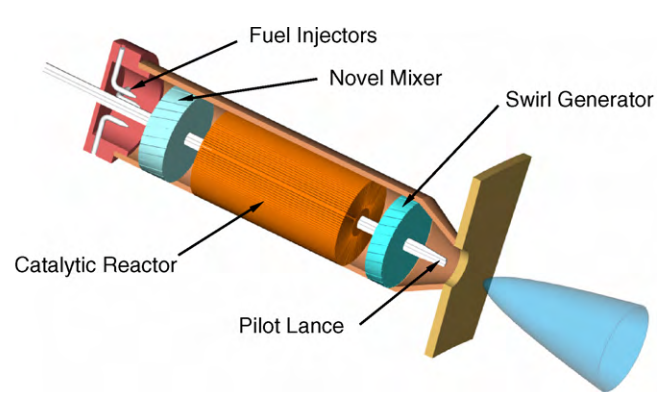


Fig. 1. Illustration of the advanced, hybrid burner and its constituent components.

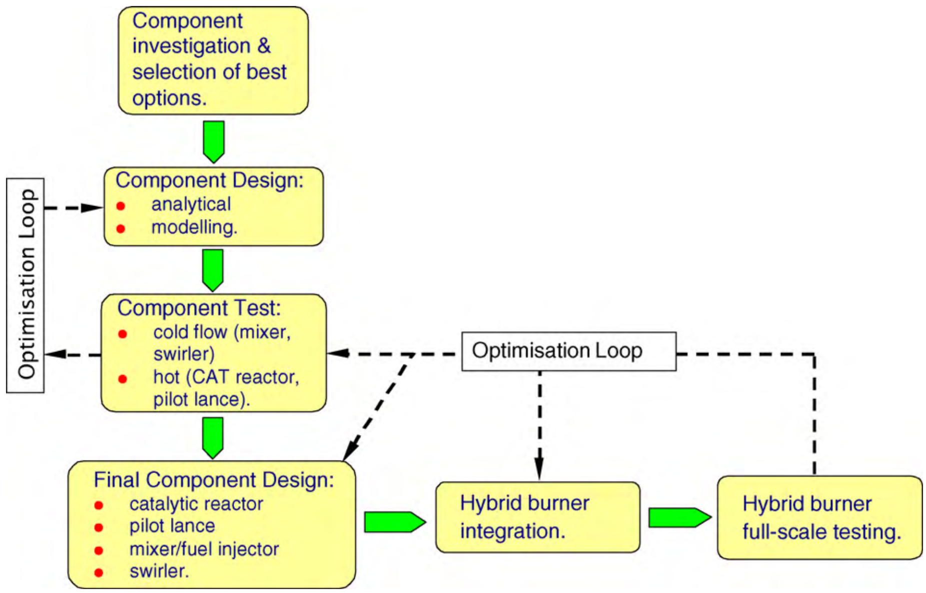


Fig. 2. Illustration of the design and integration process.

proposed, which can withstand the high temperatures (up to 950 °C) of the catalyst exhaust stream. This proposed swirler could integrate additional functions, such as arresting flashback of the homogeneous flame front, or providing further catalytic conversion. In this way, the burner offers ultra-low NO*x* capability coupled with long-term reliability.

3. Extremely high (by lean-premix standards) mixing qualities must be attained prior to entry of the fuel/air mixture into the catalytic reactor. This is necessary in order to avoid local overheating. Only a small volume is available for the fuel injection and mixing processes. High levels of fuel/air mixedness must be attained in very short distances (typically of the order of 100 mm), at high velocities, and with minimal pressure losses. The mixing process must additionally deliver a uniform axial velocity profile to the catalyst inlet zone. Devices to meet these criteria were proposed and investigated in the project [20].
4. A centrally located, low-emissions piloting lance for flame stabilization, especially at lower loads and during startup and transitions is required. This unit may contain catalytic components (as described by Griffin and Senior [21]), particularly those designed for fuel-rich heterogeneous oxidation which provides chemical flame stabilization (an example of which is outlined by Ruck et al. [22]). In the event of failure of the catalytic units, the lance must be able to revert to the partially premixed or diffusion flame modes commonly used in present applications. Furthermore, the pilot lance should also maintain the liquid fuel capability of current, lean-premix burners by injecting the liquid fuel from the lance tip into the combustion zone downstream of the catalytic reactor and swirl generator. This is to be achieved by providing sufficient space for liquid fuel ducts along the entire lance length, and for the atomization nozzle at the lance tip. Within the CATHLEAN project only the piloting lance for the support of natural gas operation was tested.

3. CATHLEAN project organization

The CATHLEAN hybrid burner consists of a number of novel components that required considerable research and development. These devices were individually designed and tested, and subsequently integrated to form the burner in its entirety. Fig. 2 illustrates the design process. Optimization of each component dictated that the design and test process be iterated until the specifications listed in Table 2 were matched. The culmination of the project was the performance of full-size atmospheric tests which revealed the need for further component optimization.

The tasks within the project were divided into discrete, self-contained work packages depicted in Fig. 3. Work packages (WP) 2–4 were focused on the development, testing and optimization of the catalytic elements within the burner. The specific goals of these work packages were catalytic material development, catalyst reactor design, and catalyst component testing, respectively. WP 5

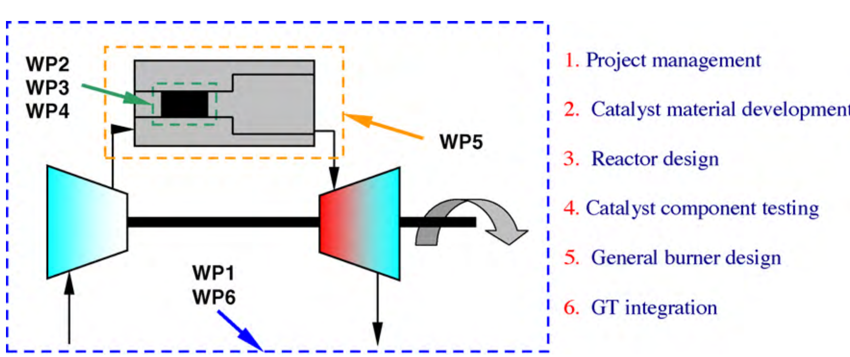


Fig. 3. Interrelationships between work packages.

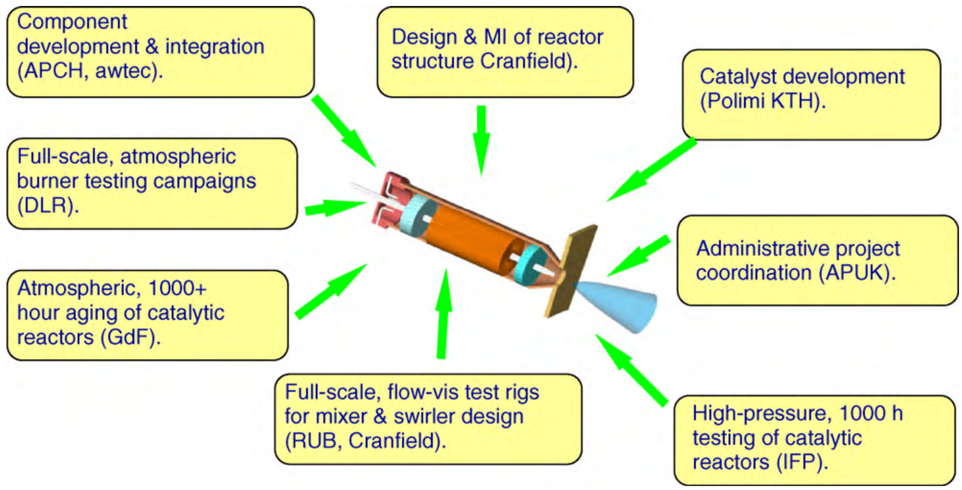


Fig. 4. Key activities of the various consortium partners.

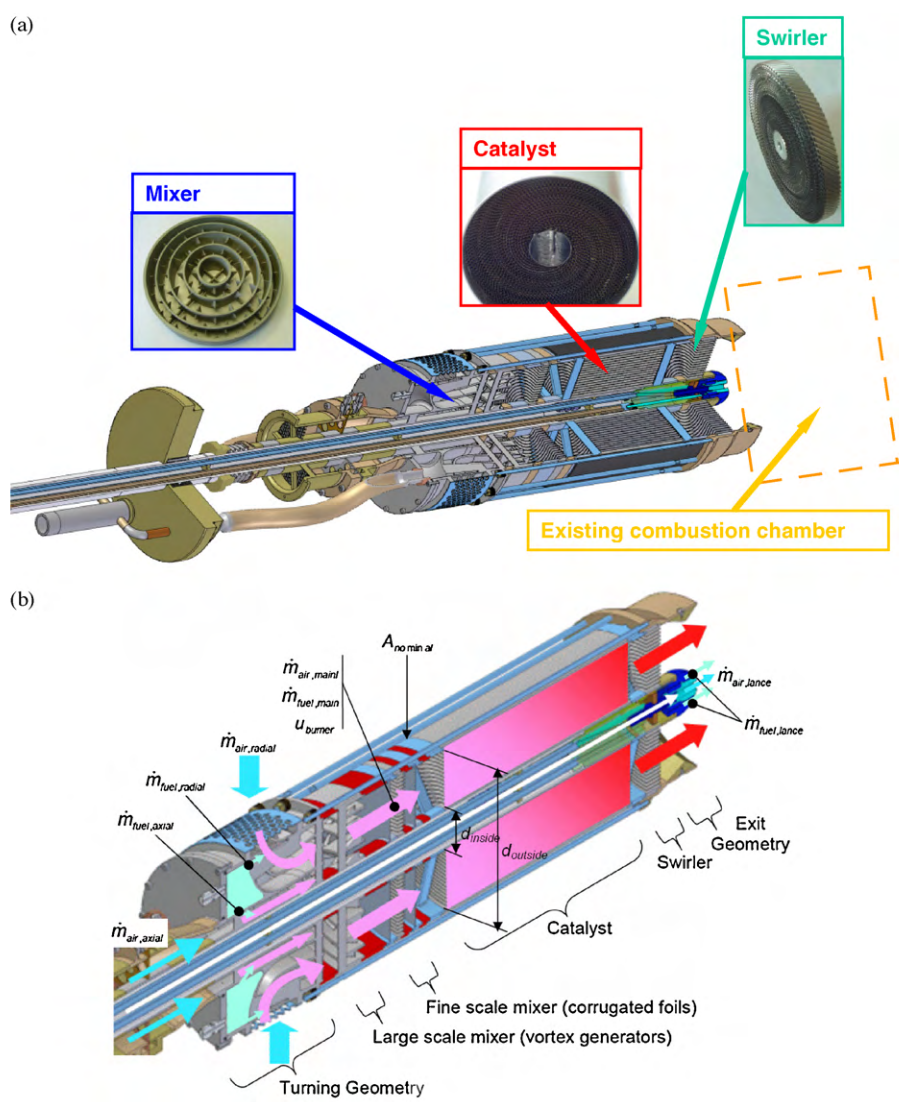
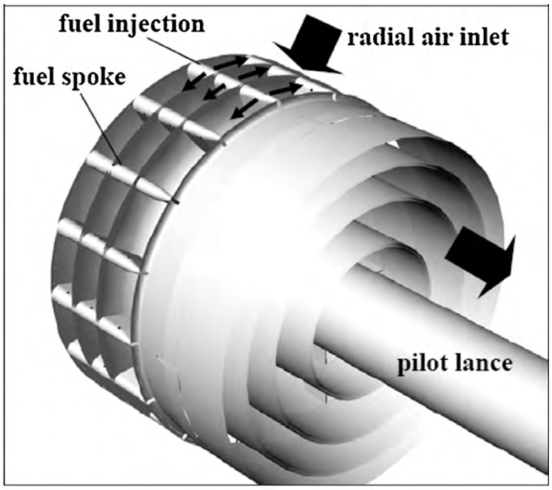


Fig. 5. (a) CATHLEAN hybrid catalytic burner design with main system components. (b) CATHLEAN hybrid catalytic burner: key operation parameters. (c) Flow turning geometry and fuel injector. (d) Selected vortex generator geometry for large-scale mixer.

(c)



(d)

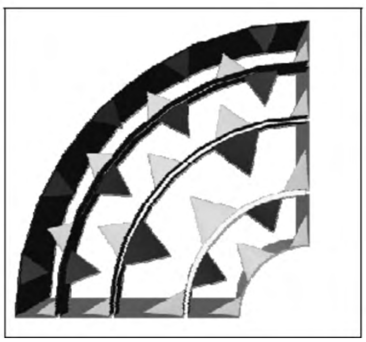


Fig. 5. (*Continued*)

focused on the aerodynamic design of the hybrid burner, including the fuel/air mixer, swirl generator, piloting lance and design of the post-catalyst burnout zone. It also involved the integration of all the catalytic and aerodynamic devices to form the full-scale, hybrid burner, and its subsequent atmospheric testing. WP 6 dealt with the integration of the burner concept within gas turbines to determine boundary conditions for design and testing, as well as developing operating concepts. Overall project management was the focus of WP 1.

The project consortium consisted of organization from six European nations. Fig. 4 provides an overview of the key partner activities.

4. Results

The CATHLEAN hybrid burner design is shown in Fig. 5a and b. Details on the specific burner design used for full-scale atmospheric testing have been given elsewhere [24]. The catalytic reactor is positioned within an annulus surrounding the central fuel-piloting lance. One of the main features of the original CATHLEAN burner design is the multifunctionality of the catalyst section.

In this design there are no moving parts to regulate the bypass of air around the catalyst. Bypassing can be used to increase the fuel concentration within the catalytic reactor to compensate for catalyst aging. With the CATHLEAN design aerodynamic stabilization compensates for catalyst deactivation insuring burner operation. The swirler located at the outlet of the catalyst bed creates sufficient swirl to stabilize the downstream combustion. The high velocities (dictated by the boundary conditions associated with retrofitting to a high-power density, gas turbine, lean-premix burner) within the CATHLEAN design place great demands on the catalyst activity and lifetime.

4.1. Mixer studies

Both large- and fine-scale mixer elements were designed by Alstom and Ruhr University, Bochum (RUB). A specific, flow correcting geometry promotes a near-uniform velocity distribution at the catalyst inlet and includes spokes for fuel injection, as indicated in Fig. 5c. Following the flow turning device large-scale mixing was enhanced by the presence of vortex generators within the concentric flow channels, as shown in Fig. 5d [20]. The details of the mixer design were optimized in a 3D-RANS-CFD study [20] to attain a spatial mixing parameter (% standard deviation from median fuel concentration) of 3.3% at a mixing length of $L/D = 0.75$. This value corresponds to an adiabatic flame temperature variation of 1808 ± 32 K and met the design target of 1808 ± 50 K. The calculated pressure drop was also well below the design target of 0.5%.

A fine-scale mixer, comprised of concentric channels formed by corrugated foils of opposite swirl orientation [23] was proposed and constructed. Due to resource constraints this design could not be optimized for the full-scale tests and was replaced by two wire mesh screens separated by 20 mm, placed downstream of the large-scale mixer.

4.2. Selection of catalyst design

At Alstom a modular, lab-scale high-pressure (*Minihochdruckanlage* MHD) test rig (rated for 30 bar operation, described in Ref. [25]) was used to screen and select the best candidate catalyst systems and, additionally, provided targets for further catalyst and reactor developments. This rig consisted of an electric heating unit, a mixing section, a catalyst test module, a burnout zone and a cooling/exhaust pipe (see Fig. 6). Three 30 kW electrical-resistance heaters preheated air to the desired temperatures. A 30 cm long series of static mixers allowed for high levels of fuel/air mixing, which is a prerequisite for stable catalyst operation without the development of hot spots. The test section and burnout zones were lined with ceramic insulation in order to promote near adiabatic operation. System pressure and velocity were regulated via a throttle, which insured that the flow was always choked, thereby simplifying the relationship between velocity, pressure and mass flow.

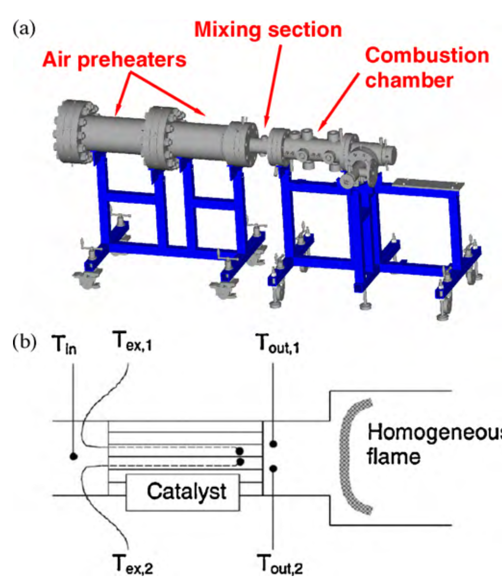


Fig. 6. (a) High-pressure test rig configuration. (b) Schematic of the catalytic test module, indicating the temperature measurement locations. Test rig operating parameters: maximum flow rate 250 g/s, pressure: 1–35 bar, maximum thermal power: 300 kW, air preheat temperature: 0–550 °C.

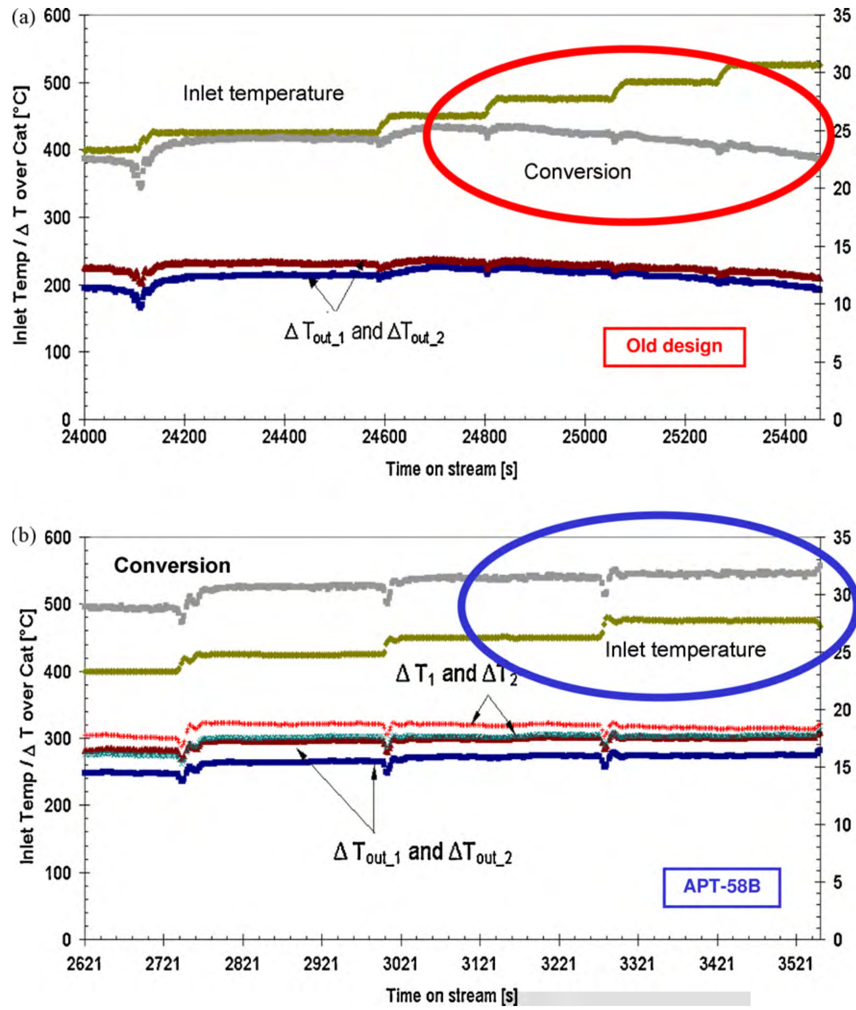


Fig. 7. Catalyst performance test with (a) old Pd/ZrO_2 design and (b) improved catalyst design at 5 bar pressure.

A homogeneous flame converted the remaining fuel in a post-catalyst combustion zone, positioned downstream of the catalyst. Anchoring of the homogeneous flame was attained via the recirculation zone created by a sudden expansion (area expansion ratio = 3.8). A 100 mm gap separated the catalyst exit plane from the expansion to minimize the impact of the flame on the catalyst.

Fig. 6 shows the Alstom test rig and the design of the catalyst bed within the reactor, with temperature measurement locations. All temperatures were measured with K-type thermocouples. The temperatures $T_{out,1}$ and $T_{out,2}$ (located approximately 10 mm downstream of the catalyst) provided the most reliable measurement of the overall catalyst exit temperature and these values were used to compute the temperature rise over the catalyst ($\Delta T_1 = T_{out,1} - T_{in}$ and $\Delta T_2 = T_{out,2} - T_{in}$).

The catalyst composition and design could be optimized by employing the MHD test rig at Alstom. (Numerical modeling of the catalyst reactors also provided valuable design insight as discussed in Ref. [25].) Tests were performed with methane fuel at air excess ratio, λ, (reciprocal of equivalence ratio, $\lambda = 1/\phi$) of 2.5 and at velocities of 15 and 30 m/s; results at 5 bar pressure are shown in Fig. 7. With the first design (Fig. 7a) containing Pd/ZrO_2 catalysts, catalyst deactivation occurred as the inlet temperature was raised above 450 °C. This deactivation was more pronounced at lower operating pressures such as 5 bar and was attributed to the PdO → Pd transition, which occurs when the catalyst reaches temperatures greater than 700 °C at 5 bar pressure. With a new design, which had a lower percentage of its surface area coated with active catalyst, in order to improve thermal management (Fig. 7b), the deactivation could be suppressed and stable operation was possible yielding >30% fuel conversion at inlet temperatures above 400 °C. This improved design was selected for high-pressure aging studies at IFP.

4.3. Aging studies of selected catalysts

At IFP a high-pressure test rig was used, which can operate with natural gas at pressures up to 20 bar, temperatures up to 1300 °C and air flow rates up to 250 kg/h [26]. The IFP test rig can accommodate catalyst honeycombs of 4 cm outside diameter and 30 cm length.

The most promising ignition catalyst design (APT-58, comprised of 10 wt% Pd on a ZrO_2 support and a 50 μm Fecralloy substrate) was chosen for high-pressure testing at partner IFP. The first 1000 h aging study was performed at 10 bar pressure, 15 m/s inlet velocity, $\lambda = 3.2$ ($\phi = 0.3125$), and with an inlet temperature of 450 °C, conditions which produce, initially, an outlet temperature of 650 °C. The results are shown in Fig. 8. The results indicate the large deactivation during the first 200 h of operation, with more or less stable operation thereafter. After the first 200 h the catalyst exit temperature was only 510 °C at 450 °C inlet and $\lambda = 3.2$.

This first aging test was performed at very conservative conditions, with equivalence ratios much lower than those expected for the gas turbine CATHLEAN burner. Thus, it is not known whether the loss in activity during the first 200 h would have been severe enough at higher fuel concentrations.

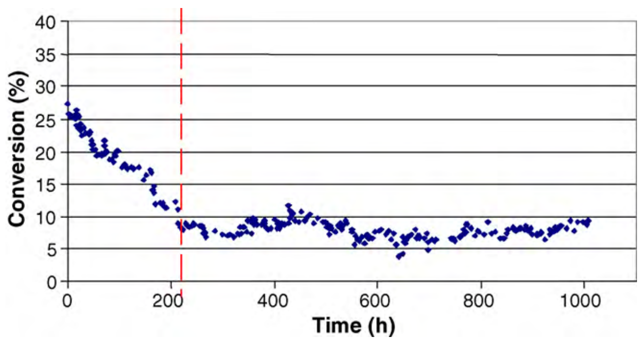


Fig. 8. First 1000 h aging test performed at ϕ = 0.33, 10 bar pressure, 15 m/s inlet velocity and 450 °C inlet temperature.

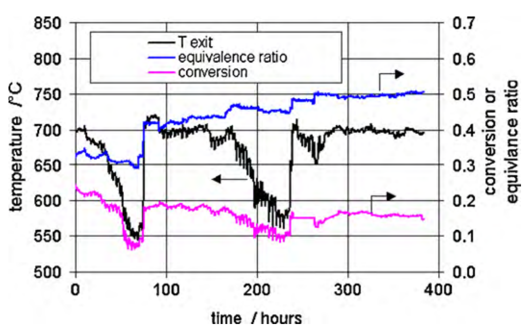


Fig. 9. Second 1000 h aging test performed at 10 bar pressure, 15 m/s inlet velocity and 450 °C inlet temperature. The outlet temperature was maintained roughly constant by increasing the fuel/air equivalence ratio.

In the second aging test the APT-58 catalyst outlet temperature was maintained more or less constant by increasing the fuel/air equivalence ratio from ϕ = 0.3125 up to roughly 0.5 during the aging process as indicated in Fig. 9. In this way the catalyst exit temperature could be maintained at 700 °C for an additional 500 h. During the first 30 h the temperature dropped only slightly, while the equivalence ratio was held approximately constant. During the time windows: 30–75 and 180–230 h, the exhaust temperature and conversion dropped sharply; these time windows corresponded to weekend operation during which it was not possible to regulate the fuel concentration. During the deactivation process pronounced oscillations of the catalyst exit temperature (up to 30 K with a period of 1–4 h) were evident and the catalyst reaction rate changed from being diffusion-controlled to kinetically controlled. A plausible explanation for such oscillations is the transition from PdO → Pd → PdO. Once aged, the light-off catalyst shows stable operation without overheating. The catalyst activity was stable during the last 250 h of operation, and the fuel concentration could be maintained at nearly constant values.

The following conclusions can be made from the aging study:

- The most significant catalyst aging occurs during the first 200 h of operation. Partner IFP were able to model their aging results using two first order decay processes, whose time constants were found dependent on the catalyst operating temperature, decreasing at higher temperatures.
- Catalyst design should be made based upon aged (at least 200 h), and not fresh catalyst performance. (The aging should be performed at the expected operating conditions of the catalysts.)

4.4. Full-scale atmospheric tests

The full-scale CATHLEAN burner was tested at atmospheric pressure at the DLR (German Aerospace Center) in Cologne. Selected operating conditions are summarized in Table 3. Fig. 5b indicates the main burner parameters, which were optimized for stable operation. The fuel split (α) and air split (β) indicate which proportion of the total flows, respectively, are injected through the central lance to create a "piloting" of the combustion. The air feed through this pilot was at ambient temperature.

The fine-scale mixer, in the original CATHLEAN concept constructed from corrugated catalyst support foils, was simulated with two sieves since time constraints did not permit several mechanical integrity issues to be fully addressed. Such a solution was adopted, despite anticipated elevated pressure losses, in order to provide uniform fuel/air mixing at the catalyst entrance. High-quality mixing was necessary in order to experimentally assess the contribution of the catalytic reactor to the combustion performance of the full-scale hybrid burner.

Based upon the aging studies, a new reactor design was chosen, which was comprised of three stages: inlet, medium and high temperature with a total length 67% greater than that tested during the aging. The purpose of this design was to ensure reasonably stable and constant catalyst performance during the testing of the full-scale hybrid burner. The catalyst bed used is shown in Figs. 10 and 11, and was constructed from corrugated Fecralloy support foils, rolled up to form a honeycomb. (The use of metal substrates allowed much greater freedom to optimize the geometry and to achieve multifunctionality.) The honeycomb was divided into three axial sections, as indicated in Fig. 11, each with different surface area fractions that were coated with active materials Pd/ZrO_2 or $Pt/MI368\text{-}Al_2O_3$.

The catalyst design employed the concept of "passive cooling", achieved by coating less than 100% of the Fecralloy substrate surfaces with active material, in order to prevent the catalyst from reaching the adiabatic flame temperature of the fuel/air mixture. To lower the risk of catalyst overheating and failure, and in an effort to ensure constant catalytic performance, the maximum catalyst bed exit temperature (T_3) was limited to a nominal 1100 K during the testing.

Table 3
Overview of atmospheric testing at DLR Cologne.

	Operation points	Characterization without catalyst	Characterization with catalyst	Comparison with and without catalyst	
		A	C	F	G
Fuel split (%)	α	0, 2.5, 5	0, 2.5, 5, 10	1.5	1.5
Air split (%)	β	2	0, 1, 2	1.5	1.5
Air inlet temperature (K)	T_3	673	673	690	690
Lambda	λ	2.2 1.7–3.1	2.6	2.4	2.0
Equivalence ratio	ϕ	0.45 0.59–0.32	0.38	0.42	0.5
Adiabatic flame Temp. (K)	T_{ad}	1700 1929–1433	1558	1621	1779

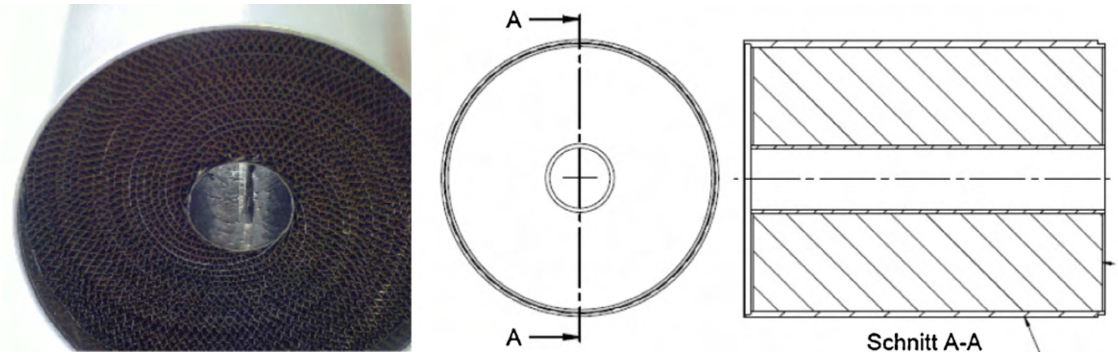


Fig. 10. Photograph of the catalyst honeycomb (left) and detail drawing of the bed.

The catalyst total bed length was increased in order to account for the observed aging. This led to the higher pressure drops measured. The commissioning tests for the burner indicated a pressure loss of 10% for the catalytic reactor at the design burner outlet velocity. Clearly, the pressure drop target for the catalytic reactor was not reached. The focus of future development must be to enhance catalytic activity and simultaneously reduce reactor length in order to satisfy the pressure drop specification. Furthermore, the sieves used to simulate the fine-scale mixer and guarantee a high uniformity of fuel concentration lead to a 14% pressure drop. It is expected that the fine-scale mixers will result in a markedly smaller pressure loss than the temporary sieve solution currently adopted.

4.5. Comparison of burner performance with and without catalytic conversion

The main purpose of the atmospheric pressure tests with the full-scale CATHLEAN burner was to quantify the combustion performance improvements due to catalytic conversion. This required tests to be initially carried out without any catalyst present. For these comparative tests, the catalytic honeycomb was replaced by spacers and four sieves which created an equivalent pressure drop.

Fig. 12 shows the burner operation without catalyst conversion as a function of the pilot fuel amount. Also indicated are the lean blow-out (LBO) limits shown as vertical dashed lines. It can be seen that increasing the pilot fuel allows operation at leaner fuel/air ratios (greater λ values) at the cost of greater NO*x* emissions. Without pilot fuel, lean blow-out occurred already at $\lambda = 2.2$ yielding approximately 3 ppm NO*x*. At $\lambda = 2.6$ operation was only possible with pilot fuel; a value of $\alpha = 2.5\%$ yielded approximately 3.2 ppm NO*x*, but with high CO emissions. Employing $\alpha = 5\%$ shifted lean blow-out to $\lambda = 3.2$, and was accompanied by very high CO emissions.

Results with catalytic conversion and various levels of pilot fuel are given in Fig. 13. (It was found that variation of the pilot air amount (β) had no significant influence on the flame stabilization process.) Stable operation without pilot at $\lambda = 2.6$ was unfortunately not possible. As described in Diers et al. [24] the flame was unstable, with the flamefront intermittently moving back and forth from the lance tip (at the burner outlet) to a downstream location. It was found that an optimal trade-off between operating stability and NO*x* emission occurred at $\alpha = 1.5\%$. Here it is seen that at $\alpha = 2.5\%$ 2–3 ppm NO*x* with equally low CO emissions were observed.

The results of these tests highlighted the problem of comparing performance with and without catalytic conversion. As indicated in Fig. 14 the burner LBO limit without catalyst occurs at a relatively low λ value; at this level of fuel concentration the catalyst exit temperature maximum value of 1100 K was reached. To reach operating

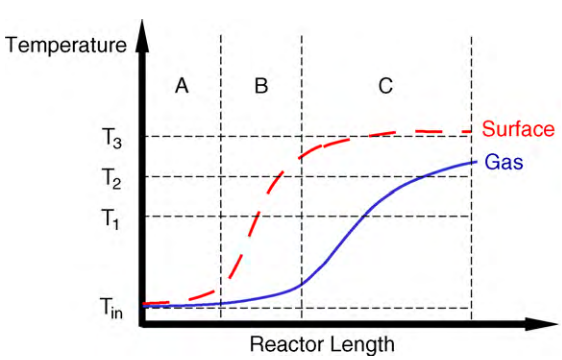


Fig. 11. Estimated gas and surface temperature distributions along the length of the catalytic reactor. Each sector of the catalyst has a different catalyst composition and coating fraction.

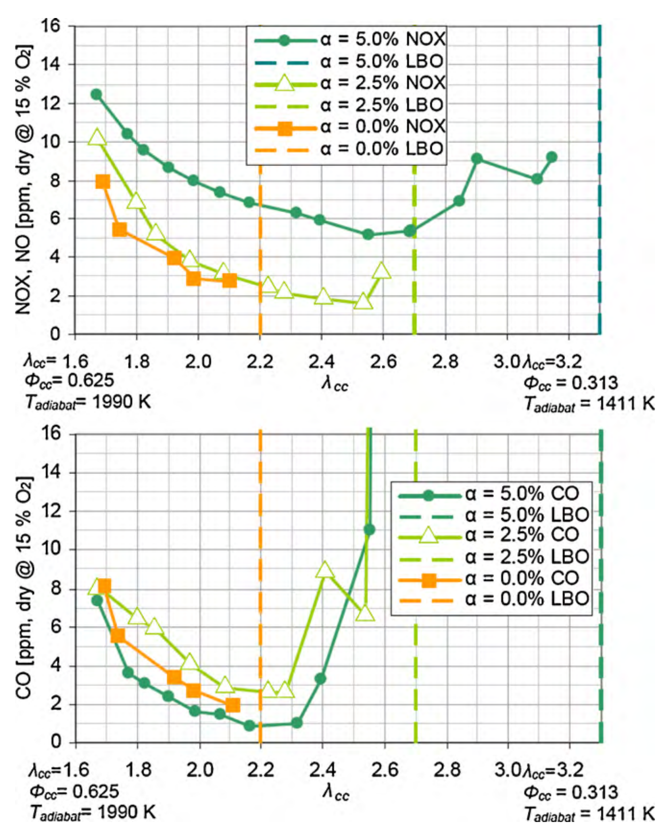


Fig. 12. Exhaust gas composition of the burner *without* catalyst at operating point A measured 600 mm downstream of burner exit.

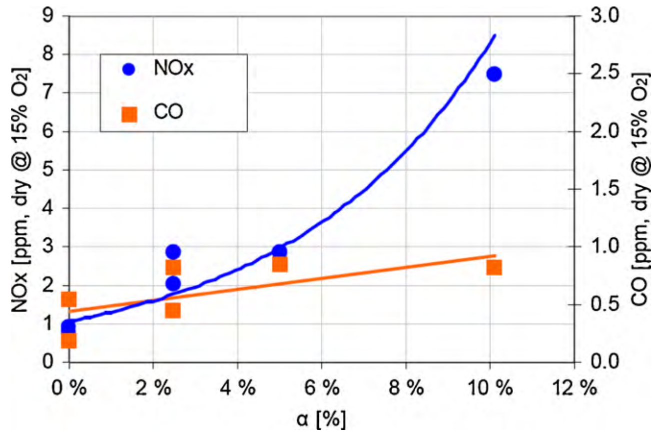


Fig. 13. Exhaust gas composition of the burner *with* catalyst at operating points C measured 600 mm downstream of burner exit.

conditions that would be stable for the burner with and without catalyst conversion the following changes in operation were made:

- The maximum allowable catalyst exit temperature was increased from 1100 to 1160 K.
- The optimal pilot air and fuel splits (α, β = 1.5%) were used.

It was also found that the catalyst aged somewhat during the first tests performed. Fig. 15 shows changes in the outlet temperature profile of the catalyst bed between the first and second test campaigns. For this reason the air inlet temperature was increased from 673 to 690 K. The dependence of the catalyst exit temperature on excess air ratio and burner velocity is shown in Fig. 16. A comparison of the 100% burner velocity data to those in Fig. 14 indicates the aging that took place.

Operational comparison was made at points F and G of Table 3. Fig. 17 shows the NOx production for all four operation points. To compare the emissions, radial profiles of exhaust gas concentrations were measured at two downstream axial locations (measured from the burner exit plane). Emission values in Fig. 17 are aver-

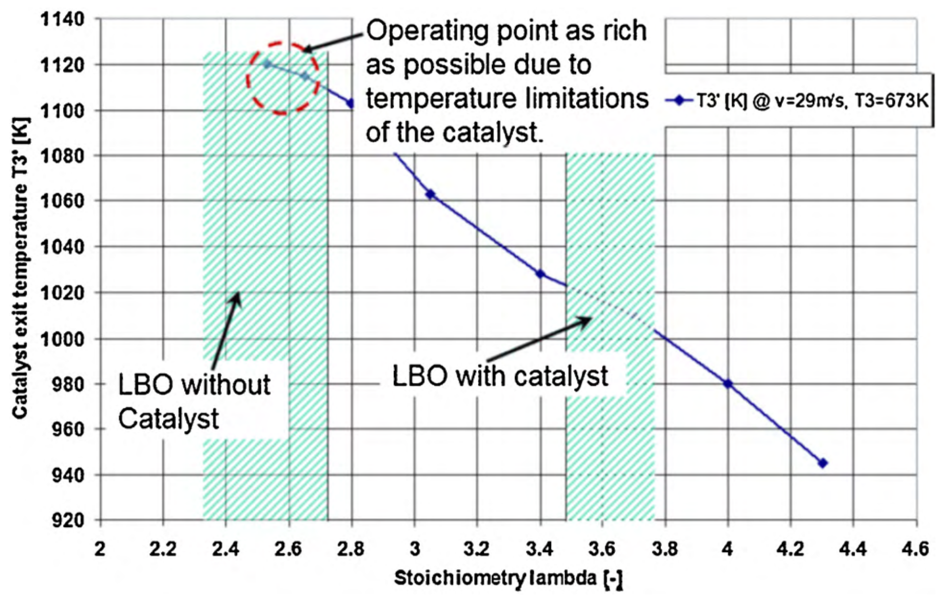


Fig. 14. Operating limits of the CATHLEAN burner with and without catalyst at an air inlet temperature of 673 K, burner velocity of 29 m/s.

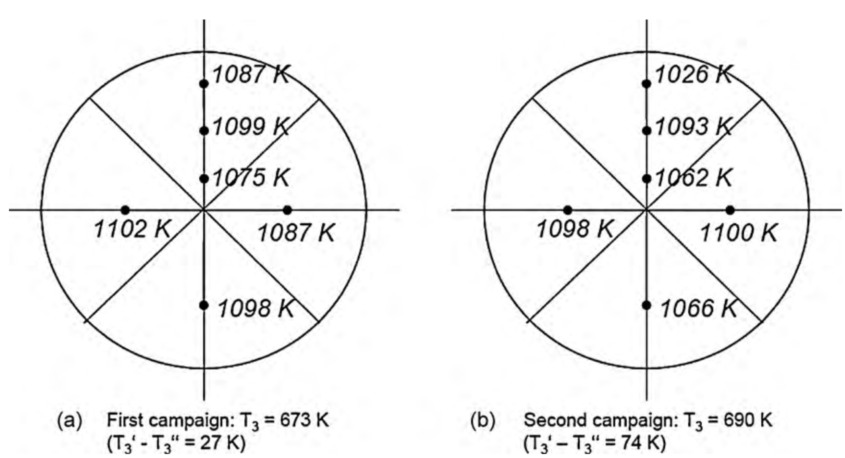


Fig. 15. Exemplary catalyst bed exit temperatures for the two measurement campaigns.

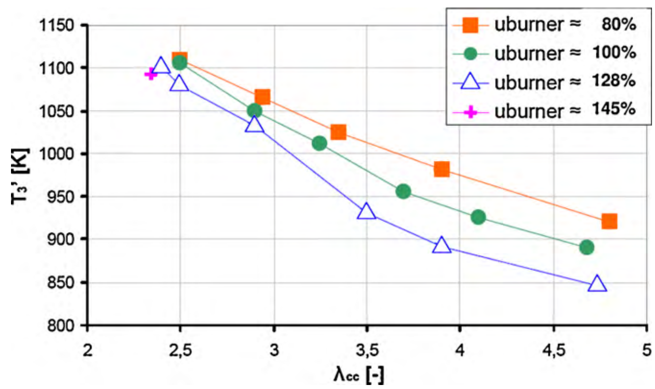


Fig. 16. Catalyst exit temperatures as a function of air excess ratio and burner velocity for an air inlet temperature of 690 K.

aged (using area-weighting) from radial measurements 650 mm downstream of the burner exit.

5. Discussion and conclusions

The main goal of the project was to assess the catalytic contribution to combustion performance, applying boundary conditions derived directly from state of the art, lean-premix, gas turbine burners. From the outset, it was clear that the overall challenge lay in developing demonstrator technology that combined the directly conflicting requirements of catalysts (low space velocities, low peak temperatures, large surface areas) and gas turbine combustors (high-power density, elevated temperatures, low pressure losses, robustness, maximum lifetime and reliability). The overall project target was achieved; the main results and findings are discussed below:

- The full-scale atmospheric pressure tests performed on the CATHLEAN burner highlighted the positive impact of catalytic fuel conversion on performance. The lean blow-out limit was extended by ca. 200 K and the NO*x* emissions were reduced by approximately 25% at conditions close to full-load, characteristic of a modern, high-power density, gas turbine burner.
- As expected, catalytic conversion of a portion of the methane fuel acted to reduce NO*x* emissions. Using the measured average catalyst exit temperature the catalytic conversion may be estimated. Assuming adiabatic conditions within the catalyst (a good assumption for the inner channels of the catalyst) the conversion is calculated as the ratio of the temperature rise within the catalyst to the total possible temperature rise (based on the calculated adiabatic flame temperature of the mixture). The results are summarized in Table 4, showing roughly 25% conversion for the two states. For the higher flame temperature, 1779 K, close to the base load operating point of a gas turbine, the NO*x* emissions are reduced by 27% (from 4.1 to 3 ppm). For a flame temperature of 1500 °C, the work of Schlegel et al. [14] (see Fig. 18) predicts a smaller decrease of NO*x* emissions at this level of catalytic conversion. Calculations of the authors in which the impact of catalytic exhaust upon NO*x* formation was assessed for a reactor model simulating a gas turbine combustor (see Fig. 19) indicate roughly 30% reduction in NO*x* emissions obtained for a case with 25% catalytic conversion, which is in line with the present experimental results. The reactor model simulated the staged combustor as a combination of a catalytic stage, a perfectly stirred reactor and a plug flow reactor as indicated in Fig. 19.
- It is expected that NO*x* emissions could be further improved with an optimized design of the central pilot, as spatially resolved measurements of the emissions indicated that a significant portion of the NO*x* emission is produced here. Although the pilot was required for flame stabilization at atmospheric pressure even with the catalyst, chemical kinetic calculations of the ignition delay times suggest that flame stabilization should be possible without the pilot at high-pressure conditions.
- The results of the project highlight the importance of catalyst aging in design. Fresh catalysts were employed in the full-scale tests and showed aging during the course of testing. In the future the design process needs to incorporate deactivation processes and “pre-aged” catalysts should be installed into components for their characterization. Considering the very long aging process (on the order of 1000 h) full-scale aging of catalytic components under actual operating conditions is not feasible. These processes

Table 4
Comparison of burner operation with and without catalytic conversion.

Operating points	State F		State G	
	w/o catalyst	Catalyst	w/o catalyst	Catalyst
Catalyst inlet Temp. (K)	690	690	690	690
Catalyst outlet Temp. (K)	–	100	–	1160
Catalytic fuel conversion (%)	–	25.5	–	26.4
Adiabatic flame Temp. (K)	1621	1609	1779	1779
NO*x*	1.9	1.0	4.1	3.0

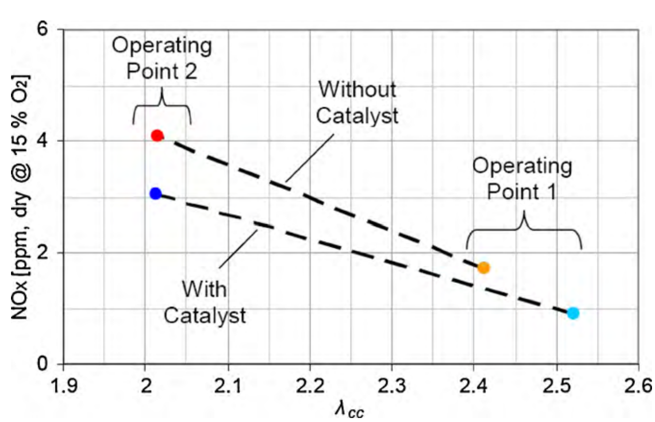


Fig. 17. NO*x* concentration values (averaged from radial measurements across the combustor cross section, 650 mm from burner exit) with and without catalyst conversion (operating points F and G from Table 4).

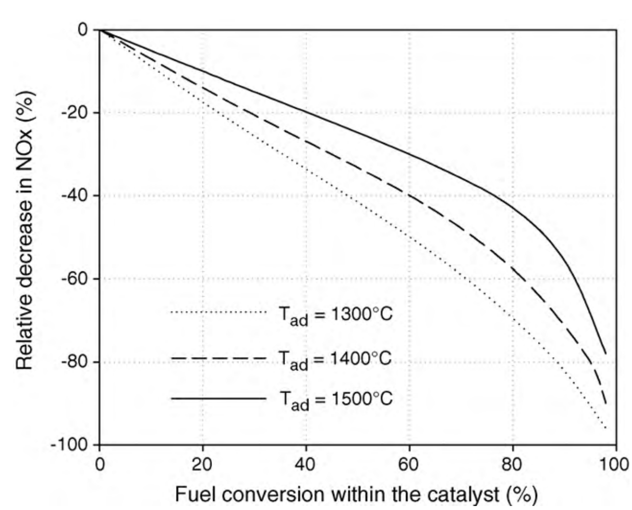


Fig. 18. Computed relative decrease in NO*x* emissions as a function of the fuel conversion within the catalyst, for different adiabatic flame temperatures (adapted from Ref. [14]): CH_4/air, atmospheric pressure, inlet temperature 380 °C and residence time 20 ms.

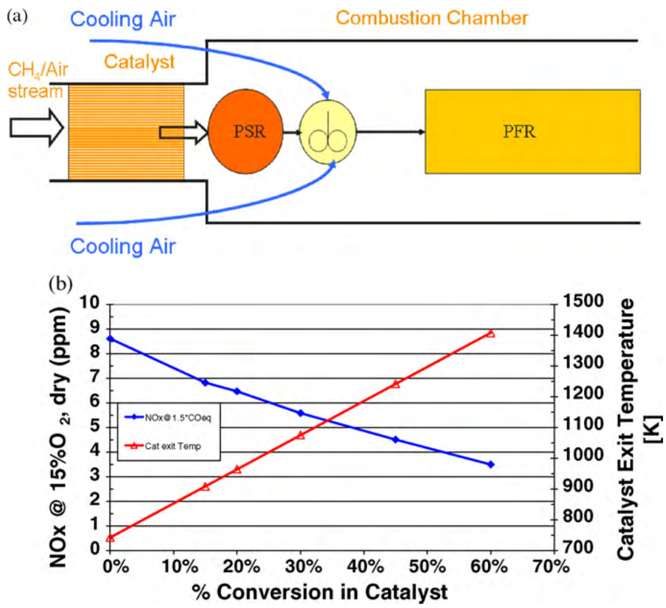


Fig. 19. Computed relative decrease in NO*x* emissions as a function of the fuel conversion within the catalyst, CH_4/air, adiabatic flame temperature of 1800 K. (a) Reactor model employed for kinetic calculation. (b) Effect of catalytic conversion on NO*x* emissions. This calculation indicates a reduction of NO*x* by ca. 30% for a case with 25% catalyst conversion.

need to be investigated in further detail and aging models should be refined, in order to define proper "pre-aging" or "accelerating aging" procedures which could be employed.

- The performance of the burner was promising, with encouraging catalytic conversion at simulated gas turbine conditions. The catalysts did work quite well, with reasonable conversion (up to the imposed maximum limit, dictated by maximum allowable catalyst temperature). There was measurable improvement in performance when the catalyst converted fuel, even though the central pilot had to remain in operation for a stable flame position.
- The burner did function cleanly and stably without catalyst conversion, using the standard aerodynamic flame stabilization. This suggests that the hybrid approach, which is most compatible with the requirements of large gas turbines, is feasible.
- Unfortunately, some components of the CATHLEAN burner could not be tested, including the fine-scale mixer, the integration of catalyst into the aerodynamic swirler and pilot. The optimization of these elements could act to reduce considerably the pressure drop required for mixing and flame stabilization.
- The impact of imperfect mixing on catalytic performance was not tested. Trade-offs between pressure drop and mixing quality (catalyst overheating) need to be investigated. Concepts for integrating mixing and catalytic conversion (catalytic coating of the fine-scale mixer, adding holes in Fecralloy catalyst substrates, allowing for mixing zones between individual catalyst beds) should be further developed.
- Risks still exist including catalyst overheating, and the operational complexity associated with the pilot. The catalyst reactor needs to be modified to reduce overall pressure drop and to better account for aging.

Whilst the most important technical goals outlined in Section 2 above were met, it should be noted that new market demands arose during the course of the project, which have made the challenge of introducing a catalytic combustor to the gas turbine market more difficult. These include: fuel flexibility requirements (from higher hydrocarbons ("C2+") to hydrogen-based synthesis gases), higher firing temperatures, higher power density, and the greater demands on component part lifetime and lower cost. Since catalysts are more or less optimized for one fuel type, fuel flexibility is a great challenge for catalytic combustion. These market demands are often more important to the customer than ultra-low NO*x*.

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