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Development of a spatially uniform low-temperature hydrogen combustor[#]

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Abstract: Examples of the use of additive manufacturing and rapid prototyping in a range of applications are of great interest in order to emphasize their role in development and production technology. In this study, a catalytic low temperature burner for H_2 on a lab scale with an integrated flow distributor was designed, manufactured, and tested for functionality. Based on a theoretical approach, a flow distributor for the burner was designed and a prototype was built using fused deposition modeling (FDM). Based on test results, an optimized version of the burner was then designed and manufactured using selective laser melting (SLM). The functionality of the designed catalytic burner was proven. Several advantages were found in comparison to conventional non-catalytic burners. In particular, flameless uniform low temperature heat generation with temperatures of about 200 °C could be realized. This contribution highlights the potential of additive manufacturing in chemical engineering. Not only was the final product built using SLM, but also during the development process, FDM was used for rapid prototyping.

Key words: Selective laser melting; Catalytic burner; Laboratory scale; Flow distributor; Hydrogen combustion
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1 Introduction

Since their invention in the 1980s, rapid prototyping technologies have become increasingly important in product design and development and for additive manufacturing (AM) technologies (Gardan, 2016). However, conventional production technologies are often still less costly when the size of the workpiece is large and/or the complexity is low (Mahamood et al., 2014). Applying AM usually requires a redesign of the workpiece and new design skills to fully exploit the benefit of this technology, namely a substantial reduction of processing steps and required parts. Since in many cases the product layout is still dominated by the ability of conventional manufacturing, a diffusion of AM into more application fields and industrial branches requires proven examples and design rules derived from them. The aim of this study was to elucidate such an example of potential use: a burner for the spatially uniform low temperature combustion of hydrogen on a lab scale.

1.1 Practical background

Hydrogen is an important energy carrier and feedstock for the chemical industry. Presently, most of the roughly 20 Mrd. m³ hydrogen generated in Germany each year (Winter and Nitsch, 2012) is based on cracking and steam reforming of fossil fuels (Yoshimura et al., 2000; Alves and Towler, 2002). If

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in the future more hydrogen will be produced from wind and solar power via electrolysis, efficient ways of using hydrogen from fluctuating sources will be required. Hence, there are many different research projects, in which hydrogen is being used as feed and/or occurs as a product off-gas.

In particular, Kelling et al. (2015, 2016) developed a ceramic counterflow reactor for the heat-integrated generation of synthesis gas ($CO+H_2$) from CO_2-rich feedstock. In their experimental setup on a lab scale, a constant flow of H_2-rich off-gas occurred that would need suitable post-treatment.

Burning of excess hydrogen accruing during chemical processes is an obvious solution, but the large explosion limits of hydrogen/air mixtures, and high flame temperatures and associated NO_x formation are drawbacks not only in industry, but also in closed laboratory premises.

Kelling et al. (2016) therefore used a dynamic dosage system, in which compressed air was used to dilute the off-gas below the explosion limit depending on the occurring H_2 mass flow. Although functionality was proven, the constant need for large amounts of compressed air, the complexity, and the necessary dynamic control algorithm for air dosage are disadvantages of this concept.

Based on these considerations, in this study we developed a catalytic burner for flameless hydrogen oxidation at low temperatures for lab-scale applications.

1.2 State-of-the-art hydrogen combustors

Several inventions have already been made in the field of hydrogen combustion on a lab scale. Overall, they differ in various ways such as:

(1) the combustion method (flaming or flameless/catalytic);

(2) the oxygen supply (pre-mixed in hydrogen or by feeding of secondary air);

(3) the combustion management (single- or two-step combustion);

(4) the purpose of combustion (heat generation or hydrogen elimination).

With respect to the method of combustion (1), the most commonly used method on a lab scale is a conventional excess gas burner in which hydrogen is burnt with an open flame. Although the implementation is certainly simpler than that of more advanced solutions, the occurrence of extremely high temperatures, the open combustion, and the potential formation of noxious NO_x in closed premises are evident trade-offs that could instead be avoided using flameless, catalytic combustion.

The oxygen supply (2), which is needed for H_2 oxidation can be supplied either by pre-mixing it with the hydrogen or by feeding it directly into the combustion zone while separating both substances as long as possible. The latter will always be the choice in this application because meta-stable H_2/O_2 could easily flashback into the supply and cause severe damage.

To ensure complete combustion and/or lower the temperature in the system, the process of hydrogen oxidation could be conducted in more than one step (3). A certain amount of the initial H_2 feed could be oxidized in the first step, lowering the temperatures compared to those of a complete one-step combustion. In the second step, and in a spatially separated area, the residual hydrogen could then be oxidized. Although better controllability of combustion and lower temperatures are obvious advantages, this approach greatly increases the complexity of the process as O_2 dosage to all separate combustion steps needs to be continuously controlled based on the current total H_2 feed.

Many publications that address oxidation of hydrogen have focused on H_2 usage as a gaseous fuel for heat generation rather than hydrogen elimination (4). Obviously, high temperatures and low energy losses are desirable in this context, but not in the context of our study.

Based on the variants described above, the hybrid atmospheric burner combined with a catalytic element proposed by Allouis et al. (2014) and Cimino et al. (2010) appears to be the most similar to this field of application. The overall size of their cylindrically shaped burner is 80 mm×80 mm and the manufacturing was carried out using only conventional machining methods. In this setup, in the first step, the feedstock is catalytically oxidized and in the second step, flame combustion takes place using secondary air for complete hydrogen oxidation. To withdraw the reaction heat from the system and thereby keep the burner at low temperatures at which NO_x formation is low, a heat exchanger for heat transfer by radiation surrounds the reactive zone.

This system is without doubt elaborate, very sophisticated, and even patented (Claudio et al., 2009). However, the occurrence of hot-spots and the overall high temperatures in the setup demand a re-design and have led to a search for alternative concepts, especially those in which AM as a potential production process is considered directly from the beginning.

1.3 Development objective

These initial considerations led to the formulation of the following requirements of a new concept:

1. Flameless, catalytic combustion for safety reasons;
2. Low temperatures to ensure the minimum NO_x formation;
3. Complete combustion to harmless products (minimization of residual hydrogen);
4. Single-step combustion to ensure simplicity of usage;
5. Continuous and robust operation for variable H_2 dosing;
6. Heat removal solely by free convection;
7. Compact design.

Although irrelevant in the case of H_2-only combustion, low temperatures would also be very desirable if the burner use-case was to be extended to a carbon containing feedstock, as it is well known that at temperatures above 450 °C metal dusting could potentially occur and damage the component (Grabke, 2003).

If a prototype catalytic burner based on the fractalized fluid flow concept is to be developed, the strengths of AM need to be exploited:

1. A hands-on prototype of a new geometry can be manufactured at negligible temporal and financial cost using fused deposition modeling (FDM);
2. The actual metal flow distributor for the test rig can then be manufactured using selective laser melting (SLM);
3. Future development steps of the burner and design variants could also be built very quickly;
4. Further fractalizations can easily be integrated in the design without jeopardizing the manufacturing process.

The desire for a compact and potentially complex device emphasizes why AM was considered the preferred production process from the beginning.

2 Preliminary design

The starting point of the design was the consideration to distribute hydrogen evenly over a catalytic surface. For practical reasons the area was set to a maximum of 80 mm×80 mm. Considering the lower heating value (LHV) of hydrogen (12.75 MJ/m^3), an energy density of 14.03 kJ/mm^2 can be expected at a maximum flow rate of 0.5 NL/min of hydrogen.

2.1 Development of base geometry

Based on this estimation, a first draft was considered, in which the feed stream leaves a reservoir through a large number of small openings (Fig. 1).

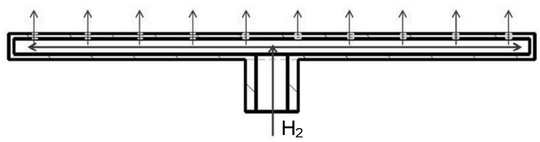

Fig. 1 First sketch of the hydrogen distribution unit involving a hollow plane

However, backmixing of air and hydrogen in the reservoir cannot be excluded, especially after shut-down of the hydrogen feed. This increases the risk of explosion in the open space of the reservoir, as an external ignition of the H_2/O_2 mixture could easily flashback. To minimize this risk, a tree-branched gas distributer structure suggested by Tondeur and Luo (2004) was considered, which drastically decreases the size of coherent cavities.

Tondeur and Luo (2004) discussed a theoretical approach for dimensioning fluid distributors. The fundamental conclusion relevant here is that by successively splitting the stream while simultaneously reducing the diameter an even flow can be achieved in all channels when certain channel lengths are applied (Fig. 2).

Respective correlations were also supplied in their publication. Based on these findings, the existing construction was modified.

According to process limitations, the smallest diameter d_8 of the flow distributor was set to 0.8 mm. Based on this fixed size, the remaining diameters were calculated as follows (the results are shown in Table 1):

$$\frac{d_0}{d_8} = 2^{8/3}, \quad \frac{d_k}{d_{k+1}} = 2^{1/3}. \tag{1}$$

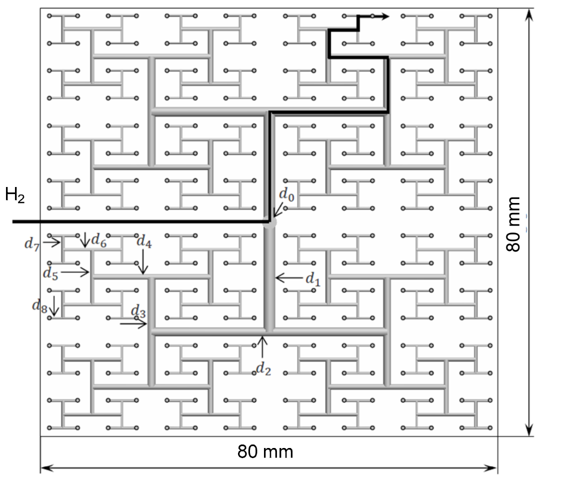

Fig. 2 Illustration of the flow distributor design proposed by Tondeur and Luo (2004). Marked is the H_2 supply at d_0 and one potential flow path that H_2 could take through the distributor

Table 1 Calculated diameters for the distributor (the positions of the pipes are shown in Fig. 2)

Diameter (mm)	Diameter (mm)	Diameter (mm)
d_0=5.1	d_3=2.5	d_6=1.3
d_1=4.0	d_4=2.0	d_7=1.0
d_2=3.2	d_5=1.6	d_8=0.8

Luo et al. (2015) recently compared their earlier proposed scheme to a conventional setup similar to the one shown in Fig. 1, but enhanced by implementation of baffles. Their reason for installing baffles was the higher pressure-drop of the tree-branch design together with fabrication tolerances which can deteriorate the quality of distribution. Nevertheless, in our work we chose the tree-like structure because pressure-drop is not critical in the considered application of hydrogen off-gas combustion in contrast to the demands of minimization of coherent cavities.

To test the functionality of the distribution approach, a first prototype was built using a Makerbot Replicator 2X by means of FDM. Due to the production limitations of the Makerbot, the selected geometry was larger than in the final product. However, the geometry was estimated with the same algorithm (Eq. (1)). Here, the feed diameter was first chosen at a fixed size of 10 mm (compare with d_0 in Fig. 2) and the smaller diameters (d_1–d_4) were calculated from the top down. The geometry is shown in Fig. 3.

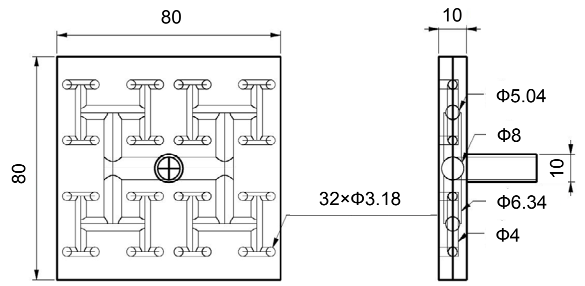

Fig. 3 Sketch of the flow distributor prototype, built using a Makerbot Replicator 2X by means of fused deposition modeling (FDM) (unit: mm)

Several tests were performed, in which smoke was fed through the prototype, and the even distribution over all holes was confirmed visually.

Oxidation of hydrogen is a strongly exothermic process. Thus, heat removal is crucial to avoid overheating of the burner. Therefore, ribs were considered on the backside of the distributor.

To ensure steady state operation without extinction of the oxidation reaction, a permanent supply of oxygen is essential. One possible solution would be the use of a second, identically constructed distributor which could feed secondary air directly into the hydrogen outlet holes of the first distributor. Doing so, the system could be used for other use-cases in chemical engineering in which concrete ratios between the educts have to be ensured. However, this would not only double the production cost (two distributors instead of one), but also require constant adjusting of the oxygen-containing feed. Therefore, a rather simple approach was put into effect, in which the burner gas distributor was placed vertically into a channel through which air flows due to natural convection (chimney effect). Doing so, the additional components needed are of very simple geometries and can hence be produced by conventional means. Furthermore, this approach is self-regulating.

Ambient air contains a high excess of oxygen (~20%), which means that the supply of 'new air' from the bottom of the chimney will in most cases not be the limiting quantity. However, at some point during operation the total amount of O_2 available will be noticeably reduced as oxygen is nevertheless consumed. During operation, the air in the chimney will be heated up by the exothermic process whereby it will ascend to the top. For reasons of continuity, oxygen-rich ambient air will then be drawn inside the chimney from the bottom. As the heat release depends directly on the amount of oxidized H_2, this effect will be the stronger the more hydrogen is converted and hence the more oxygen has been consumed.

The resulting complex structure of the flow distributor highlights the necessity of using AM as the preferred fabrication method. One might argue that the proposed design includes geometric features (e.g. holes and shallow grooves) that seem perfectly suitable for a process chain based on computerized numerical control (CNC) machining and mechanical assembly, which would likely be preferable in terms of accuracy, surface finish, structural integrity, and price.

With conventional production methods at least two parts are required that need to be assembled and sealed off hydrogen-leak-proof in all thermal states of the burner: cold, steady state hot and all intermediate states with non-homogeneous temperature fields and thus thermal warping. The only way to put this into practice would be to use a soft elastic seal which is of limited lifespan and requires maintenance, especially when exposed to hydrogen and high temperatures. Furthermore, surface finish was not considered to be an issue in this particular application as the increased roughness will not deteriorate the burner performance, but will improve the heat removal by increasing the external heat transfer coefficient.

Incomplete hydrogen combustion due to insufficient H_2 residual time at the catalyst can lead to flammable hydrogen that remains in the off-gas. Wolfgang and Josef (1989) addressed this issue for a catalytic heating panel by separately dosing hydrogen and oxygen into a diffusion layer. This has also been adopted here. As in their study, we mounted a highly porous layer (a catalytically activated metal filter) onto the outlet of the tree branch structure. Yet in contrast to their work, only H_2 was fed actively into the filter and oxygen was supplied passively by the chimney effect.

Although hydrogen is convectively transported throughout the filter, the pressure drop leads to a drastic reduction in velocity which results in a sufficient residual time at the catalytic layer.

Diffusive back-mixing of aerial oxygen into the tree structure during times when no H_2 is dosed would also pose a potential risk, as oxyhydrogen is highly explosive. However, this can most likely be excluded due to the diffusion limitation caused by the filter.

The proposed design for the distributor with minimized coherent cavities could also be used for other fields of application such as the gassing of bubble columns, where back-mixing too, can be a potential safety risk.

2.2 Similar applications of AM

With the presented H_2-burner, an example for a class of fluid systems is presented, known as fractalized fluid flow systems, which find applications in all kinds of cooling and chemical reactions. The design is bio-inspired from, for instance, blood vessels progressively splitting up to uncountable very fine ducts. Despite showing here only the upper layers of the fractal fluid distribution, this demonstrates the direction, where only AM processes can help manufacture these systems.

The demand for low temperatures requires a uniform distribution of hydrogen in air over the catalyst. This approach basically demands the manufacture of small channels in a metal flow distributor, with small but homogeneously distributed openings. Additive manufacturing is best suited to manufacturing such structures with complex shaped internal channels, as has been proven for other similar applications.

Both Mazur et al. (2016) and Dimitrov et al. (2010) investigated standard and conformal cooling channels for application in the field of injection moulding tools.

Another example of application optimized cooling channels is described by Mueller et al. (2013), who investigated the potential of AM in respect of the hot sheet metal forming process of press hardening, and by Miksche (2014) who focused on die casting moulds.

In another application, a complex spin pack was successfully manufactured for three different polymers with >14 m of internal heating/cooling channels and a thermal insulation grid (Hufenus et al., 2012).

Furthermore, for fuel combustion on an industrial scale, AM has already been successfully tested by Euro-K using an EOS M 290 3D printer (Euro-K, 2015). Euro-K designed a novel micro-burner with an optimized footprint and functionality which is capable of using gaseous and liquid fuels. It focused especially on the issue of optimizing the surface area increase of liquid fuels to enable ignition. The burner geometry is cylindrical. Fuel is injected inside, where non-catalytic combustion takes place with open flames.

The biggest benefits of this new concept are the flexibility of using liquid and gaseous fuels, the reduction of the combustion chamber size by 20% compared to conventional designs, economical efficiency due to low unit costs despite small batch sizes, and the reduction of NO_x emissions due to comparably low temperatures.

Although having widely proven functionality for combustion scenarios for the envisaged purpose, this concept has several disadvantages. First, as stated, a catalytic, flameless oxidation is clearly preferred for safety reasons. Another drawback linked to open flames would be the reduced robustness in the case of hydrogen feed cutout and subsequent resetting, which would very likely require continuous supervision and some sort of ignition spark.

The proposed setup by Euro-K could be extended by a catalyst to avoid having open flames, and the lower combustion temperatures could decrease the NO_x formation even further. However, catalyst assembly would be far more difficult compared to our flat design.

Furthermore, a scale-up of the burner would be more challenging compared to our flat design. Either the number of burners would need to be increased (numbering-up), which would significantly increase the space needed for operation, or an augmentation of the overall burner geometry would be needed which would most likely increase manufacturing costs due to an increase in component height. Our setup as presented in Fig. 4, however, will enable scale-up far more easily than by numbering-up. By simply mounting more flat burners into the chimney-like retainer, the increase in the overall installation space will be much reduced and the burner printing height will remain constant.

As shown above, our presented geometric solution of a metal flow distributor on a lab scale is similar to other use-cases and demonstrates the potential of AM in a different field, using the possibility of functional design, where geometry rigorously follows process requirements.

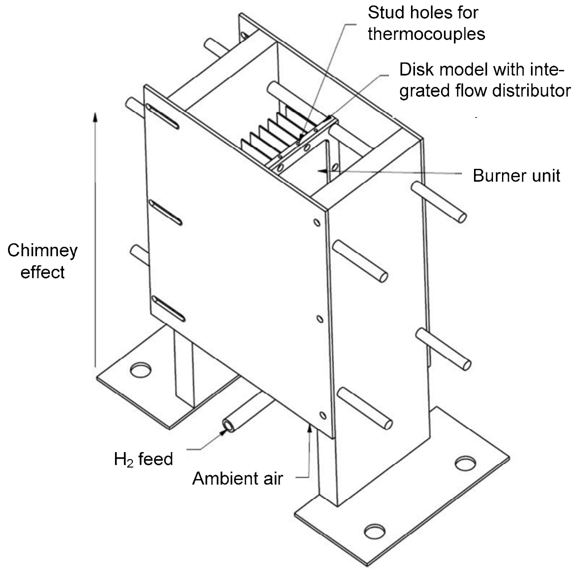

Fig. 4 Sketch of the burner unit including flow distributor and containment

3 Detailed design

This section describes the detailed design of the flow distributor and complete burner unit as well as the setup of the experimental plant.

3.1 Flow distributor

The flow distributor consists of a plate with an integrated pipe structure (Fig. 2). The design is symmetric since the upper half is a mirror image of the lower half. To reduce producing costs, only one half plate of Fig. 2 was fabricated, with hydrogen feed in the center of the half plate (supplementary Fig. S1). To retain the required chimney effect, the half-plate was later mounted vertically with the long side in the air flow direction. The estimated diameters in Table 1 are not affected by this bisection.

The detailed dimensions chosen in this work can be seen in Fig. 5. Manufacturing the chosen design by conventional methods would be very complex as will be described in Section 3.2. However, AM technologies offer significant advantages for producing metal parts with extreme complexity.

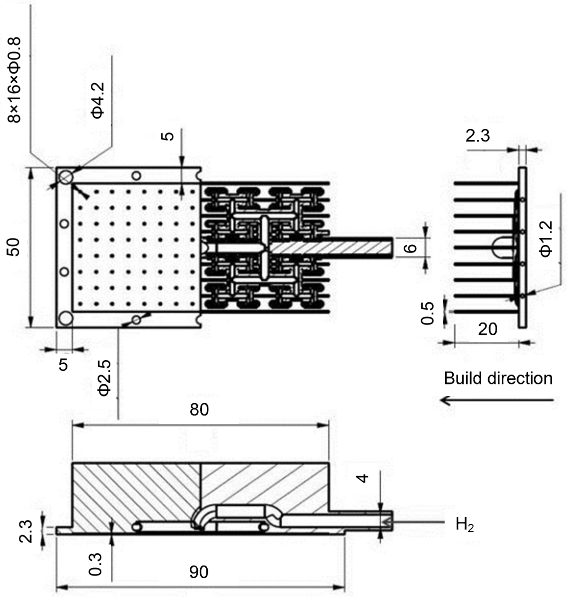

Fig. 5 Detailed drawing of the flow distributor (the build direction is marked by the arrow) (unit: mm)

The design of Fig. 5 was produced in stainless steel 316L (1.4404) using selective laser melting (SLM) technology at inspire-icams (St. Gallen, Switzerland). The raw material was selected because austenitic steel is known to be resistant against hydrogen embrittlement. Further requirements like stiffness, elastic limit, or temperature stability were not taken into account due to the practically unpressurized operation and modest working temperature (≤400 °C). The powder material used shows a particle size distribution, with a volume-based D_{10}=13 μm, D_{50}=22 μm, and D_{90}=41 μm (supplementary Fig. S2).

A Concept Laser M2 machine was used with the processing parameters (supplementary Table S1). These parameters were developed prior to this project, reaching a material density for such uncritical structures of about 99.5%. Such a processing window enables a build process to be designed that should not lead to significant defects and which enables the proper manufacturing of internal channels with small diameters as shown in Fig. 5, thereby avoiding the formation of too many adhering particles and melt-droplets in the top area of such channels.

To guarantee the optimal part quality with respect to the surface quality of the blades and to reduce the build time, the part was built in a horizontal orientation (supplementary Fig. S3). The cavity on the down-side was filled with a lattice-support structure, including the junction, and an additional base of 0.5 mm was used enabling the removal of the part from the base-plate by electrical discharge machining (EDM). The remaining support structures were removed by conventional cutting processes after the SLM-process and EDM cutting of the flow distributor from the build base-plate. This approach of build orientation and finishing enables the production of a good, flat surface with well-defined channel-openings for H_2.

The top vaults of the channels are crucial points for SLM, as they are overhanging surfaces. Although the quality of these surfaces will be worse than that of the vertical side wall (with a typical surface roughness R_a≈5 μm to 6 μm (Spierings et al., 2011), the top quality was still acceptable (supplementary Fig. S4).

In cases where the flow resistance of the channel plays a crucial role, suitable post processing could be achieved by extrude honing processes (Schrader and Elshennawy, 2000), which are also able to deal with continuously changing cross-sections. An alternative measure to increase the channel top surface quality for future applications, if necessary, would be to use a channel cross-section (supplementary Fig. S5). This kind of tangenting triangle would avoid the problem of flat-angles in the area of overhangs, and as the application presented here can be regarded as unpressurized, the emerging notch would not inflict any weak spot on the part.

The build direction shown in Fig. 4 was chosen because construction height is one of the most cost-determining parameters in SLM (Rickenbacher et al., 2013). This build direction is also advantageous with regard to distortion of the lamellae, as in this build orientation no significant distortions will arise.

Due to the good and large area of contact of the flanges with the build plate, the warpage of the whole part could be reduced to a minimum of about 1 mm. However, warpage in SLM cannot be fully avoided because residual stresses arise during the welding-like SLM process. Therefore, the scanning strategy (e.g., orientation of the scanning lines in each layer) will also have a certain effect on warpage. In the current case, the standard scanning strategy involving scanning small 5 mm×5 mm islands with alternating scanning directions between the layers was used to minimize stress accumulation over the whole cross-section of the part. Excess powder material remaining in the created channels could easily be removed from the tree-structure using a vibrating table.

The contact surface of the flow distributor on the printing bed was machined off conventionally in the aftermath. The completed part is shown in Fig. 6.

The SLM part was then tested. Smoke was fed through the inlet and an even distribution over all holes was observed (Fig. 7).

3.2 Burner unit

As a next step, the catalyst has to be applied to the flow distributor. Therefore, a sintered metal filter was sputtered with a Pt/Pd alloy using a Polaron SC7680 sputter coater at the Deutsches Zentrum für Luft- und Raumfahrt (DLR, Stuttgart, Germany). Scanning electron microscope (SEM) pictures of the filter before and after sputtering can be found in supplementary Fig. S6.

Note that by sputtering, only a relatively small amount of the catalytic material is applied. In this specific application for hydrogen combustion, this amount was considered sufficient to ignite the reaction.

The sputtered filter was then mounted on the flow distributor (Fig. 8). The filter evenly slows down the residence time of the gas in the filter, which results in a contact time sufficient for catalytic reaction.

The proposed design places very little demand on the geometric tolerances of the SLM-processed part. Basically, only the contact surfaces of the distributor and filter need to be reasonably flat. This is ensured because the support structure originally located here, was machined off using a CNC-machine. A flexible graphite seal between the frame and the distributor/filter was used to ensure gas tightness. As mentioned earlier, in the presence of hydrogen this would require permanent supervision of the seal to ensure functionality. We chose this design for our prototype to provide more flexibility. However, in future development steps, the distributor/catalytic filter/frame unit could easily be sealed by one singular circumferential, continuous welding seam.

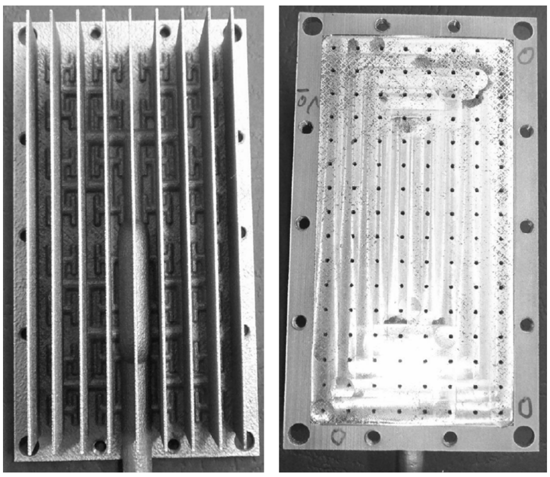

Fig. 6 Photos of the completed flow distributor directly after manufacture: backside (left) and catalyst side (right)

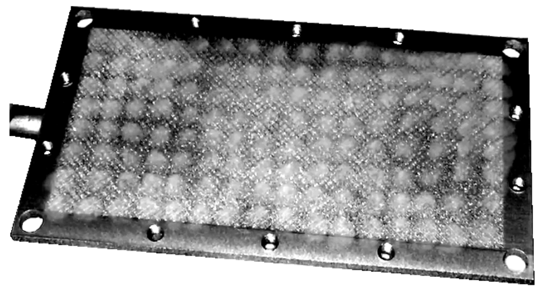

Fig. 7 Photograph of the smoke test (for clarity, the contrast of the picture has been amplified)

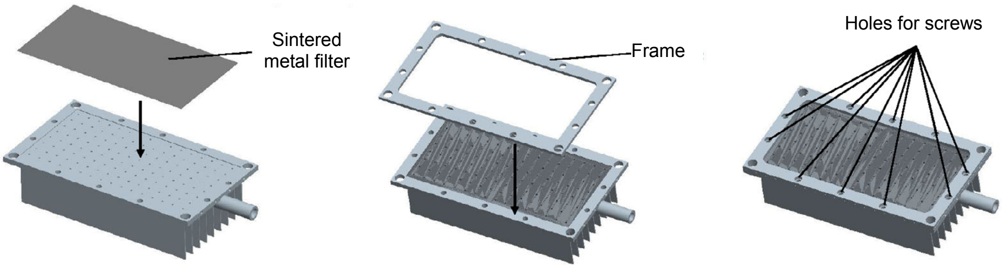

Fig. 8 Assembly of the catalytically sputtered sintered metal filter on the flow distributor

The flow distributor had to be tested for gas tightness on the backside of the catalyst because hydrogen leakage, as mentioned earlier, would present a safety risk. As the operation is aspired to be practically unpressurized, a simple bubble test was considered to be sufficient. Therefore, an un-catalyzed filter was mounted on the distributor, which was then placed inside a container of water, and 0.5 NL/min of air was fed into the equipment. As no bubbles were observed on the backside, the system was considered gas tight.

After that, the complete flow-distributor/catalyst unit was mounted inside a containment. This ensures the above-mentioned chimney effect and a continuous oxygen supply by ambient air. The final setup can be seen in Fig. 4.

3.3 Burner test unit

In a last step, the burner unit has to be placed inside a suitable experimental setup to ensure functionality and safety. The respective process flowchart is shown in Fig. 9.

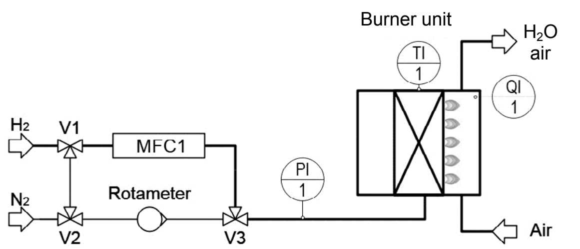

Fig. 9 Process flowchart of the complete experimental setup

Gas dosage of either hydrogen or nitrogen is realized by a mass flow controller (MFC1). N_2 may also be purged into the burner through a rotameter.

In the initial state of operation, nitrogen (N_2) is purged through the burner unit (V2, V1, V3). This ensures that no air (containing O_2) can back mix from burner side into the piping, which would increase the risk of premature ignition. When hydrogen is to be burnt, the respective three-way valves are switched, so N_2 is dosed through MFC1 instead of the rotameter. Then by switching valve V1, N_2 is replaced by H_2 and the burning starts if the catalyst is sufficiently hot.

A pressure gauge (PI1) ahead of the burner unit indicates the overall pressure drop, and the temperature of the flow distributor can be measured using a Type-K thermocouple (TI1). The concentration of unburnt H_2 in the nearby ambient air due to slip is measured using a mobile detection device (QI1).

When the combustion is to be terminated, switching back V1 replaces the hydrogen with nitrogen and the combustion extinguishes.

4 Functional test

After assembling the test unit the burner was tested for functionality. In the first step, 0.5 NL/min H_2 was fed into the system with the burner unit being at room temperature. However, the temperature (TI1) and the rise of H_2 concentration (QI1) indicated that ignition was unsuccessful. Therefore, a short flush with a Bunsen burner was used to start the reaction. The temperature of the catalytic burner then quickly (~20 min) increased up to 120 °C in the center of the catalyst. Steady state was reached at about 200 °C at the center of the catalyst and at an edge temperature of 170 °C. In comparison, the temperature of H_2 combustion without the catalytic sinter metal filter mounted showed values up to 400 °C. Systematic measurements of the whole catalyst area revealed that no hotspots occurred. The H_2 concentration on the catalyst side of the burner remained lower than 30 ppm whereas on the backside it was within the scope of measurement accuracy. This indicates almost complete combustion and can also be regarded as a validation of the bubble test performed as described in Section 3.2, as a hydrogen leakage on the catalyst backside would have resulted in drastically higher H_2 concentrations, especially on the backside.

A further requirement of the experimental plant was the oxidation of hydrogen without the occurrence of flames. As hydrogen flames are colorless, a piece of paper was held against the catalyst surface. It did not ignite. This test was repeated without the sintered metal filter mounted. The setup then worked as a conventional non-catalytic burner, which was confirmed by ignition of the paper.

Finally, the autonomous reignition of the burner after an interruption of hydrogen feed should be mentioned. If under steady state operation with a measured temperature of the flow distributor of

170 °C the hydrogen flow was shut off, the reaction reignited after resetting the hydrogen feed, as long as the catalyst temperature remained above 30 °C.

Obviously, the heat storage due to the thermal mass of the system in combination with the catalyst is a further advantage compared to conventional burners that either do not reignite or constantly need a pilot flame or ignition spark to avoid extinction.

In future versions of the burner, this lower temperature limit might even be extended by augmentation of the metal filter loading with catalytic material. Although requiring a larger hardware and control effort, an electrical heating element which keeps the catalyst temperature at about 40 °C could also be a reasonable extension of the setup. It would guarantee permanent operation if hydrogen was emitted only periodically during a lab experiment.

5 Conclusions

In this contribution, a lab-scale catalytic flameless low temperature hydrogen burner was designed and manufactured using AM. A theory-based branched-tree approach was realized in order to distribute the gas evenly over a catalytic surface. Such fractalized fluid flow systems could also be of interest in other chemical process engineering applications. Constant oxygen supply was ensured by an enclosure generating a chimney effect for ambient air. The concept was successfully tested for functionality.

In the future work, several improvements could be applied to the existing burner unit. First of all, the additive manufacturing of porous structures as catalyst carriers could be integrated into the process (Spierings et al., 2014). This has the advantage that such structures could be produced with defined porosity, a more deterministic channel size, and reliable strength. Depending on the thickness of the porous layer, the flow into the chimney could be distributed more evenly. Subsequent applications of the catalyst could again be applied with sputtering or wet impregnation.

The formation of special holes for the placement of thermocouples in the center of the distributor could also be easily implemented in the manufacturing process.

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List of electronic supplementary materials

Fig. S1 Sketch of the final flow distributor, representing one half of the distributor in Fig. 2
Fig. S2 q_0 particle size distribution of the powder raw material used to produce the flow distributor
Fig. S3 Representation of the flow distributor at a height of 2.5 mm and support-structures needed to support overhanging planes below the part and the flange
Fig. S4 Surface quality of hole cross-sections on a test-piece made by SLM in 1.4404 with diameters from 1 mm to 5 mm
Fig. S5 Schematic sketch of an alternative channel cross-section
Fig. S6 SEM pictures of the sintered metal filter (catalyst support) before and after sputtering with Pt/Pd alloy
Table S1 SLM processing parameters used on a ConceptLaser M2 machine

中文概要

题　目： 空间均匀低温氢气燃烧器开发

目　的： 设计制造低温的催化氢气燃烧器，并对其进行相关功能性测试。

创新点： 成功设计并制造出一个集成流量分配器的催化低温氢气燃烧器。

方　法： 1. 基于树状分叉方法，设计燃烧器的流量分配器，均匀分配气体到催化表面，并利用熔融沉积成型技术制备原型样机。2. 基于测试结果，利用选择性激光熔化技术对燃烧器进行最优化设计。3. 对设计的催化燃烧器的相关功能进行验证。

结　论： 1. 设计的低温催化燃烧器与传统的非催化燃烧器相比具有很多优势，尤其是实现了无焰均匀低温（约 200 °C）的产热；这一技术有望运用于化工领域的增材制造。2. 本文不但用选择性激光熔融技术制备了最终产品，而且利用了熔融沉积成型技术进行快速的样机制备。3. 催化剂多孔载体的调控和催化剂的负载方式研究有望进一步提升燃烧器的综合性能。

关键词： 选择性激光熔化；催化燃烧器；实验室规模；流量分布器；氢气燃烧