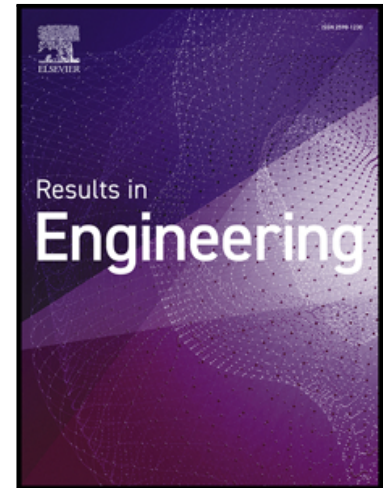


Induction Heating of Washcoated Metallic Structures for the Dry Reforming of Methane

Thomas C. Kieble, Jannik Betz, Damian Mertens, Stefan Heißler, Christoph Klahn, Gregor D. Wehinger

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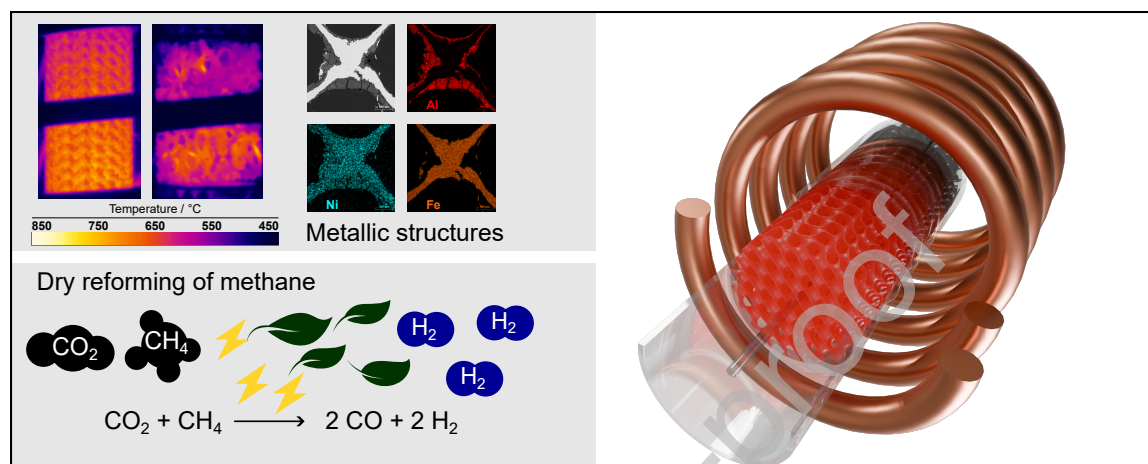
Highlights

- Washcoated metallic structures can simultaneously serve as inductively heated susceptors and catalyst supports.
- Fiber Bragg Gratings enable axially resolved temperature measurements in induction heating.
- FeCrAl foams exhibit optimal heating performance in the ferromagnetic regime.
- Additively manufactured structures show efficient eddy current heating at high temperatures.

Graphical Abstract

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ABSTRACT

Decarbonization strategies targeting CO₂ and CH₄ conversion are essential for reducing the environmental footprint toward climate neutrality. Dry reforming of methane (DRM) is an attractive route for CO₂ valorization and syngas production, offering a substitute for carbon-intensive feedstocks. Endothermic DRM requires significant energy input, making electrification an appealing option for direct and potentially dynamic energy supply. Combining induction heating (IH) with metallic structured packings offers a promising approach for integrating heat generation and catalyst support within a single reactor component.

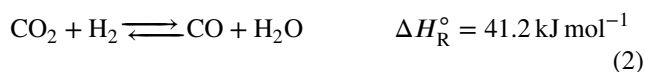
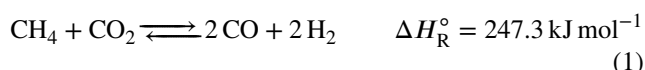
Here, we report two classes of metallic structures: open-cell foams made of FeCrAl and additively manufactured 316L periodic open-cell structures (AMS), employed as susceptors for IH. For each class, several geometric configurations are investigated, all coated with a catalytic alumina washcoat, enabling a large active Ni surface area. FeCrAl foams exhibit optimal heating performance in the ferromagnetic regime <600 °C, which is not favorable under equilibrium-limited DRM. In contrast, AMSs demonstrate efficient eddy current heating at elevated temperatures <800 °C, resulting in a maximum transfer efficiency of 18%. Under optimized operating conditions, conversions close to unity and H₂/CO ratios close to one were achieved. However, careful control of the operating conditions was required, as carbon formation, predominantly in the form of filamentous carbon, was observed. Furthermore, spatially resolved temperature measurements using a Fiber Bragg Grating (FBG) setup proved suitable for IH and confirmed the synergy between AMS and IH. Overall, the results demonstrate the importance of susceptor design for heat generation and temperature distribution in inductively heated catalytic reactors.

1. Introduction

Europe pursues the ambitious goal of becoming the world's first climate-neutral continent by 2050. Achieving this requires substantial efforts across multiple sectors, including energy, mobility, and agriculture. The chemical industry is a major energy consumer and must therefore decouple greenhouse gas emissions, particularly CO₂, from its products. In principle, renewable electricity should be employed for the production of fuels and chemicals [1]. The electrification of chemical and catalytic reactors offers considerable potential to reduce the environmental footprint of a wide range of chemical products worldwide [2]. Among the different concepts of electrified reactors, induction heating (IH) exhibits several advantages, such as a reduced environmental footprint through renewable electricity, direct magnetic field based heating and operational flexibility, which makes it a compelling option for future chemical reactor technologies [3]. To fully exploit the potential of IH, it is advantageous to apply it to highly endothermic reactions such as the dry reforming of methane (DRM, Eq. (1)), in which carbon dioxide (CO₂) and methane (CH₄) are converted into syngas (CO and H₂) at elevated temperatures of 600 – 800 °C [4]. This creates a significant demand for

efficient and scalable reactor concepts that can successfully combine the IH susceptor with an appropriate catalyst and reactor configuration [5].

The overall reaction equation for DRM is shown in Eq. (1), which is accompanied by the Reverse Watergas Shift (RWGS, Eq. (2)) reaction leading to the production of water as byproduct [4]:



Only a few authors showed the potential of induction heating in combination with DRM, while mostly powder catalysts or unstructured packed bed reactor concepts were investigated. Varsano and co-workers used a cobalt/nickel nanoparticle (NP) system synthesized via arc melting in a powder bed for IH DRM [6, 7]. Reaction temperatures larger than 850 °C were achieved and methane conversion and hydrogen production occurred with yields comparable to those obtained by conventional heating. A similar bimetallic Ni/Co NP formulation for IH DRM was synthesized by spray drying presented by Küchen et al. [8]. High conversion and coking resistance was achieved at a specific Ni/Co ratio. However, the stability of the NPs could not be maintained at the high

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reaction temperatures, at which point sintering began and the alloy catalyst was deactivated by coking.

As reported by Mazarío et al., NP based systems are widely used in IH due to the well characterized and understood heating mechanism, mostly due to hysteresis and relaxation losses in the small nm range [9]. In particular, Ni/Co nanoparticle formulations with combined catalyst and susceptor properties have also proven effective in other reactions, such as the steam reforming of methane [10, 11, 12, 13, 14, 15]. However, even when using Co or Co alloys, high reaction temperatures are limited by the corresponding Curie temperature, which is 1121 °C [16] for pure Co. In case of DRM, the NP systems are therefore constrained by the need for precise NP tuning, labor-intensive synthesis, and restricted material freedom, making them both costly and inflexible.

Furthermore, eddy currents can serve as an induction heating mechanism in larger macroscopic millimeter range objects [9]. The formation of eddy currents depends highly on the conductivity, shape, frequency, and size [17]. This combination of predominant eddy current-heated systems for DRM was investigated by only a few authors: Yang et al. utilized FeCrAl monoliths combined with a Ni/MgO washcoat for induction-heated DRM [18]. While their results highlight the promise of macroscopic metallic susceptors, the study solely investigated monoliths with straight channels. Truong-Phuoc et al. showed the combination of DRM and silicon carbide foams combined with a few layer graphene coat to facilitate eddy current heating [19]. Lin et al. demonstrated high-frequency metamaterial reactors with structured conductive foams enabling volumetric induction heating for the RWGS reaction, while spatial temperature measurements highlighted the strong coupling between reactor geometry, IH power delivery, and heat distribution [20].

Dedicated scale-up analyses further indicate that efficiency increases with reactor scale. In addition, radial temperature gradients can be mitigated by tailoring the effective conductive pathways within the structured susceptor [21]. Very recently, Tanksale and co-workers demonstrated the use of eddy current-based complex metallic structures for IH in DRM [22]. Additively manufactured Inconel structures with a NiO/Ce_{0.8}Gd_{0.2}O_{2-δ} washcoat were tested within IH and demonstrated stable activity for 42 h. Triply periodic minimal surface (TPMS) geometries, particularly gyroid structures, exhibited superior heating performance. These studies highlight the high potential of macroscopic eddy current-based systems and reveal a clear dependence of heating behavior on structural design. However, a fundamental correlation between structural parameters and heating efficiency is still lacking. In addition, the internal temperature distribution within the metallic structures remains unclear, as only the surface temperature was measured.

Temperature measurement in IH reactors is an under-developed topic. Thermocouples interact with the magnetic field and therefore cannot be used without further data processing [23]. Therefore, in most of the above mentioned studies, optical temperature measurements were used, such

as thermal imaging (TI) [24] or pyrometers [14], which can only access a surface temperature of the susceptor/structure. Positioning of the coil reduces the optical accessibility and emissivity coefficients of the solid surface are often unknown and/or can also change during the reaction. The before-mentioned direct heating poses new questions in terms of temperature gradients and reactor design, which makes a thorough temperature measurement crucial. This is particularly relevant for complex metallic structures, where eddy currents are highly dependent on the structure's geometry and the heating is not uniform.

Although several studies have demonstrated the feasibility of induction-heated DRM, the fundamental relationship between structured package geometry, eddy current formation, temperature distribution, and catalytic performance remains largely unresolved. Previous studies identified trends between structure type and heating behavior, but no predictive framework or design principles have yet emerged. Consequently, it is still unclear which structural characteristics govern efficient IH and how these characteristics translate into reactor performance. Addressing this knowledge gap is one of the central motivations of the present work.

Structured catalysts and reactor packings with conventional wall heating were developed for different reaction systems by several research groups, among others by Moulijn and Kapteijn [25] and Tronconi and co-workers [26, 27, 28, 29]. In the latter group, various metallic structures with porous catalyst washcoats were studied featuring increased heat transfer properties and high specific surface area. Giani et al. used FeCrAl alloy foams with a porous washcoated alumina layer derived by sol-gel deposited pseudoboehmite for the catalytic CO oxidation [30, 31, 32]. FeCrAl in general is highly suitable as a catalyst support due to the inert alumina layer formed through oxidation on the surface making it temperature resistant and suitable for alumina washcoats at high reaction temperatures up to 800 °C. [33, 34, 35].

Besides open-cell foams, additively manufactured macroscopic metal structures (AMS) are very interesting in process intensification [36] and IH, as they provide flexible design regarding intensified lateral mixing, reduced pressure loss [37], and potentially beneficial eddy current formation. Additively manufactured POCS made of AlSi7Mg0.6 by laser-based powder-bed fusion (PBF) greatly enhanced the radial heat transfer in conventionally heated Fischer-Tropsch synthesis [38]. Furthermore, conventionally heated DRM was carried out in additively manufactured honeycomb monolith reactors with a Ni/CeO₂–ZrO₂ washcoat [39] and electrochemical deposition of Ce and Ni [40], showing high conversion under stable reaction conditions.

The novelty of this work lies in combining spatially resolved temperature measurements via Fiber Bragg Grating (FBG) and TI with catalytic DRM testing to establish structure–heating–performance relationships for two fundamentally different classes of metallic packings. The study aims to identify design principles for induction heated structured reactors that can guide the design of efficient future electrified reactors.

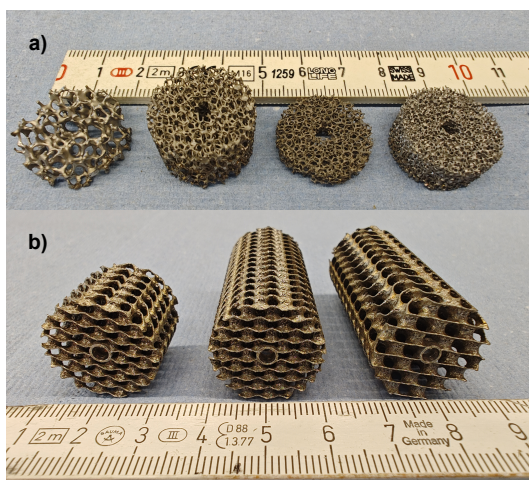


Figure 1: a) FeCrAl foam (10, 20, 30, 50 pores per inch (PPI)) disks with an outer diameter of 21.6 mm and drilled 3 mm holes. b) 316L AMSs with 1.8 mm hole and 3 mm groove with 20 and 50 mm total lengths and 6 and 8 mm cell length.

2. Materials and Methods

2.1. Metallic structures

Two types of metallic structures were used for the IH experiments: open-cell foams and AMS TPMS structures. Open-cell foams were chosen as a highly scalable and cost-effective commercial structure with a long history in catalytic applications [41]. In contrast, the AMS represent a highly tailorable platform that allows systematic adjustment of geometry and electromagnetic properties.

The first structure consists of commercially available (Hollomet GmbH, Dresden) iron chrome alumina (FeCrAl) foams in 10, 20, 30, and 50 pores per inch (PPI) as shown in Figure 1a). The foam plates of different heights (7-16 mm) were first laser cut to produce disks with a diameter of 21.6 mm. To ensure a smooth insertion of the temperature measurement capillary, a 3 mm hole was drilled through the disks. The parameters of the different foam geometries are listed in Table 1.

The second type of structures were TPMS shown in Figure 1b) additively manufactured by PBF. The TPMS geometry consists of a periodic lattice of interconnected diamond unit cells with a uniform wall thickness of 0.3 mm (see Supporting Information, Section S1). Diamond cell type was chosen as it features no direct shoot trough in axial direction to avoid the formation of gas bypassing. At the same time, the washcoating procedure (see subsection 2.2) may benefit from pores that are accessible in the radial direction and extend through the structure (“see-through” pores). Different lengths (20, 50 mm) and cell sizes (6, 8 mm) were investigated and named AMS_{cell length x height}, see Table 2.

The AMS were produced on a Realizer SLM 125 machine from austenitic stainless steel 316L (X2CrNiMo17-12-2) powder. Manual finishing included mechanical support removal, depowdering by aspiration cleaning, and an

Table 1

FeCrAl foams with characteristic design parameters and measured porosity.

pore density / PPI	10	20	30	50
disk height / mm	10	16	7	10
number in stack	3	2	4	3
total stack height / mm	30	32	28	30
porosity (from manufacturer)	0.900	0.900	0.930	0.880
porosity (pycnometry)	0.906	0.904	0.947	0.923

Table 2

AMS 316L structures with characteristic design parameters and measured porosity.

name	AMS _{6x50}	AMS _{8x50}	AMS _{6x20}
cell length l_c / mm	6.0	8.0	6.0
wall thickness / mm	0.3	0.3	0.3
height h / mm	50	50	20
outer diameter / mm	21.6	21.6	21.6
porosity (CAD)	0.882	0.910	0.880
porosity (pycnometry)	0.835	0.877	0.835

ultrasonic bath to ensure free pores and windows. The structures were designed with a 1.8 mm hole in the center and a 3 mm groove to lock on additional parts upstream and downstream. Ceramic static mixers were used in every experiment and acted as a flow homogenizer, a heat shield, and with the printed funnel as a guidance for the penetrating FBG capillary unit (more details see SI section S2). A detailed sketch and a photograph of a typical reactor setup are shown in Figure 2 (a-b). Helium pycnometry (Micromeritics Accu-Pyc 1330) was carried out on all structures to measure the porosity, see Table 1 and Table 2.

2.2. Alumina washcoat and wet impregnation

All structures were washcoated with a commercial Alumina washcoat (Disperal HP 14, Sasol). Prior to the washcoating process, all structures were cleaned two times in ethanol. The FeCrAl foams were also precalcined to ensure a sufficient oxidation layer formation on the surface as recommended by [30]. The structures were fully submerged into the washcoat. Excess washcoat was blown out with air and drying was performed at room temperature with a fan under constant rotation for at least 2 h in line with [42]. For all AMS as well as 10 and 20 PPI foams a 30 wt% Disperal HP 14 solution was used. The 30 and 50 PPI foams were washcoated with a 20 wt% Disperal HP 14 solution, since their pore windows were too narrow to remove blocked pores by air blow-out. Subsequently, all structures were calcined at 900 °C for 6 h with a temperature ramp of 3 K min⁻¹ in a muffle oven. Wet impregnation was carried out by fully submerging the structures in Ni nitrate (Emsure) overnight (0.1 M). Excess liquid was removed by air blow-out followed by drying at room temperature under constant rotation similar to the washcoating process. Some samples were impregnated with a higher Ni loading of 0.5 M to increase visibility in the

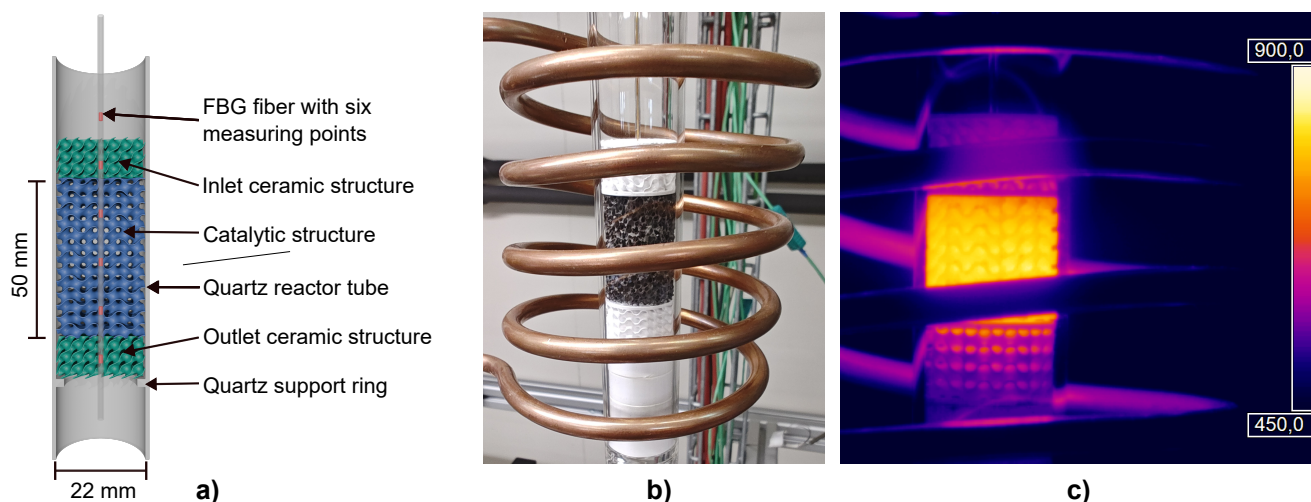


Figure 2: a) Schematic reactor setup with FBG capillary and stacked inlet/outlet and catalytic structures. b) Photograph of typical reactor setup with ceramic inlet/outlet structures, two stacked 20 PPI FeCrAl foams and additional ceramic spacer rings at the bottom to increase packing height and surrounding copper coil for IH. c) Thermal image of an AMS_{6x20} during DRM experiments with temperature scale in °C.

EDX scans, presented in subsection 3.1. Furthermore, the samples were dried at 120 °C for 5 days and then calcined two times at 350 and 450 °C (3 K min⁻¹ heat-up with 200 min hold each) in line with [42]. Loading was evaluated by weighing the samples with the alumina washcoat solely and with washcoat and Ni (oxide).

Characterization of the alumina washcoat was carried out by nitrogen adsorption measurements (Micromeritics ASAP 2020). For this purpose, 20 PPI FeCrAl washcoated foams were calcined at various temperatures and durations, and the washcoated samples were examined in a modified adapter with an inner diameter of 22 mm. The specific surface area (SSA) was determined by the BET method, and the pore radius distribution was obtained using the BJH method. Further details are provided in the SI section S3. To verify a uniform distribution of the alumina washcoat and successful impregnation, a series of SEM/ESEM and SEM/ESEM-EDX analyses were carried out on a 20 PPI FeCrAl foam and a 316L AMS_{6x20} on a Zeiss LEO 1530 Gemini (SEM) and FEI Quanta FEG 650 (ESEM). The samples were partially embedded in epoxy and ground flat to enable a precise determination of the washcoat thickness.

2.3. Reactor setup with induction heating unit and FBG temperature measurements

For all investigations a technical Lab Plant was used running with Beckhoff TwinCat and NI LabView. Gases, i.e., Ar, CH₄, CO₂, CO, H₂, N₂ each 20-1000 Nml min⁻¹, were dosed with a MFC unit (Bronkhorst). Reaction species were analyzed with a Hiden HPR-20 mass spectrometer (Faraday detector) with a heated inlet. Pipes and valves after the reactor were heated to 120 °C to prevent the condensation of water originating from the RWGS reaction.

The induction heating unit consists of an Ambrell Easy-Heat 2.4 kW power generator (power head capacity 0.66 μF,

$I = 0 - 400$ A, $f = 150 - 400$ kHz) with appropriate water cooling (Lauda Variocool VC 3000 set to 20 °C) of the coil. To ensure a homogeneous magnetic field, an inhouse produced copper coil with five windings, a diameter of 78 mm, and a height of 93 mm was employed. The setup further included a brass adapter with 6 mm Swagelok fittings and a soft copper pipe (6 mm outer diameter). Further information is available in the SI S4. The coil frequency was measured by the Easy Heat power Supply Unit (PSU), whereas the input power of the whole PSU was measured by a Beckhoff KI3403 power measurement bus terminal. The reactor consists of a cylindrical quartz glass tube (Schott Ilmasil PN) of 800 mm length, 22 mm inner diameter, and a wall thickness of 2 mm. A quartz support ring of 2 mm was fused into the tube to support the structure stack, see Figure 2a).

The FBG temperature measurement was implemented by a commercial FBG system (Fisens FiSpec FBG X100). The FBG fiber consists of a grating 0.150 mm quartz fiber encapsulated in a 0.440 mm quartz tubing. A quartz tube of 500 mm length with an outer diameter of 1.5 mm was used to protect the fragile fiber. This capillary was inserted into the reactor stacking, see Figure 2b), and sealed via Swagelok Ultra Torr 1/16 inch fittings. It was therefore possible to easily relocate the capillary package to fit the desired axial position. In this vertical arrangement, additional mechanical load on the capillary is minimized, ensuring that the temperature measurements remain unaffected by mechanical stress. Ceramic spacer rings (aluminiumoxide) were used in several cases to match the stack height according to the FBG measuring zone (also displayed in Figure 2b). The fiber includes six gratings distributed equidistantly on 60 mm length. To ensure accurate measuring, the temperature range was recalibrated to 900 °C in a muffle oven and a quadratic fit was applied to match the shift in wavelength to

the according temperature. The FBG calibration exhibited a standard deviation of 8.3 K, while initial temperature readings before and after the experiments differed by less than 5 K, indicating good reproducibility of the sensor response. Further information can be found in the SI section S5.

For supplementary validation, thermal imaging was carried out to identify possible hot spots and to assess the qualitative heating behavior. An Optris PI 1M (27° x 17°) with PIX Connect ($\epsilon = 1$) software was used, which can measure the temperature (450–1800 °C) of the metallic structures through the quartz glass, as illustrated exemplarily in Figure 2c). To determine $T_{TI,max}$, the maximum temperature within the structure was localized using PIX Connect.

2.4. DRM experiments

Prior to all reactive tests, a careful positioning was carried out to ensure that the heated metallic sample is located in the center of the induction coil and also accessible for the TI camera. Next, the sample was flushed thoroughly with argon to ensure any remaining air was removed. All samples were subsequently reduced in the reactor to convert nickel oxides formed during calcination into active metallic Ni. Therefore, the sample was heated to 450–500 °C and reduced with a total flow of 400 Nml min⁻¹ at 10% H₂ in Ar for 45–60 min. Next, the reactive tests were carried out at different heating powers and flow rates under stoichiometric conditions of CH₄ and CO₂, diluted in 50% argon. In the following, only the flow rate of CH₄, $\dot{V}_{CH_4}$, is reported. The experimental procedure followed the three steps:

1. Vary the coil current I at a constant low flow rate of $\dot{V}_{CH_4} = 100$ Nml min⁻¹ up to a methane conversion close to 90%.
2. Vary the flow rate from 100 – 400 Nml min⁻¹ at two constant coil currents I to check the reactor performance under high gas hourly space velocities.
3. Check for stability of the catalyst and carbon deposition at $\dot{V}_{CH_4} = 200$ Nml min⁻¹ and $T_{FBG,max} \approx 650$ °C for 3–4 h time on stream.

After reaction, the samples were removed from the reactor and deposited carbon was analyzed via Raman and SEM, see details in subsection 2.5

Methane conversion, X_{CH_4} , was calculated regarding the inlet molar flow rate $\dot{N}_{CH_4,in}$ dosed by the mass flow controller and the outlet molar flow rate $\dot{N}_{total,out}$ together with the molar fraction y_{CH_4} measured by the mass spectrometer.

$$X_{CH_4} = \frac{\dot{N}_{CH_4,in} - y_{CH_4} \cdot \dot{N}_{total,out}}{\dot{N}_{CH_4,in}} \quad (3)$$

Furthermore H₂/CO-ratio was calculated based on the outlet ratio of the measured molar flows $\frac{y_{H_2}}{y_{CO}}$. When not noted otherwise, the maximum temperature of the six FBG measuring points $T_{FBG,max}$ was used as a comparing temperature. The equilibrium conversion, $X_{CH_4,eq}$, was calculated based on a cantera routine (GRI-Mech 3.0 [43], ideal

gases, NASA-7 polynomials, minimizing of the Gibbs Free Energy). Analogously, the transferred energy into the reactor $P_{reaction}$ was calculated based on the gas enthalpy flows. Specific enthalpies of all components h_i were calculated via the cantera routine. For $h_{i,in}$ room temperature was assumed and for $h_{i,out}$ the maximum recorded temperature $T_{FBG,max}$ was used, as it was deemed the best approximation of the outlet temperature of the metallic structure.

Reactor pressure was considered in the calculations although normally close to atmospheric pressures. All gases were treated as ideal.

$$P_{reaction} = \sum_i \dot{N}_{i,in} \cdot h_{i,in} - \sum_i \dot{N}_{i,out} \cdot h_{i,out} \quad (4)$$

With the measured input power of the Easy Heat PSU in the induction heating system, P_{PSU} , the transfer efficiency, η_{trans} , was determined according to [44]:

$$\eta_{trans} = \frac{P_{reaction}}{P_{PSU}} \quad (5)$$

2.5. Carbon characterization

Carbon characterization was performed via Raman spectroscopy and additional SEM images. Raman spectra were measured by Micro Raman spectroscopy with a Bruker Senterra 2 Raman spectrometer (Bruker Optics, Ettlingen, Germany). As excitation source a green diode pumped solid state NdYAG laser (wavelength: 532 nm) was used. The excitation beam as well as the Raman backscattering radiation was guided through a 20x objective, NA 0.45 (OLYMPUS, Tokyo, Japan), to the spectrograph. The spectra were obtained with a resolution of 4 cm⁻¹ in the range of 100 cm⁻¹ to 4100 cm⁻¹ at 250 μW laser power. The integration time was 80 s with 4 co-additions (4 times 20 s). The background signal was collected every time prior the sample measurement was started. The spectra were evaluated using the Bruker OPUS software. FeCrAl foam samples were analyzed directly in the Raman microscope. The outlet foam part in the stack was used. Carbon deposits collected from the AMS were mechanically scraped from the reactor and analyzed separately. To account for potential heterogeneity of the deposited carbon, four to five measurement spots distributed across the structure were selected, and the corresponding spectra were evaluated statistically.

For carbon characterization, the D, G, and 2D Raman bands were evaluated (see Table 3). Baseline correction was performed using an asymmetric least-squares algorithm followed by Savitzky–Golay smoothing. The degree of graphitization and structural disorder was assessed using the ratios $\frac{I_D}{I_D+I_G}$, $\frac{I_D}{I_G}$, and $\frac{A_D}{A_G}$, where (I) and (A) denote peak intensity and integrated peak area, respectively. Herein, only the mean values averaged over all measurement points are reported in Table 4

3. Results and Discussion

3.1. Washcoat and catalyst characterization

The typical DRM operation temperatures are in the range of 600–800 °C, which requires a high thermal stability

Table 3
Raman bands used for carbon characterization.

Band	Wavenumber / cm^{-1}	Assignment
D	1250–1450	Defects in graphitic carbon
G	1535–1610	Ordered sp^2 graphitic carbon
2D	2500–2800	Graphitic stacking

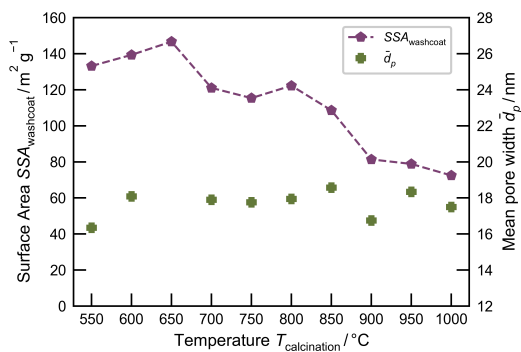


Figure 3: Specific Surface Area (SSA) of the washcoat and mean pore diameter ($\bar{d}_p$) of washcoated FeCrAl 20 PPI foam at different calcination temperatures $T_{\text{calcination}}$. Calcination under air atmosphere (linear heating ramp of 3 K/min followed by 12 h hold).

of the washcoated catalyst. Therefore, the specific surface area (SSA) and the mean pore diameter $\bar{d}_p$ of the 20 PPI FeCrAl foam (acting as a representative of all structures) were analyzed at different calcination temperatures to ensure washcoat stability, see Figure 3.

The SSA decreases with increasing temperature from approx. 140 to $80 \text{ m}^2 \text{g}^{-1}$ at 900°C , which is caused by sintering and phase transition processes. The formation of metastable $\gamma\text{-Al}_2\text{O}_3$ from aluminiumhydroxide (pseudoboehmite) takes place at around 500°C [45, 46]. At higher temperatures, formation to other metastable phases (δ, θ) can occur, since the transformation process is topotactically [46]. $\alpha\text{-Al}_2\text{O}_3$ is the only stable phase that requires complete recrystallization and is therefore kinetically hindered by the relevant presence of process temperatures above 1000°C [47, 48]. From a technical point of view, the Alumina washcoat produced in this work is suitable as a catalyst carrier in the high temperature DRM providing a stable SSA of $80 \text{ m}^2 \text{g}^{-1}$. Therefore, the calcination temperature of all structures was set to 900°C to ensure it exceeds the highest reactor temperature during DRM experiments. Time dependent SSA revealed an ideal calcination time of 6 h. Additionally, the constant pore diameter of around 18 nm implies that the mesoporous pore network is stable, with little to no species mass transport limitations due to collapsing pores. The mean pore diameter was used to calculate the pore effectiveness factor, η , which is nearly unity up to a washcoat thickness of $100 \mu\text{m}$, see SI section S6 for more information. In comparison with the supplier's data [49], $160 \text{ m}^2 \text{g}^{-1}$ could be reproduced although the

suggested 3 h of calcination at 550°C was not sufficient to achieve a stable conversion.

With SEM and ESEM-EDX, a qualitative distribution of the washcoat on a 20 PPI FeCrAl foam is shown in Figure 4. It is clearly visible that the FeCrAl foam struts are hollow, which originates from the manufacturing route via coated polyurethane particles. As evident from the EDX-Al scan, a strong Al signal is observed both at the outer surface and within the hollow interior of the strut, indicating that a substantial fraction of the washcoat is deposited inside the strut cavity. Furthermore, the structure inventory provided in the Supporting Information (S8) shows that the catalyst loading, calculated from the mass of Ni oxide relative to the washcoat mass, is substantially higher for the FeCrAl foams than for the AMS. While the catalyst loading of the AMS ranges from approximately 1–1.7 wt%, significantly higher values are observed for the FeCrAl foams, increasing from about 2 wt% for the 10 PPI foam to approximately 5 wt% for the 30 and 50 PPI foams. This increased catalyst loading suggests that a considerable amount of Ni is deposited within the internal cavities of the foam struts during the fully submerged wet-impregnation process.

The internal cavities are less likely to contribute to the overall reaction rate due to mass transfer limitations. A further study should characterize the inner deposited part either via detailed modeling or highly resolved 3D X-ray Tomography measurements as reported in Zanota et al. [50] to estimate the inaccessible cavity part. Also removing the inner cavity part via vacuum infiltration could be applied, similar to works of Vogt et al. [51].

An additional connection between the two struts is observed, see the EDX-Al image in Figure 4, due to partial pore blockage. Given the complex foam morphology and the variation in pore window sizes, some blockage was unavoidable despite careful air blow-out. However, it is considered acceptable, as most pore windows remain unobstructed. More images are presented in the S7 in the SI.

Similar trends are seen for the 316L AMS displayed in Figure 5, where a thin washcoat layer covers the structure. The washcoat thickness on the strut walls ranges from 10 to $50 \mu\text{m}$, while at the junctions of the diamond cells it increases up to $300 \mu\text{m}$. EDX-Al and EDX-Ni scans reveal that the layer is entirely made up of alumina that has been fully dispersed with Ni, which implies that the washcoat procedure was successful, resulting in a high catalyst surface area. The EDX Ni signal in the 316L structures is also prominent, as 316L contains 12 wt% Ni, which is comparable to the washcoat loading, that was specifically adjusted to enhance EDX signal intensity. Several cracks can also be seen, which originate from shrinking during drying and calcination, which may be beneficial for the species transport within the washcoat.

Overall, the samples were all washcoated with the same procedure. The only exceptions were the 30 and 50 PPI foams, for which a washcoat with lower solid concentration was used, as described in subsection 2.2. Several authors have proposed alternative washcoating procedures, such as

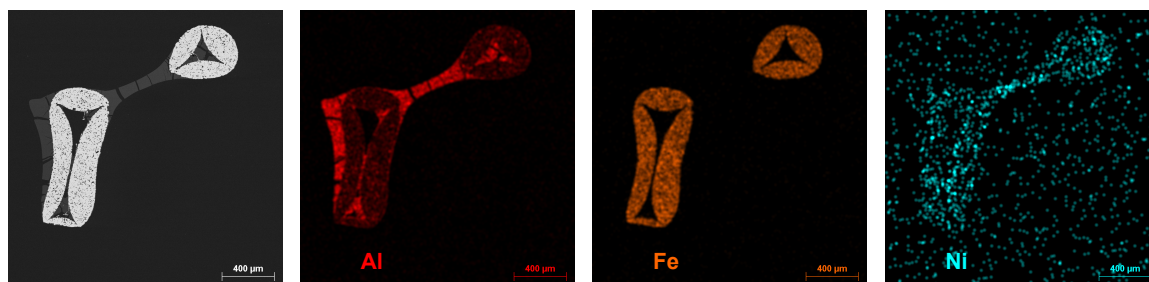


Figure 4: ESEM image (Backscattered Electron Detector) of a 20 PPI FeCrAl foam embedded in epoxy, showing a visible internal hollow region in the foam struts. EDX-Al mapping showing the washcoat dispersed on the outer surface and internal hollow region. EDX-Ni signal is low due to the 0.1 M impregnation. Layer thickness of the washcoat is between 10-30 μm . Connection between two struts is observed due to partial pore blockage, as seen in the EDX-Al.

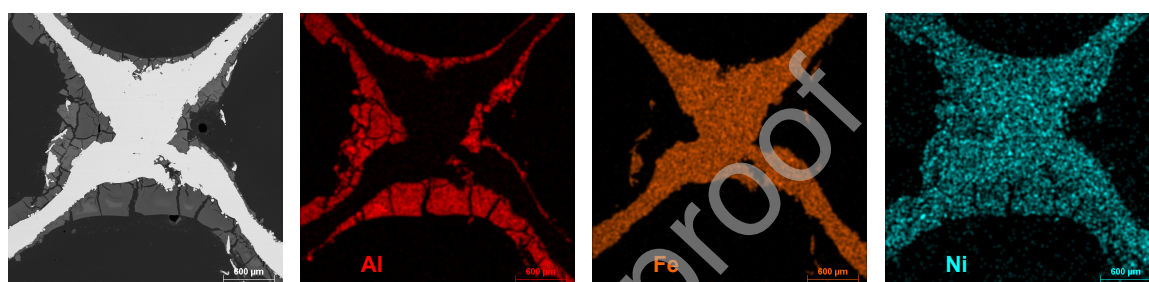


Figure 5: ESEM image (Backscattered Electron Detector) of a AMS_{6x20} part (impregnated with 0.5 M Ni Nitrate for increased EDX-signal, not used during the reactive tests). EDX-Al and EDX-Ni mapping revealing the washcoat layer of Al with Ni dispersed throughout the washcoat. EDX-Ni signal in the 316L part is equally high as the surrounding washcoat due to the 12 wt% Ni content. Layer thickness of the washcoat between 10-50 μm at the walls with locally higher washcoat thickness at the junction of the diamond cells.

spin-coating developed by Ambrosetti et al., which results in a more homogeneous washcoat layer even on high-PPI foams [52]. More recently, μCT measurements were included early in the washcoating procedure of a honeycomb catalyst to control layer thickness by adjusting recipe and process parameters [53].

3.2. Temperature measurements during IH experiments

As reported earlier, temperature measurements are crucial in IH to evaluate the induced power of the susceptor and to assess temperature gradients within the reactor, particularly for the macroscopic structures investigated in this work. Several impact factors have to be considered when discussing temperature in IH:

- IH governed by eddy currents and hysteresis heating. Both influenced by the material, temperature and in case of the eddy currents also by the geometric structure.
- The endothermic DRM and RWGS reactions, which strongly depend on the availability of active catalyst sites, temperature, local concentrations, and thermodynamic equilibrium

- Heat transport within the structure, taking into account both interconnected structures (AMS) and loosely stacked FeCrAl foam configurations.
- Heat losses to the environment in the uninsulated system via convection, conduction, and radiation, which are difficult to quantify due to unknown emissivity and unconstrained conductive pathways.

Most of the above mentioned factors are coupled leading to an highly complex interplay.

Using the FBG measurement technique, the core temperature profile of the metallic structures is obtained, largely unaffected by heat losses to the environment.

Figure 6a) displays the axial temperature for all AMS and the 20 and 50 PPI foam structures at the highest investigated flow rate of $\dot{V}_{\text{CH}_4} = 400 \text{ Nml min}^{-1}$ and a constant coil current of 140 and 160 A, respectively, under reactive conditions. The temperature profile was shifted such that the beginning of the metallic structure corresponds to an axial position of 0 mm.

Temperatures within the metallic structures vary significantly, with the long AMS configurations (AMS_{6x50} and AMS_{8x50}) exhibiting the highest overall structure temperatures. In contrast, AMS_{6x20}, FeCrAl_{20PPIx32}, and FeCrAl_{50PPIx30} show comparable temperature levels, which are notably

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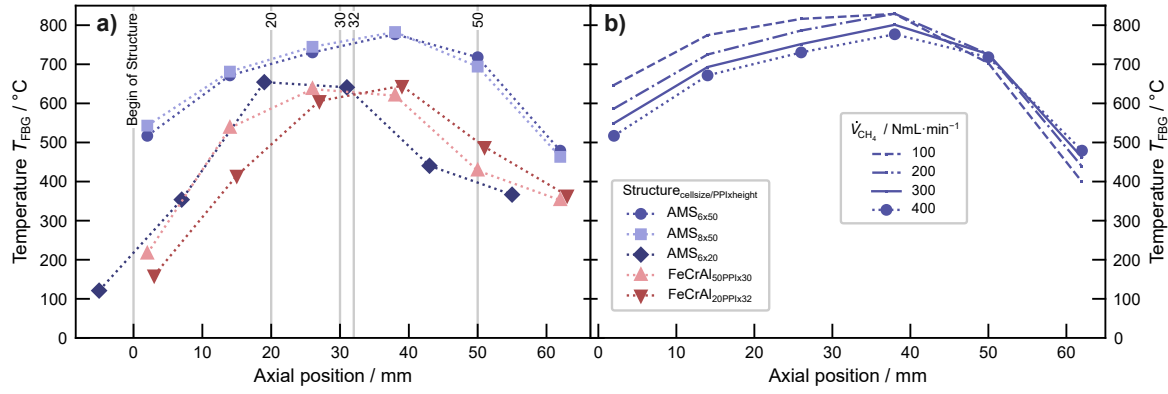


Figure 6: a) Axial temperature profiles of selected structures at $\dot{V}_{CH_4} = 400 \text{ Nml min}^{-1}$. AMS_{6x50}, AMS_{8x50} and AMS_{6x20} at 140 A. FeCrAl_{50PPIx30} and FeCrAl_{20PPIx32} at 160 A. b) Axial temperature profiles of AMS_{6x50} at $\dot{V}_{CH_4} = 100 - 400 \text{ Nml min}^{-1}$ at 140 A. All temperature profiles have been shifted such that the beginning of the metallic structure corresponds to an axial position of 0 mm. The structure length is also indicated, with the corresponding positions marked by vertical grey lines.

lower than those observed for the long AMS configurations. This indicates that the structure and the stack composition affects the heating behavior, which will be evaluated further in subsection 3.3.

When comparing the short AMS_{6x20} with tall AMS_{6x50}, it is clearly evident that the initial temperature gradient at axial position of 0 mm is significantly lower for AMS_{6x20}. Also AMS_{6x50} displays a significant higher initial temperature of approx. 300 K at the axial position of 0 mm. This indicates that there is significant axial heat transfer in the metallic structure through the continuous TPMS structure.

All structures exhibit their highest local temperatures at the end of the metallic section. The only exception is FeCrAl_{50PPIx30}, which shows its maximum temperature slightly downstream of the metallic structure, within the ceramic outlet region. This may be due to the 12 mm spacing of the FBG fiber, which is too coarse to resolve the local temperature peaks at the end region.

Figure 6b) displays the temperature distribution for AMS_{6x50} at different flow rates, i.e., $\dot{V}_{CH_4} = 100 - 400 \text{ Nml min}^{-1}$ at a constant coil current of 140 A. As seen in Figure 6b), for all flow rates an increase in temperature can be noted until $T_{FBG4,38\text{mm}}$, where the maximum temperature is recorded. Subsequently, a slight decrease is observed up to $T_{FBG5,50\text{mm}}$, followed by a pronounced drop of 200–300 K at $T_{FBG6,62\text{mm}}$. For the low flow rate of $\dot{V}_{CH_4} = 100 \text{ Nml min}^{-1}$, measured temperatures in the structure are all larger than 640 °C with a maximum temperature $T_{FBG4,38\text{mm}} = 829 \text{ °C}$ resulting in the later mentioned conversion of 92%. A decrease in temperature is generally noted as flow rates increase. Temperatures at $T_{FBG1,2\text{mm}}$ drop from 647 to 517 °C and at $T_{FBG4,38\text{mm}}$ from 829 to 777 °C respectively.

As the inlet gases enter the reactor at room temperature, significant heating of the gas phase is required before the equilibrium-governed dry reforming of methane (DRM) can proceed. High localized heating, as well as axial heat transfer within the structure, is therefore essential to establish an extended zone of elevated temperature with high reaction

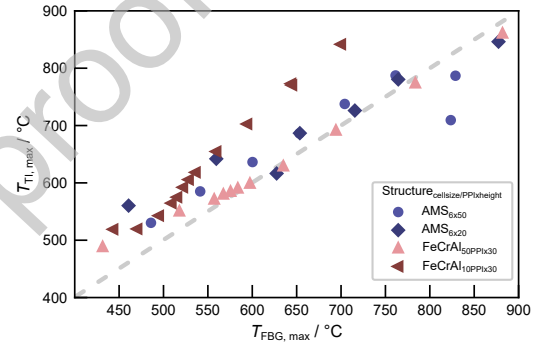


Figure 7: Temperature $T_{TI,max}$ over $T_{FBG,max}$ for selected structures (AMS_{6x50}, AMS_{6x20}, FeCrAl_{50PPIx30}, and FeCrAl_{10PPIx30}) at $\dot{V}_{CH_4} = 100 \text{ Nml min}^{-1}$. Identity line plotted for clarity.

rates for the equilibrium limited DRM. This is evident for the long AMS configurations (AMS_{6x50} and AMS_{8x50}), where a stable conversion of approx. 80% is observed at the highest flow rate. This can be attributed to both localized intense heating and efficient axial heat transport within the structure, discussed in detail in subsection 3.6.

While FBG measurements provide access to the internal core temperature of the metallic structures, thermal imaging (TI) captures the surface temperature without affecting the structure and is thus the established standard measurement technique in induction heating.

A full comparison of the two techniques is not completely valid due to significant heat losses as stated, the formation of temperature gradients within the structure, and the use of different probing locations. Therefore, the maximum recorded temperatures, $T_{TI,max}$ and $T_{FBG,max}$, are compared instead.

Figure 7 compares the maximum temperatures measured with TI and FBG for a constant flow rate of $\dot{V}_{CH_4} = 100 \text{ Nml min}^{-1}$. AMS_{6x20}, AMS_{6x50}, FeCrAl_{10PPIx30}, and FeCrAl_{50PPIx30} are included at varying coil currents. It can be noted that the temperatures generally follow the identity

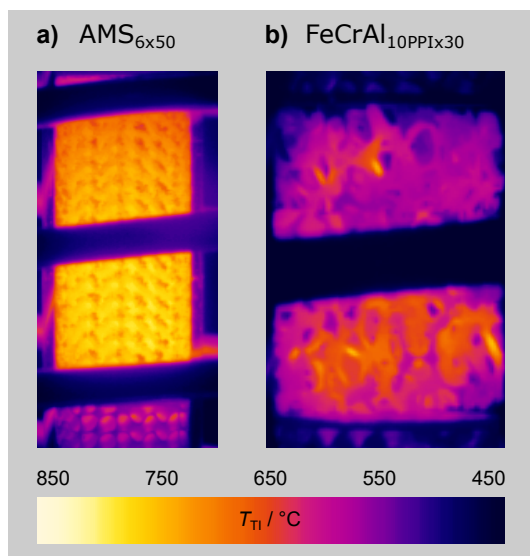


Figure 8: Thermal images: a) AMS_{6x50} at 140 A and $\dot{V}_{\text{CH}_4} = 100$, Nml min⁻¹ with the diamond cell structure visible as cooler regions in contact with the outer quartz wall, b): FeCrAl_{10PPIx30} at 180 A and $\dot{V}_{\text{CH}_4} = 400$ Nml min⁻¹. Localized temperature hot spots within the struts appear as bright regions.

line, although several deviations are apparent: The 10 PPI foams generally show the largest deviations, with approx. 150 K, which may result from the coarse foam geometry, which does not allow sufficient heat transfer to the FBG capillary. In contrast, the finer 50 PPI foams exhibit smaller temperature deviations, which can be attributed to their finer pore structure.

AMSs tend to exhibit smaller deviations, with a maximum of approximately 50 K, except for AMS_{6x50} at the highest temperature of $T_{\text{FBG,max}} = 820$ °C. This may be attributed to localized heating patterns formed by eddy current heating. In addition, carbon formation, which was observed during the measurements, may explain the significantly lower $T_{\text{TI,max}}$ compared to $T_{\text{FBG,max}}$, as carbon deposits affect optical camera access.

While evaluating qualitative heating via the TI in Figure 8, several other aspects could be noticed:

For the AMSs, locally varying surface temperatures were observed, which are attributed to spatially dependent eddy current heating and individual metal walls in contact with the outer quartz wall acting as heat-exchange elements, leading to cooler outer regions (see also Figure 2c)). The AM diamond cell structure can thus be readily recognized from the cooler shaded border regions.

Furthermore, the 10 PPI foams show local hot spots in the foam struts, which are unevenly distributed throughout the structure. This may be caused by current channeling in thinner struts, resulting in locally enhanced heat generation, and is also consistent with the large deviation of $T_{\text{TI,max}}$ compared to $T_{\text{FBG,max}}$ shown in Figure 7.

Based on this comparison, several conclusions can be drawn regarding temperature measurements in IH. Fiber

Bragg Grating (FBG) measurements in the uninsulated setup are highly suitable, as they are only weakly affected by heat losses to the environment. However, a modified configuration ensuring sufficient thermal coupling to the structure remains necessary. This limitation becomes particularly evident for coarse structures, such as the 10 PPI foams, where effective heat transfer to the sensing location is restricted.

Optical temperature measurements can be used to compare local temperature distributions within the structure, which is particularly important in this work due to the complex macroscopic metallic geometries. In addition, they enable the resolution of pronounced local hot spots, i.e. within the struts of the 10 PPI FeCrAl foam. Nevertheless, optical temperature measurements are limited by complex radiation transmission through the reactor wall, uncertainties in emissivity coefficients, and dynamically changing surface conditions arising from carbon deposition as well as oxidation and reduction processes in the catalyst and structured packing. In addition, optical access inherently complicates the implementation of insulated measurement setups and optimized coil geometries with narrow spacing. To ensure adequate comparability between the different structures, the subsequent evaluation in this work is therefore based on the maximum recorded FBG temperature, $T_{\text{FBG,max}}$.

3.3. Reactive tests with FeCrAl foams

The FeCrAl foams were used for reactive tests at low flow rates of $\dot{V}_{\text{CH}_4} = 100$ Nml min⁻¹ with varying coil currents to assess how heating and reaction performance depends on morphology. A sufficient number of foam disks were stacked to approx. 30 mm, see Table 1. Over the reactor length, a temperature profile develops due to the complex interplay between the magnetic field, convective and diffusive heat transport, as well as the local heat of reaction. Temperature profiles measured with the FBG system are presented and discussed above, see section subsection 3.2. The maximum recorded temperature of the stack, $T_{\text{FBG,max}}$, is used for the following evaluation. Figure 9 shows methane conversion X_{CH_4} and the maximum temperature $T_{\text{FBG,max}}$ over coil current, I , and therefore varying magnetic field strengths. Also methane conversion X_{CH_4} in reference to $T_{\text{FBG,max}}$ is shown with the corresponding equilibrium conversion.

In Figure 9a), the temperature increases non-linearly for all investigated foam sizes with increasing coil current. All curves are characterized by a steep temperature increase at low coil currents, followed by a temperature plateau, and finally a linear increase for high coil currents. For the 20 and 50 PPI foams, temperatures of 500 °C at 50 A and maximum temperature of 800-900 °C at 180 A were observed. Both foam sizes display a temperature plateau between 60 and 100 A followed by a nearly linear heating behavior, whereas the 50 PPI foam shows the highest temperatures for constant coil currents. In contrast, the 10 and 30 PPI foams exhibit lower temperatures over the entire coil current range. The temperature plateau of the 10 PPI is approx. 100 K lower than for the 20 and 50 PPI. For the 30 PPI, this plateau is not

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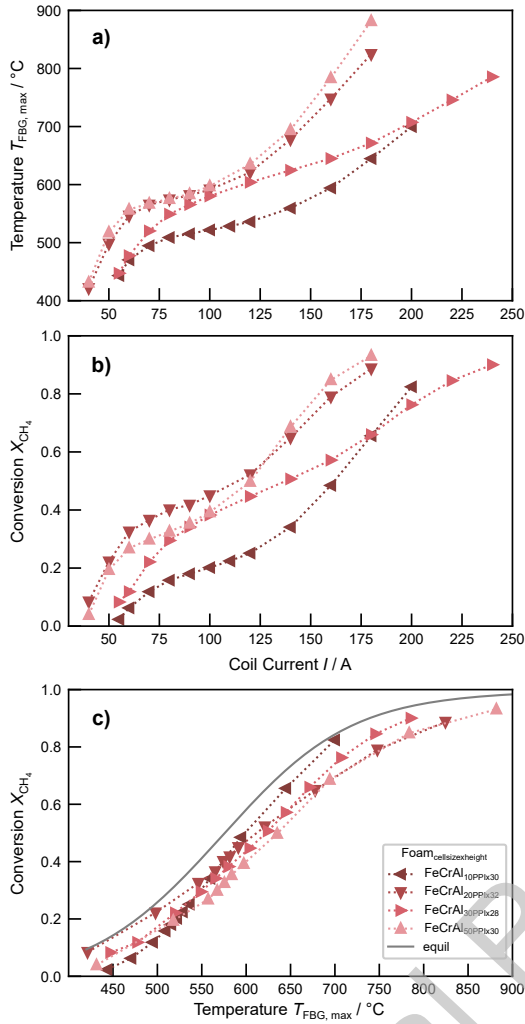


Figure 9: a) Maximum reactor temperature $T_{\text{FBG,max}}$ and b) methane conversion X_{CH_4} over coil current I for different foam structures at $\dot{V}_{\text{CH}_4} = 100 \text{ Nml min}^{-1}$. c) Methane conversion X_{CH_4} as a function of $T_{\text{FBG,max}}$ with equilibrium conversion calculated at $T_{\text{FBG,max}}$ and reactor pressure $p = 1.12 \text{ bar}$.

as prominent. Linear trends for the heating at high currents above 120 A can also be observed with the slope of the 30 PPI being the lowest followed by the 10 PPI and exceeded by the already mentioned 20 and 50 PPI foams.

The methane conversion profiles over the coil current (Figure 9b) behave similarly to the temperature. Conversion for the 20 and 50 PPI foams is generally higher compared to the 10 and 30 PPI foams. However, over the temperature plateau section, the conversion continuously but slowly increases. 80% conversion can be achieved for the 20 and 50 PPI foams at 160 A, noticeably lower than 200 A for 10 PPI and 220 A for 30 PPI foams, respectively. For low coil currents the 10 PPI generally shows the lowest conversions until 200 A, where it starts to overpass the 30 PPI foams. The 30 PPI foam shows comparable conversions to the 50 PPI foam in the range of 80–100 A, but its performance deteriorates significantly at higher currents. When compared with

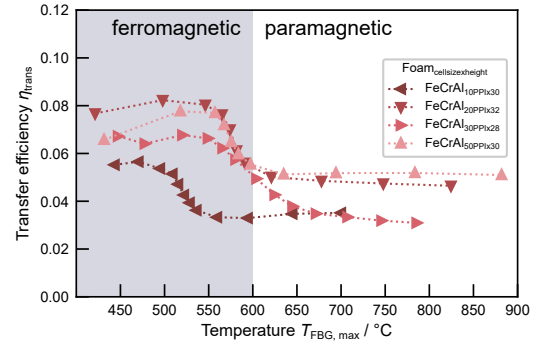


Figure 10: Transfer efficiency η_{trans} over maximum reactor temperature $T_{\text{FBG,max}}$ for different foam structures at $\dot{V}_{\text{CH}_4} = 100 \text{ Nml min}^{-1}$. Ferromagnetic and paramagnetic regimes marked with Curie Temperature of 600 °C according to [54].

the thermodynamic equilibrium in (Figure 9c), all observed conversions are slightly below their equilibrium conversion. The typical sigmoidal characteristic can be seen clearly for all foam structures. The deviation in conversion from the equilibrium one may be related due to kinetical effects. In conclusion, the methane conversion is not substantially limited by the reaction kinetics for the investigated flow rate of $\dot{V}_{\text{CH}_4} = 100 \text{ Nml min}^{-1}$. Rather, it is limited by the power input and thereby the generated structure surface temperature. As a consequence, the qualitative heating-conversion performance is as follows: 50 PPI > 20 PPI > 30 PPI > 10 PPI. This behavior will be further examined in the following.

Figure 10 shows the transfer efficiency, Eq. (5), over the maximum reactor temperature for the different foams. For all foams and temperatures below 500 °C, efficiencies range from 5% for the 30 PPI foams to 8% for the 20 PPI foams. While the temperature increases, all foams show a characteristic drop in efficiency of approx. 2-4%. The 20 and 50 PPI foams show a step sequent drop between 550 and 600 °C from around 8% efficiency to 5%. In contrast, the 30 PPI foam exhibits a gradual decrease from 7 to 3% in the temperature range between 550 and 660 °C. Similarly, for the 10 PPI foam, the efficiency decreases from 5 to 3% in the temperature range of 500 to 550 °C. For the 50 PPI foams, the efficiency remains nearly constant at about 5% for temperatures >800 °C. The characteristic decrease in efficiency can be attributed to the transition in magnetic behavior from ferromagnetic to paramagnetic at the Curie temperature, T_C , of the material, which is approx. 600 °C [54]. IH below T_C can be linked to hysteresis and eddy current heating, respectively. In contrast above T_C , IH relies solely on eddy current heating, which performance is highly dependent on the foam morphology and stack structure, i.e., height and number of stacked discs. This explains the temperature plateau or slight incline between approx. 500-600 °C in Figure 9b) as it is just below the Curie Temperature, where heating input power is decreasing towards reaching T_C .

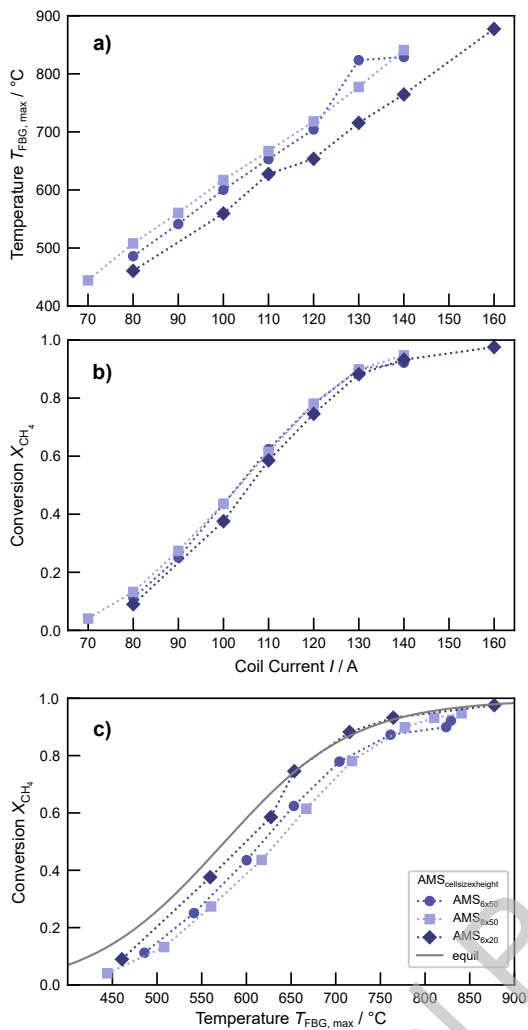


Figure 11: a) Reactor temperature $T_{\text{FBG,max}}$, b) reactor methane conversion X_{CH_4} over coil current I for different foam structures at $\dot{V}_{\text{CH}_4} = 100 \text{ Nml min}^{-1}$. c) Methane conversion X_{CH_4} as a function of $T_{\text{FBG,max}}$ with equilibrium conversion (calculated at $T_{\text{FBG,max}}$ and reactor pressure $p = 1.12 \text{ bar}$) for different AMS at $\dot{V}_{\text{CH}_4} = 100 \text{ Nml min}^{-1}$.

In summary, high methane conversions can be achieved with FeCrAl foams, demonstrating the general feasibility of combining this material class with IH for DRM. Furthermore, the results indicate that the maximum transfer efficiency of FeCrAl foams is confined to the ferromagnetic temperature range. In the case of DRM, this is disadvantageous, as the thermodynamic equilibrium allows relevant conversions only for temperatures above 600°C .

Finally, the optimal performance of the FeCrAl foam stacks is limited in this high temperature application due to the material properties, the microstructure, and the stack composition.

3.4. Reactive tests with 316L AMS

The three different additively manufactured structures, i.e., AMS_{6x50}, AMS_{8x50}, and AMS_{6x20}, were tested under IH reactive conditions with the same low flow rate of $\dot{V}_{\text{CH}_4} =$

100 Nml min^{-1} as the foams in subsection 3.3. The results of the measured temperature and methane conversion are presented in Figure 11. For all AMS, the maximum temperature increases nearly linearly with varying coil currents. In contrast to the FeCrAl foams, no plateau-like behavior is observed as the coil current increases. The 50 mm tall structures reach 50 K higher temperatures than the 20 mm structure. The influence of the cell size for the same structure height is minor. A larger cell size results in approx. 10 K higher maximum temperatures for the same coil current. At a coil current of 140 A, the maximum temperatures for AMS_{6x50} and AMS_{8x50} are 829°C and 841°C , respectively, whereas AMS_{6x20} reaches only 764°C under the same conditions. As the AMS are manufactured from 316L stainless steel, there is no ferromagnetic regime. The solely paramagnetic AMS can therefore be heated only by eddy currents, which depend mostly on the varying magnetic field strength regulated by the coil current and the coil frequency, which is constant. The linear relationship may be attributed to the complex interplay between increasing input power, increasing heat transfer driving force, increasing losses to the environment by radiation and convection, as well as the endothermic catalytic reaction. Further analysis is difficult, since especially heat losses of the uninsulated reactor are complex to characterize. Detailed modeling will be therefore developed in follow-up studies. However, the linear trend is clear and can also be perceived for the FeCrAl foams in the eddy current-dominated regime above 600°C , see Figure 9.

The conversion profiles for all AMSs, see Figure 11b), are very close and have the typical S-shape reaching full conversion for coil currents larger 150 A.

When compared with the thermodynamic equilibrium in Figure 11c), it can also be seen that the curves follow the sigmoidal shape of the equilibrium methane conversion. AMS_{6x50} and AMS_{8x50} do not reach full equilibrium conversion, respectively, which can be explained by inaccurate temperature measurements due to the reasons discussed in subsection 3.6. The short AMS_{6x20} shows equilibrium conversions at temperatures $>650^\circ\text{C}$, implying that $T_{\text{FBG,max}}$ refers to the actual catalyst temperature.

As stated in subsection 3.1, internal mass transport limitations within the washcoat layer are not expected. Furthermore, when comparing the conversions of AMS_{6x50} and AMS_{6x20}, which share the same cell structure, the conversion in Figure 11c) for the short AMS_{6x20} is slightly higher than that for the tall AMS_{6x50}. Therefore, limitations due to external mass transport or reaction kinetics can be excluded at the low flow rate of $\dot{V}_{\text{CH}_4} = 100 \text{ Nml min}^{-1}$.

Overall, it can be said that for the low flow rate of $\dot{V}_{\text{CH}_4} = 100 \text{ Nml min}^{-1}$, all structures reach equilibrium conversion. Therefore, conversion is only affected by temperature, meaning that input power is the crucial factor here. Hence, a closer look at the transfer efficiencies for the AMSs is essential. The results are presented in Figure 12 and show comparable efficiencies for all AMSs. A slight increase from 4 to 5% can be seen for the AMS_{6x50} and AMS_{8x50} over $T_{\text{FBG,max}}$, respectively. AMS_{6x20} displays the

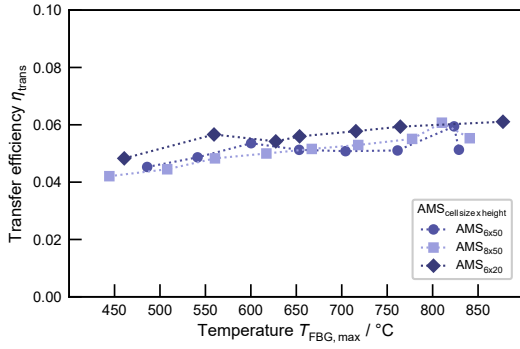


Figure 12: Transfer efficiency η_{trans} over maximum reactor temperature $T_{\text{FBG,max}}$ for different AMSs at $\dot{V}_{\text{CH}_4} = 100 \text{ Nml min}^{-1}$.

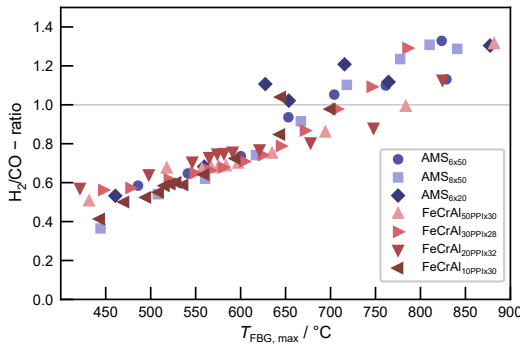


Figure 13: H_2/CO -ratio for all structures at varying temperatures $T_{\text{FBG,max}}$ at $\dot{V}_{\text{CH}_4} = 100 \text{ Nml min}^{-1}$.

highest efficiencies of nearly 6% for temperatures $>800^\circ\text{C}$ resulting from a lower PSU Power, which is discussed below. In contrast to the FeCrAl foams (Figure 10), no distinct dependence of transfer efficiency and temperature can be noted. This is in accordance with the eddy current heating of paramagnetic 316L, which is only weakly dependent on temperature [55].

3.5. Carbon analysis and DRM side reactions

As mentioned earlier, carbon formation occurred during the experiments, which is a well known phenomena for Ni-catalysts in DRM [56]. Main side reactions for carbon accumulation are methane cracking and Boudouard reaction, the former thermodynamically relevant at process temperatures above 700°C and the latter one dominating at low process temperatures below 700°C . [57]. To determine the influence of the abovementioned side reactions and the RWGS reaction, Figure 13 shows the H_2/CO -ratio for the conducted experiments at low flow rates of $\dot{V}_{\text{CH}_4} = 100 \text{ Nml min}^{-1}$.

As shown in Figure 13, a clear linear trend is observed for the H_2/CO -ratio. Values below unity are obtained at temperatures below approximately 700°C , whereas values exceeding unity are observed at higher temperatures. This behavior suggests a significant contribution of the RWGS reaction at low to intermediate temperatures, resulting in H_2/CO -ratio values below one. A contribution from the Boudouard reaction cannot be unambiguously confirmed.

Table 4

Results of Raman analysis for the spent carbon

Structure	$\frac{I_D}{I_G}$	$\frac{I_D}{I_D+I_G}$	$\frac{A_D}{A_G}$
FeCrAl 10 PPI	0.68 ± 0.47	0.36 ± 0.19	0.90 ± 0.47
FeCrAl 20 PPI	0.57 ± 0.12	0.36 ± 0.05	1.10 ± 0.09
FeCrAl 30 PPI	1.31 ± 0.12	0.57 ± 0.02	1.80 ± 0.16
FeCrAl 50 PPI	0.97 ± 0.02	0.49 ± 0.01	1.15 ± 0.02
AMS _{6x20}	1.14 ± 0.26	0.53 ± 0.06	1.55 ± 0.32
AMS _{6x50}	0.77 ± 0.18	0.43 ± 0.06	1.08 ± 0.21
AMS _{8x50}	0.67 ± 0.25	0.39 ± 0.08	0.91 ± 0.28

Nevertheless, the low H_2/CO -ratio is consistent with its occurrence, as carbon formation through CO consumption would further decrease the ratio, in agreement with previous reports [57]. At temperatures approaching 800°C , H_2/CO -ratio values greater than unity indicate the onset of methane cracking, leading to an excess production of hydrogen. In summary, the H_2/CO -ratio is strongly temperature-dependent; however, values close to unity can be reached by selecting an appropriate reaction temperature.

The carbon characterization results obtained by Raman spectroscopy are presented in Table 4, Figure 14, and Section S10 of the SI. For the AMS structures, where carbon deposits were analyzed after scraping from the surface, relatively consistent $\frac{I_D}{I_D+I_G}$ ratios between 0.39 and 0.53 were obtained. These values indicate the presence of graphitic carbon species containing a substantial degree of structural disorder [58]. Furthermore, the pronounced 2D band observed in the spectra is characteristic of defective graphitic carbon and is commonly associated with filamentous carbon, carbon nanofibers (CNFs), or nanotube-like deposits [59]. These findings are in good agreement with the known carbon formation behavior of Ni-based catalysts, for which the formation of filamentous carbon is frequently reported [57].

The results obtained for the FeCrAl foams show somewhat larger variations, with $\frac{I_D}{I_D+I_G}$ ratios ranging from 0.36 to 0.57. Nevertheless, these values likewise indicate the presence of defective graphitic carbon species. The results for the 20 PPI foam should be interpreted with caution, as the Raman signal intensity was relatively low compared to the background signal. To complement the Raman analysis, SEM investigations were performed on entire foam sections, which could be directly accommodated within the microscope chamber. This approach enabled verification of the carbon species on the washcoat surface and allowed a qualitative assessment of their spatial distribution. The majority of the observed carbon deposits exhibited filamentous morphologies, as shown in Figure 14, which is in good agreement with the Raman results and the broad variation in $\frac{I_D}{I_D+I_G}$ ratios. Several distinct filamentous carbon morphologies were identified, although no clear dependence on the foam PPI was observed. This finding is consistent with the broad distribution of Raman-derived disorder parameters and suggests the presence of heterogeneous carbon species

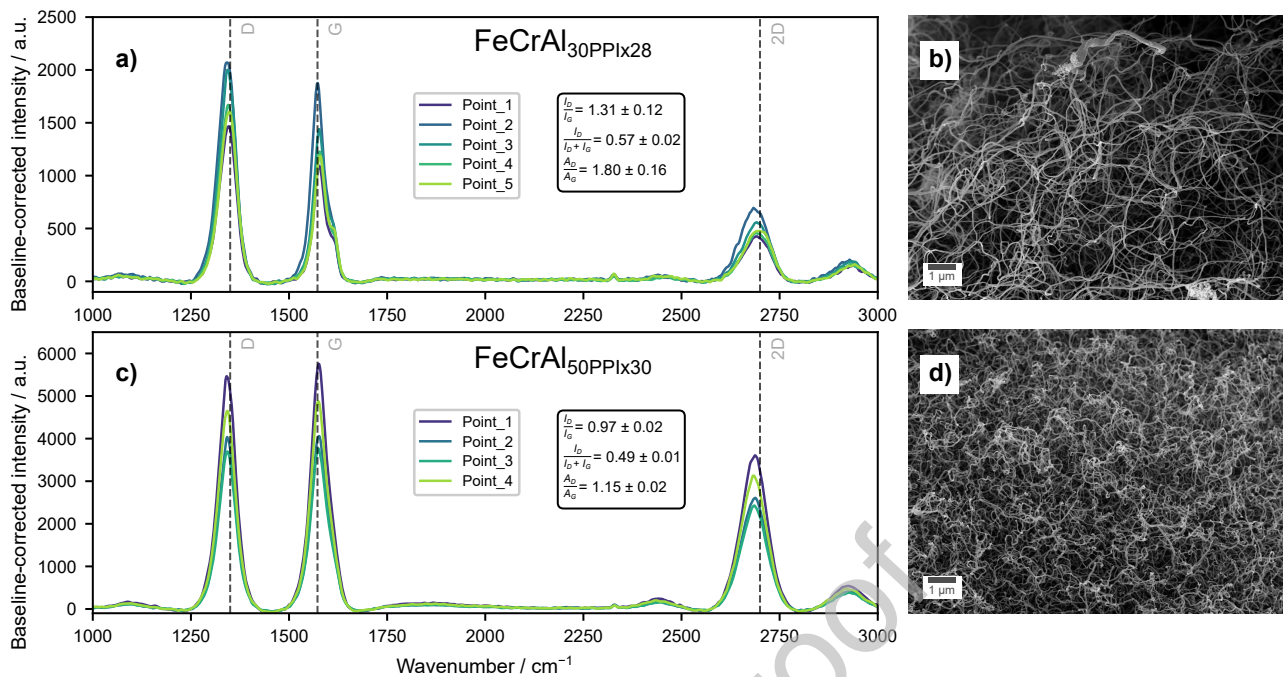


Figure 14: a) + c): Raman spectra of all FeCrAl foams (30, 50 PPI) with D, G and 2D band marked. c) + d): SEM images of spent carbon for each foam respectively. $\frac{I_D}{I_G}$, $\frac{I_D}{I_D + I_G}$, and $\frac{A_D}{A_G}$ from Table 4 have been included.

[60]. For example, comparison of carbon on the 30 PPI foams in Figure 14b) and the 10 PPI foams in SI S8 b) reveals pronounced differences in filament morphology, ranging from thin fibers to considerably thicker filaments, sometimes even within the same sample. No distinct spatial trends in carbon deposition were identified. Carbon filaments were observed both directly on the washcoat surface and within existing washcoat cracks, albeit with varying local densities, as illustrated in Figure 15a), c), d), and f). In addition, localized CNFs were detected at several positions, as shown in Figure 15b). Partial foam window blockage by carbon deposits was observed, particularly for the 50 PPI foam (Figure 15e). As carbon formation is strongly influenced by local temperature, Ni particle size, and the local C/H ratio, no direct relationship between structure geometry and carbon formation can be established from the present data [61, 60]. Nevertheless, the observed CNFs are known to contribute to catalyst deactivation through pore blockage, mechanical damage to the washcoat, and encapsulation of active Ni particles [57]. Due to the screening approach applied in this study, the metallic structures were exposed to different local concentration and temperature profiles. Future investigations should therefore assess carbon formation in situ to identify carbon-forming reaction conditions and to correlate local temperature and concentration effects with the carbon formation rate.

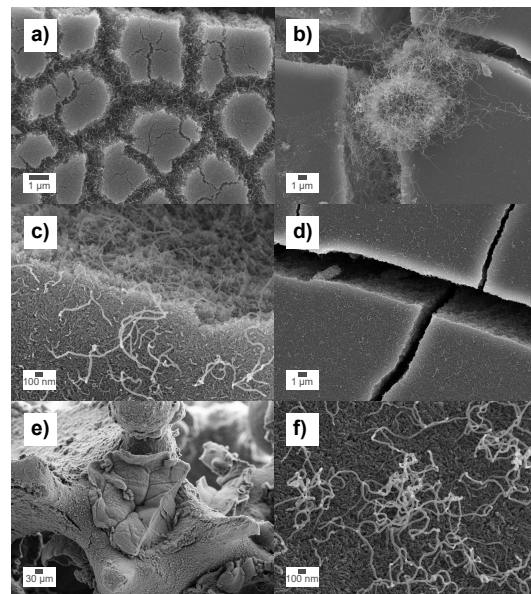


Figure 15: SEM images of a) + b): 20 PPI FeCrAl foam, c) + d): 10 PPI FeCrAl foam, and e) + f): 50 PPI FeCrAl foam.

3.6. Comparison of structures and transfer efficiencies in reactive tests conducted at elevated flow rates

As reported, FeCrAl foams and AMSs are capable of achieving full conversion under various coil currents and magnetic field strengths. For technical applications, high flow rates are preferred in order to obtain high transfer

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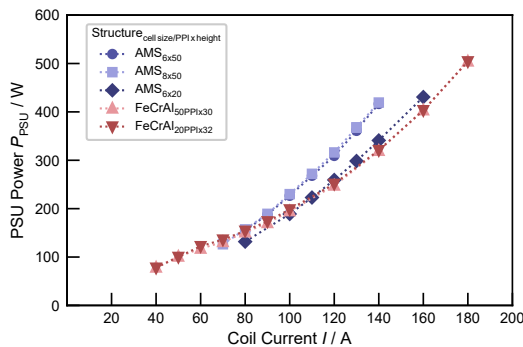


Figure 16: PSU power, P_{PSU} , as a function of set coil current, I , for different AMSs and FeCrAl foam structures. Since PSU power did not depend noticeably on the flow rate, all values are shown for a flow rate of $\dot{V}_{\text{CH}_4} = 100 \text{ Nml min}^{-1}$.

efficiencies and high production capacities. Therefore, experiments were carried out over a wide range of flow rates ($\dot{V}_{\text{CH}_4} = 100 - 400 \text{ Nml min}^{-1}$). For the sake of clarity, the 10 and 30 PPI foams have been omitted in further discussions, as their performance is significantly lower than the 20 and 50 PPI foams. The transfer efficiency is mainly influenced by the PSU power, the conversion, the temperature, and the gas/molar flow rate. PSU power is strongly dependent on the set coil current, which in turn affects the magnetic field strength and thereby the temperature and thus the conversion. Figure 16 displays the monitored PSU power, P_{PSU} , as a function of the coil current, I , for the various investigated structures.

Figure 16 shows that at a given coil current and magnetic flux densities, the AMSs require the most electrical PSU power. The larger AMS_{6x50} and AMS_{8x50} have a notably higher PSU power than the shorter AMS_{6x20}. AMS_{6x20} and the foams, i.e., FeCrAl_{50PPIx30} and FeCrAl_{20PPIx32}, show comparable PSU powers. Consequently, PSU power is generally dependent on the geometry and volume of the structure. A strong eddy current formation, which results in high transfer power and elevated temperatures, also distorts the magnetic field (according to Lenz's law) and requires higher PSU power to compensate for this effect. Larger structures also increase the PSU power explaining the larger PSU power of AMS_{6x50} compared to AMS_{6x20}. In conclusion, the PSU power depends on the overall setup and catalyst structure (as well as the coil geometry, which is fixed in this case). However, the influence of the structural configuration is relatively small compared to the influence of the coil current.

Furthermore, it is necessary to compare not only the PSU power and coil current, but also the transfer efficiency for different catalyst structures. Consequently, Figure 17 shows the transfer efficiency η_{trans} and conversion of specific structures, i.e., all AMSs and 20 and 50 PPI foams, at their respective optimal coil currents, which were selected to achieve substantial conversion in the former paragraph. All configurations in Figure 17 are set to a constant coil current with best efficiency at high conversions and varying flow

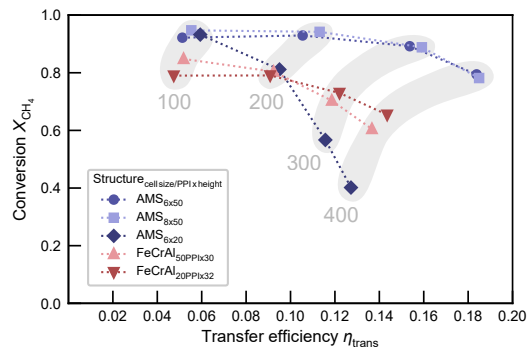


Figure 17: Methane conversion over transfer efficiency for various structures at increasing flow rates from $\dot{V}_{\text{CH}_4} = 100 - 400 \text{ Nml min}^{-1}$. Connected flow rates marked in grey. Each structure was set to the coil current with best efficiency at high conversions: AMS_{6x50}, AMS_{8x50} and AMS_{6x20} at 140 A. FeCrAl_{50PPIx30} and FeCrAl_{20PPIx32} at 160 A.

rates from $\dot{V}_{\text{CH}_4} = 100 - 400 \text{ Nml min}^{-1}$. For low flow rates of $\dot{V}_{\text{CH}_4} = 100 \text{ Nml min}^{-1}$, conversion is higher 80% for all structures with a transfer efficiency of approx. 5%. At this point, all the structures operate within a performance range where high reactions temperatures ($> 700 \text{ }^\circ\text{C}$) are reached. Due to similar PSU power at the different selected coil currents (140 A for the AMS and 160 A for the FeCrAl foams) and comparable high conversions of 80-90%, all efficiencies for low flow rates of $\dot{V}_{\text{CH}_4} = 100 \text{ Nml min}^{-1}$ are in the range of 5%. For increased flow rates, a general decrease in conversion and increase in efficiency can be reported. The decreased conversion can be attributed to a decreased structure temperature limiting the reaction due to the chemical equilibrium and partly due to decreased reaction kinetics. Interestingly, the decrease in conversion is highly dependent on the reactor structure. AMS_{6x20} shows a rapid decrease with a low conversion of 40% and an efficiency of 13% at $\dot{V}_{\text{CH}_4} = 400 \text{ Nml min}^{-1}$. In comparison, the foam structures, i.e., FeCrAl_{50PPIx30}, FeCrAl_{20PPIx32}, perform more stable with conversions between 60-70% and efficiencies of 14-15% at $\dot{V}_{\text{CH}_4} = 400 \text{ Nml min}^{-1}$. The FeCrAl_{20PPIx32} foam outperforms the FeCrAl_{50PPIx30} foam based on the higher conversion and efficiency.

Since the feed gas enters the structure near room temperature, an initial preheating zone is present before substantial DRM occurs. At higher gas flow rates, this zone extends further into the structure and reduces the fraction operating at elevated temperature. For the FeCrAl foams, this inlet region lies within the ferromagnetic regime below $600 \text{ }^\circ\text{C}$, where higher transfer efficiencies were observed. This can contribute to the higher conversion and efficiency compared with AMS_{6x20}. However, the structures also differ in length, with the FeCrAl foams extending over 30–32 mm compared with 20 mm for AMS_{6x20}. The observed performance differences can therefore be attributed to the combined effects of low-temperature heating behavior and available structure length.

The long AMSs (AMS_{6x50} and AMS_{8x50}) display the overall highest conversions and efficiencies for high gas flow rates. The elongated structure provides adequate preheating and a sufficiently long reactive zone to achieve high and stable conversions of 80% and efficiencies of 18% at a high flow rate of $\dot{V}_{\text{CH}_4} = 400 \text{ Nml min}^{-1}$.

Noble et al. reported energy efficiencies for laboratory-scale radio frequency induction heating reactors in the range of approximately 0.7–22% [62], in line with the reported maximum of about 23% [12] from Almind et al., depending strongly on the reaction, reactor configuration, operating frequency, and definition of efficiency. In this context, the maximum transfer efficiency of 18% obtained in the present uninsulated setup lies toward the upper end of the range reported for laboratory-scale IH reactor systems, supporting the conclusion that the AMS provide pronounced heating and catalytic performance. Lin et al. demonstrated a highly optimized IH system also targeting the electrical power supply and showed 85% for the RWGS reaction [20]. Although this is not fully comparable due to differences in PSU design, reaction, operating conditions, and milder temperature regime, it still illustrates the considerable potential for further efficiency improvements through optimization of both the reactor and the electrical system.

When comparing the tall AMSs (AMS_{6x50} and AMS_{8x50}) in Figure 17, it can also be observed that external mass transport limitations are absent. Since the external specific surface area of AMS_{6x50} is significantly larger than that of AMS_{8x50}, a higher conversion would be expected if external mass transport was limiting at the comparable temperatures reported above. As this trend is not observed, external mass transport limitations can be excluded. Consequently, the Damköhler number for the AMS must be close to zero, consistent with the behavior discussed in subsection 3.4.

As the investigated structure lengths differ, the hydrogen energy yield was additionally calculated from the data in Figure 17 and is reported in Section S11 of the Supporting Information. This metric combines structural, heating, and catalytic performance and confirms that the longer AMS configurations exhibit the highest overall performance under the investigated conditions.

Additionally, methane conversion was compared as a function of gas hourly space velocity (GHSV) in Figure 18, providing a more meaningful basis for comparison than the volumetric flow rate alone.

For each structure, the coil current was adjusted to achieve near-complete methane conversion at the lowest flow rate. The elongated AMS configurations (AMS_{6x50} and AMS_{8x50}) maintain methane conversions above 80% over the investigated GHSV range up to 5050 h⁻¹. Similarly, the FeCrAl foams exhibit stable conversion levels at GHSVs of up to approximately 8000 h⁻¹. Owing to its short length of only 20 mm, AMS_{6x20} reaches substantially higher GHSVs of up to 12627 h⁻¹. Under these conditions, methane conversion decreases to approximately 60%, reflecting the combined effects of limited residence time and the previously discussed preheating requirements.

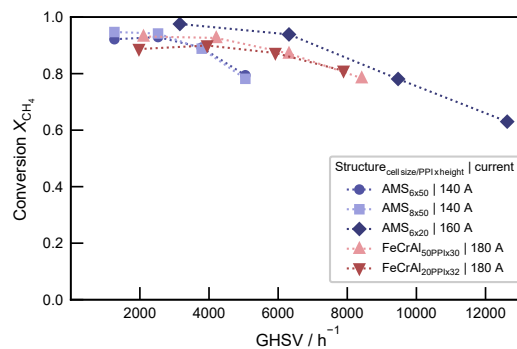


Figure 18: Methane conversion over GHSV for various structures at increasing flow rates from $\dot{V}_{\text{CH}_4} = 100\text{--}400 \text{ Nml min}^{-1}$. Each structure was set to the coil current with best efficiency at high conversions: AMS_{6x50} and AMS_{8x50} at 140 A. AMS_{6x20} at 160 A. FeCrAl_{50PPIx30} and FeCrAl_{20PPIx32} at 180 A.

Despite its significantly higher GHSV, AMS_{6x20} achieves methane conversions comparable to or exceeding those of the FeCrAl foams at similar coil currents. This observation highlights the superior high-temperature heating characteristics of the AMS structures, which are dominated by eddy current heating in the temperature range relevant for DRM. The results further indicate that reactor performance is not solely governed by catalyst loading, but also by the ability of the structure to efficiently generate and distribute heat, which includes heat transport by convection, conduction, and radiation. To further exploit the advantages of AMS structures while minimizing preheating limitations, future studies should investigate partially coated long AMS configurations, following the concept proposed by Zheng et al. [44]. Such an approach may allow efficient utilization of the coated structure while maintaining high methane conversion.

A basic heat loss model, which is described in the SI S9, indicates that for the AMS_{6x50} only 32 – 37% of the actual transfer energy is used. Based on the estimated loss contributions, overall transfer efficiencies of approximately 50 – 58% could potentially be achieved through appropriate thermal insulation of the reactor setup. In a recent study, maximum efficiencies of 11% under comparable DRM reaction conditions were reported [22]. An increase in transfer efficiency with increasing flow rate was also confirmed. For catalytic steam reforming of methane, Almind et al. showed maximum efficiencies of 23% with a sophisticatedly insulated NP based heating system [12]. This can be mainly attributed to the insulation, while this presented work is carried out in an uninsulated quartz reactor. Asmat et al. also reported that sufficient insulation can approx. double the transfer efficiency [22], which is consistent with the predictions of the heat loss model in SI S9.

Further improvements could be achieved by optimizing the catalytically active sites, for instance by employing noble metal catalysts (e.g., Rh, Ru), which exhibit enhanced catalytic stability, as reported by several authors [4, 63]. Such a transition would also increase catalytic activity, making

it particularly attractive for achieving higher hydrogen production capacity within a limited reactor volume, and thus supporting decentralized hydrogen production.

4. Conclusions

This work investigated electrified dry reforming of methane (DRM) using induction heating in combination with structured metallic catalysts. Two fundamentally different classes of structures, namely commercial FeCrAl open-cell foams and additively manufactured 316L structures (AMS), were evaluated with respect to their heating behavior, temperature distribution, catalytic performance, and energy transfer characteristics.

While FeCrAl foams performed well within their ferromagnetic operating range below 600 °C, they are less suitable for DRM due to the elevated temperatures required for industrially relevant methane conversions and the formation of hot spots arising from their inhomogeneous material geometry. In contrast, AMS structures proved to be highly effective susceptors at temperatures between 600 and 800 °C and under high flow rates, exhibiting comparatively homogeneous heating patterns. Transfer efficiencies of up to 18% were achieved at methane conversions exceeding 80%, demonstrating the superiority of structured metallic packings over NP based powder beds for induction-heated DRM.

Combined thermal imaging and Fiber Bragg Grating measurements revealed complex temperature distributions and localized heating effects within the structures. The results demonstrate that reactor performance cannot be explained solely by catalyst loading or geometric surface area, but is strongly governed by the interplay between structure geometry, eddy current formation, and heat transport phenomena.

Post-reaction characterization revealed predominantly graphitic filamentous carbon species, consistent with side reactions such as methane cracking and the Boudouard reaction. These findings emphasize the importance of catalyst optimization and long-term stability studies, for example through the use of noble-metal catalysts or tailored active-phase design.

A simplified heat-loss analysis indicated that only a fraction of the transferred energy is ultimately utilized for gas heating. This highlights the importance of thermal insulation and heat integration strategies for maximizing the efficiency of induction-heated reactors. Overall, the study demonstrates that electrified IH requires a fundamental redesign of the interplay between reactor geometry, catalyst, and heating mechanism.

Establishing quantitative structure–heating–performance relationships will be crucial for the rational design of next-generation induction-heated reactors and motivates future experimental and modeling studies at the interface of additive manufacturing, induction heating, and reaction engineering.

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Declaration of interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.