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Multidisciplinary Design Analysis for Wash-Coating of Additively Manufactured Metallic Monoliths

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Abstract

For the sake of environmental reasons, additive manufacturing of catalytic converters has been an attractive topic in both computational research and product development fields within the last few years. With respect to structured catalysts, monolithic designs manage convenient characteristics by providing a large surface area for catalytic reactions within a defined volume. Because of manufacturing limitations, conventional metallic monolithic geometries were limited to sinusoidal channels. Accumulation of the catalyst and washcoat in the corners of sinusoidal channels have negative influences on the washcoat geometry and mass-transfer. These reasons together with high manufacturing costs had limiting effects on expanding applications of metallic monoliths. With the emergence of rapid prototyping technologies, complex unity structures of metallic monoliths can be printed at once. Although flexibility in design and ease in fabrication both are satisfied with additive manufacturing, still operational conditions and reaction requirements must be met for approval of any prototypes. This research uses a multidisciplinary approach to study monolithic designs suitable for a specific reactor size. Besides, these designs must be 3D-printable and suitable for washcoating. Considering design challenges and innovations in additive manufacturing of metals, relationships between mathematics, structural integrity, coating and material aspects are considered for analyzing the multi-channel monoliths.

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Nomenclature

<i>KBE</i>	Knowledge-Based Engineering
<i>TPMS</i>	Triply Periodic Minimal Surfaces
<i>OFA</i>	Open Frontal Area
<i>CAT</i>	Catalytic Converter
<i>COAT</i>	Catalytic Coating

1. Introduction

Additive manufacturing (AM) technologies have created a revolution in product design and manufacturing. Highly complex and intricate designs can be incorporated into product development which were previously hard or impossible to be considered in traditional manufacturing methods. This increased level of ease in design and manufacturing can deliver products which are more functional, efficient and suitable for particular applications. Accuracy and customization are specifically important in healthcare, energy, aerospace and automotive industries. For these cases, customized, exact and reproducible

components are required and so demanded [1,2].

Production complexity and innovation are driven by the perception of competitive market pressures. To move at the same pace as the market, companies have to consistently offer new products with enhanced features, unique functionality and improved user experiences; while keeping the prices affordable. Development of complex products often requires exchange and collaboration between cross-functional groups from material science, engineering, design and software development areas. Therefore, effective multidisciplinary communication is critically important to successfully pitch an innovative idea to the pilot and ultimately to the competitive market [3,4].

Adopting modern manufacturing methods, such as AM, can impact the product development process. While highly customized designs can be rapidly prototyped via AM, development timeline can also be extend. Since with the increased complexity come bigger risks and uncertainties, there are also greater responsibilities for quality control [5,6]. Complex products in aerospace, automotive and energy industries must meet stringent quality requirements and safety standards. So, companies must invest in comprehensive quality assurance processes and demonstrate compliance with industry regulations.

This article points out importance of the washcoat as an inseparable part of catalytic converters, which is very often missing in current knowledge-based engineering (KBE) developments in additive manufacturing of catalytic converters. On the

other side, by integrating multidisciplinary results in practical workflows, using specialized software tools and fostering collaboration, development of catalytic converters via metal additive manufacturing can benefit from a holistic KBE approach. Such an approach ensures that expertise from different domains contribute synergistically to the optimization of catalytic converter designs for high-temperature applications.

Air quality standards ensure that more and more devices are equipped with catalytic converters (including non-road mobile machinery, ships and airplanes) [7–10]. Considering this need, modern versions of catalytic converters can be seen as an important application for AM. With this objective, we propose comprehensive multidisciplinary workflows to deal with mathematical, design, material and manufacturing aspects together. A simple architecture is also suggested for including all the required conditions in just one software. This helps in reducing manufacturing time, costs and risks along with ease of knowledge transfer.

2. State of the Art

Knowledge-based engineering (KBE) is a growing modelling approach in additive manufacturing, where integrating knowledge into design and production workflows can accelerate the efficiency and optimization of a process. KBE with the encapsulation of fluid dynamics (CFD), finite element analysis (FEA) and generative design tools creates the best practices for automating repetitive tasks to enable rapid innovations in the design and production of additively manufacturing parts [11,12].

Nevertheless, current applications of KBE are mainly looped around AI-based data mining, automation of designs and simulations, and consequent topology optimizations. They often lack a practice-customised approach, so design activities are started before freezing practical requirements of the preferred components. By the integration of initial and fluid requirements (from the user or/and device) into KBE, this gap can be bridged [13,14].

Deployment of AM technologies in energy sectors including catalytic converters is not without challenges. Material properties, surface quality and dimensional accuracy are strongly dependant on AM-process planning before being integrated into an application [15]. Here, KBE is a good strategy for automatic estimation of functional properties of AM-parts. Besides, in the manufacturing of catalytic converters, washcoating is fundamentally important. The washcoat by providing a porous and high-surface-area substrate allows for more catalytic active sites, promoting efficient catalytic reactions [16]. Therefore, AM-manufactured monolithic parts are required to be washcoated after simulation-based design/topology optimizations and before any catalytic applications. Here, a combination of KBE from proven concepts and unique experiments can be seen as a multidisciplinary approach for design analysis, continuous learning and improvements.

While research and development in additive manufacturing and chemical engineering are considerably improving, the seamless integration of these fields faces obstacles related to multidisciplinary and collaborative-decision-makings workflows along with the complex nature of chemical reactions and processes. Hybrid workflows based on both KBE and prototyp-

ing can facilitate knowledge exchange and bring significant advantages to the product development. Based on our experience in the mentioned topic, a hybrid workflow is introduced. This workflow tries to mention all the inclusive subjects and suggest how to approach them. Besides, scalability considerations are not underestimated.

3. Methodology

Metal additive manufacturing of structured catalytic converters deals with multiple perspectives. Considering both challenges and innovations in additive manufacturing of metals, relationships between mathematics, structural integrity, coating and material aspects are considered for analyzing the multi-channel monoliths. The goal is to utilize additive manufacturing for 3D-printing of metallic monolithic bodies. However, production is not completed without coating them via a selected catalyst and final testing of the catalytic performance under a defined reaction.

This initial workflow is shown in Figure 1. Although this idea in the nutshell seems quite straightforward, complexities are hidden inside single blocks while interacting with neighboring components.

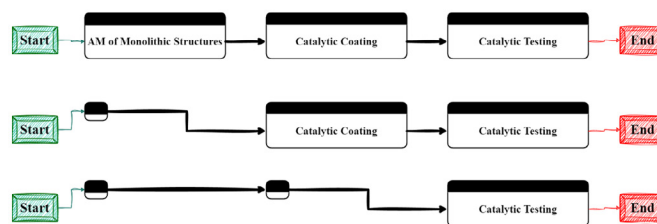


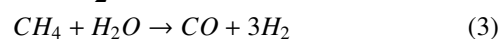
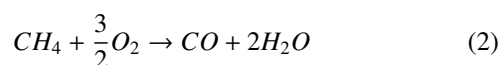
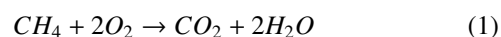
Fig. 1. Initial building blocks for AM of structured catalytic converters

3.1. Start Block

The starting point (or motivation) is defined by the reaction, through chemical engineering/chemistry rules. Considering catalytic oxidation of methane, the probability function P is defined. Variables a, b, c, d, e are respectively oxygen: methane feed ratio, loading of precious metal on the support, nature of the support (alumina, zirconia, etc), particle size of the precious metal, and catalyst pre-treatment. C_1 is a constant defining the precious metal. In this example by using palladium, $C_1 = Pd$.

$$predict_reaction_type = P(C_1, a, b, c, d, e)$$

The results from probability function P_1 can fall into one or more of reactions 1-5 (reaction notations from [17]):



In principle, selection of a catalyst is a matter of the specific

chemical reaction, temperature range, poisoning resistance, and composition of the involved reactants. For catalytic oxidation of methane, palladium and/or platinum catalysts supported in alumina and zirconia are appropriate [18]. This means based on the proven concepts, compatible metals with alumina and zirconia are required for AM of the monolithic reactors.

3.2. AM of Structured Catalytic Converters

After freezing the reaction type (complete oxidation of methane or reaction 1), design and manufacturing starts. In fact, design and topology are not apart from mathematics and manufacturing is the matter of AM process parameters and material selections. Nonetheless, simulations by incorporating fluid dynamic rules and reaction kinetics which are happening in the proposed monolithic reactor structures (a combination of geometrical design and surface texture characteristics which are dependant on AM process parameters) are patterns for predicting and optimizing performance of the catalytic converter designs.

The result from *-AM of Monolithic Structures-* are communicating with the results from *-Catalytic Coating-* and continuously learning from them. Considering the experimental challenges of coating step (coming from chemistry and nature of the materials), this block has a low-level tendency towards learning and therefore dominates the manufacturing. In the meantime, although the variety of materials for AM are increasing, they are still limited. Consequently, the next two blocks in Figure 1 have more interactions with each other to be considered separately. Figure 2 demonstrates this newly defined relationship which is multidisciplinary, interactive and learns from experimental Demos.

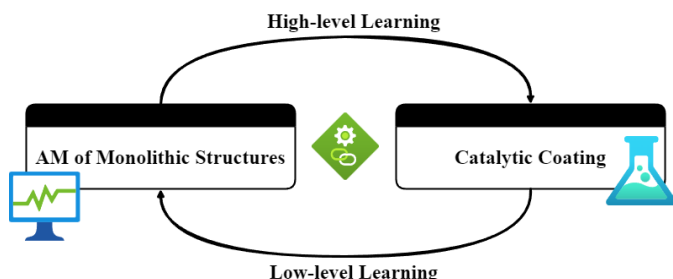


Fig. 2. Required iterative interactions between AM and coating steps

A combination of low-level image processing algorithms such as Sobel operator for edge detection, with a high-level approach which is a combination of generative design and reinforcement learning for topology optimization to maximize the surface area for washcoating are both involved in AM of catalytic converters. In this case, convolutional neural networks (CNNs) can be used for demonstration of a hierarchy of features, starting from washcoat thickness to topology optimization based on metal-ceramic adhesion (a measure of coating consistency) predictions.

For instance, open frontal area (OFA) plays a crucial role in the performance of catalytic converters. OFA refers to the portion of the surface of the catalytic substrate which is available for flow of the gas systems. Shape and distribution of OFA are simply outcomes of design and topology. This single parameter is also a crucial factor in the multifaceted performance of catalytic converters, influencing gas flows, thermal dynamics, back-pressure and the effectiveness of the catalytic coating

[19].

Fig 3 shows how easily by just changing arrangement of the circular channels, topology of a monolith can change. This simple change affects the value of OFA, cpsi (cells per square inch) and geometrical surface area [20].

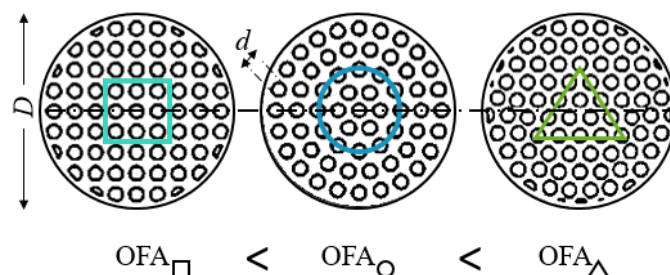


Fig. 3. Simple topology optimization by changing the arrangement of circular channels from rectangular to circular to triangular to increase OFA. Diameter of the circular cross-section (D) is constant, and all the circular channels are the same.

To our knowledge, specific standards that define which geometries are the best for designing of catalytic converters do not exist. Instead, geometry and design of catalytic converters are typically guided by engineering principles, practical considerations and industry practices. To create the highest possible surface-area within a limited cylindrical volume, circular channels are not the best; since a big portion of OFA stays untouched (shown in Fig 4). To address this limitation structures with more intricate geometries are required.

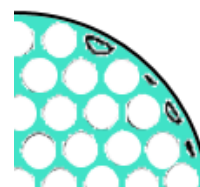


Fig. 4. Incompatibility of circular channels for catalytic coating. Only the white areas (OFA) contribute in reactions.

Triply periodic minimal surfaces (TPMS) have the big advantage of providing a high surface-to-volume ratio. This feature makes TPMS structures suitable designs for flow distribution, electricity and heat conduction applications. But when it comes to structured catalytic converters, corresponding KBE studies relying solely on pure FEA/CFD simulations together with generative design algorithms not always proceed towards successful results in practice. To study these complex designs, the importance of multidisciplinary design evaluations is significantly important. Consequently, studying the washcoat via advanced microscopes and image processing algorithms play crucial roles.

Theoretically, an ideal design for a monolithic catalytic converter is a design with the lowest possible pressure drop while having the highest possible mass-transfer. A detailed relationship between pressure drop and mass transfer inside structured monolithic catalytic converters depends on flow conditions (Reynolds number), fluid properties (the gases in reaction 1); and characteristics of the porous medium (alumina washcoat) such as pore size distribution, tortuosity and permeability. However, often optimizing pressure drop may adversely affect mass-transfer (and vice versa). This trade-off poses the first challenge in designing monolithic designs due to the need of

balancing these two conflicting requirements.

Theoretically, TPMS designs can offer unique advantages of high surface-to-volume ratio, enhanced mass transfer and optimized flow distributions within monolithic catalytic converters [21]. Beyond KBE design and numerical optimizations, metal additive manufacturing of high resolution TPMS designs with thin-and-porous walls (about 0.2 mm for laser powder bed fusion-LPBF) and the corresponding post-processing steps are not challenge-free.

If additively manufactured TPMS parts are meant for catalytic applications, they are required to be washcoated. Here, taking a multidisciplinary approach based on practice is required to plan the best coating approach(es). Two examples of our optimized TPMS-based topologies are shown in Figure 5. To increase the accessible surface-area for the washcoat suspension, rotation of Gyroid-based TPMS structures is a promising approach. Although TPMS structures based on Schwarz-P cell have very low pressure drops [22], support removal and post-processing of small size samples was costly. Therefore, by using the same methodology an alternative version was printed by a ReaLizer SLM 125 machine.

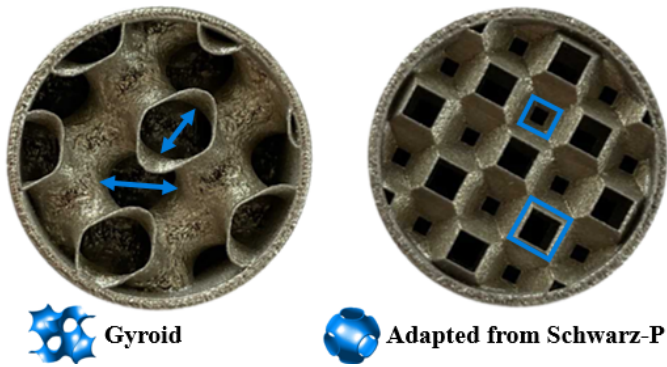


Fig. 5. Topology optimization for the purpose of facilitating washcoating (left) and reducing the support removal and post-processing (right). The blue color is used to refer to accessible open areas required for washcoat deposition (reproduced from [10]).

After optimizing topology of the TPMS designs, the question of coating reflects back. Adhesion of the washcoat ceramic (such as alumina) to the 3D-printed monolithic structures is indeed a critical step in the manufacturing of catalytic converters. Effectiveness of the catalytic converters heavily depends on the uniformity and durability of the washcoat. In relation to KBE approaches for design optimizations, it is essential to evolve beyond idealized cases. Integrating real-world considerations - such as non-uniform distribution of the washcoat thickness, methods and material aspects for improving adhesion - into simulations are crucial for more accurate and predictive models.

Apart from its vital importance, KBE approaches for design optimization only utilize FEA/CFD simulations based ideal cases with uniform distribution of the washcoat thickness, and the material aspects of adhesion are not fully considered.

The mentioned explanations were supposed to put emphasis on collaborative research for metal additive manufacturing of structured catalytic converters. Due to the many overlaps between computer-based and practical aspects, an iterative design and incremental build model (more information in [23]) is more suitable than independent decision-making strategies. Figure 6 shows an alternative agile workflow instead of the initial wa-

terfall model in Figure 1. In the next step, we define AM_CAT and CAT_COAT libraries and their corresponding modules and functions.

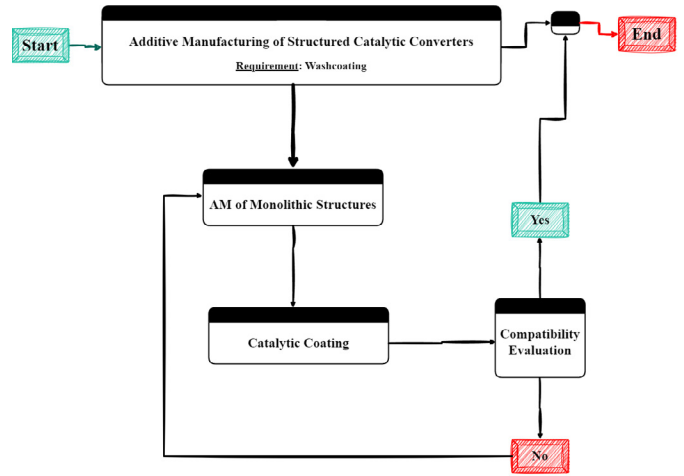


Fig. 6. Agile workflow: two-sided arrows show bidirectional flow of information

3.2.1. Additive Manufacturing of Monolithic Structures

This block is represented by a library called AM_CAT which has three modules shown in Fig 7. A combination of Deslib, Simlib and Manlib should be sufficient for algorithmic based on parametric design operations, to implement virtual experiments (FEA, CFD) and for the selection of AM process parameters and materials (process and material modelling).

Design library or Deslib supports parametric, 3D and surface modelings, customization and scripting like a computer-aided design (CAD) software. Simulation library or Simlib integrates FEA tools for material modelling, structural analysis and solid mechanics simulations. Multi-physics simulation tools for studying heat and mass transfer, fluid flow and other multi-physics phenomena are also included.

While Deslib and Simlib are both merged into one package, the compatibility problems when transferring files from one software (design files to FEA/CFD software) to the other are automatically solved. In a normal case, FEA/CFD simulations which are done inside professional software such as Abaqus/COMSOL do not always lead to the same results as FEA/CFD simulations which are performed with available CAD software. This is coming from different order of elements (linear in CAD software versus quadratic in Abaqus), meshing strategies and solver algorithms. However, these issues can be fixed by using the AM-CAT library.

Manufacturing library or Manlib is a customizable applica-

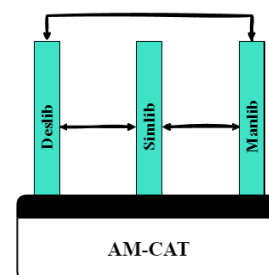


Fig. 7. AM-CAT library and its three modules

tion programming interface (API) similar to slicing software that allows users to interact with the 3D-printing machine. It also has an additional feature related to the washcoating process, which is explained in the next section.

3.2.2. Catalytic Coating

In practice, washcoating involves applying a thin layer of catalytically active materials (suspensions of porous ceramics with a certain percentage of precious metals) onto the surface of a ceramic/metallic substrate via a coating method (dip-coating, stencil printing, electrophoretic deposition, etc). The washcoat by providing a large, porous and structured surface supports accommodating nanoparticles of precious metals to improve the reaction kinetics. This results in more effective conversion of pollutants into less harmful substances (conversion of hydrocarbon like methane).

Surface finish (porosity and roughness) of additively manufactured metallic parts is indeed more compatible towards coating than the texture of conventional metals [10]. However, still porosity and roughness of the washcoat ceramic and metal powders for AM are not in the same scale. An alumina washcoat can have a particle size of $0.05 \mu\text{m}$ [24], while most of the metal powders for laser powder bed fusion of metals, binder jetting and direct energy deposition have particles in the order of $10\text{-}50 \mu\text{m}$ [25,26]. Therefore, an additive manufacturing technology which can utilize small (ideally $0.05 \mu\text{m}$) metal particles is fitting better manufacturing of catalytic converters. Therefore, Manlib also considers a compatibility relationship between washcoat particle size, metal particle size and the applied AM technology.

CAT-COAT block is rather considering practical aspects and imposing its needs on Manlib library, in parallel with that, design and simulations can upgrade to more realistic cases. In practice, porosity, specific surface area and thickness of the washcoat have strong influences on catalyst selectivity, reaction kinetics, diffusion and mass transport phenomena inside monolithic reactors. Thicker washcoats may impact selectivity, limit diffusion and alter mass transport characteristics [27]. In particular about washcoated monoliths made by metal AM, the thickness of the washcoat can interact with the complex geometries and impact diffusion of the reactants and products.

Thickness and uniformity of the washcoat is a matter of the contact angle between the washcoat suspension and AM-metal, wetting properties of the AM-metal and capillary rise in porous walls of multichannel monoliths. Among common metals, aluminum and nickel have relatively high surface energies and good wettability [28]. Therefore, metal alloy powders for AM which cover a percentage of these elements should also lead to better washcoat depositions. Design aspects of AM can also improve coating adhesion by influencing capillary action.

The deposited suspension has to be cured at high temperatures (above 500°C for γ -alumina [29]). The compatibility between 3D printed metal and ceramic washcoat at high temperatures requires exact monitoring in terms of thermal expansion coefficient, ceramic phase changes, oxidation and corrosion of metal monoliths at high temperatures. Besides, metals generally have higher ductility and toughness, while ceramics are known for their hardness and brittleness. In summary and as can be seen in Figure 8, CAT-COAT and AM-CAT blocks have many back-&-forth interactions until when the compatibility evaluation block

is satisfied.

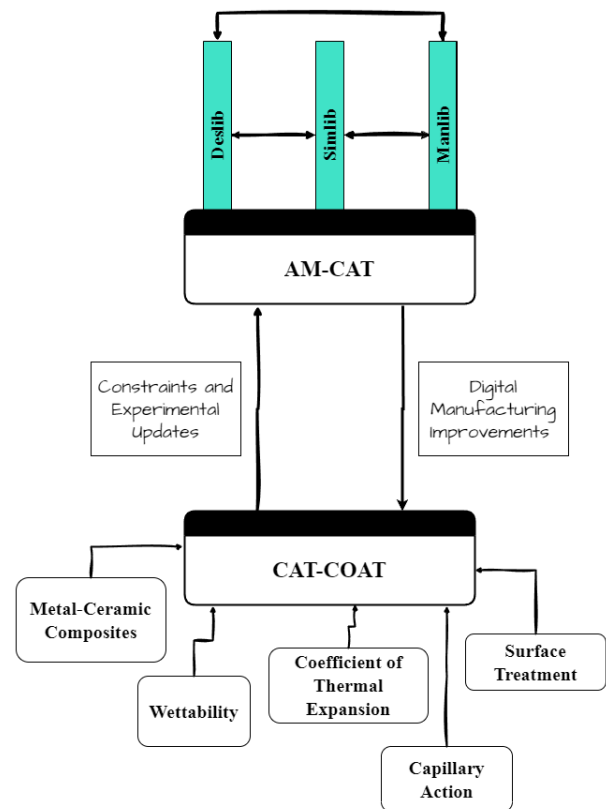


Fig. 8. Interactions between AM-CAT and CAT-COAT libraries

3.2.3. Catalytic Testing

This block as the final production step is dependant on compatibility evaluation block. Similar to the CAT-COAT, catalytic testing block is composed of numerical simulations and experimental validations. In fact, it is an experimental version of the previously performed CFD simulations in Simlib. The results of reactor tests, such as residence time distribution (RTD) and pressure drop can be used as a final statement for evaluation of design/topology of additively manufactured metallic monoliths. Gas conversion can be measured through analytical techniques such as gas chromatography (GC). A high conversion rate suggests that the catalytic active sites are effectively utilized. So the washcoat has been uniformly distributed and it has a good adhesion to the channels of the monolith.

4. Discussion and Conclusions

Multidisciplinary design analysis for washcoating of additively manufactured metallic monoliths is a dynamic field which requires collaboration and integration across diverse domains. Meanwhile that developments in computational modeling and AM technologies have propelled design of catalytic converters, several challenges are still persisting. For comprehensive understanding of the design requirements, critical considerations including intricacies of material-process interactions, fluid dynamics affecting coating uniformity and comprehensive optimization algorithms are needed. For the reliability of multidisciplinary design analyses, bridging the gap between simulated ideal cases and real-world complexities is indispensable. For implementing a good manufacturing practice in AM

of catalytic converters, design and simulation aspects, material control in 3D fabrication and coating, reaction conditions and testing must be all considered simultaneously and comprehensively. By far, bi-functionality has been covered up-to some extent by generative design tools. However, an informative platform or a software package which covers all the reality system requirements is missing. A unity multidisciplinary software package combined with practical obstacles can avoid interoperability issues happening while transferring files (design, simulation, data) between different software while learning from experiments. Considering the motivation, this work gave an introduction about objective constraints for design and topology of monolithic catalytic converters. It highlighted the need for structuring the monoliths, which means washcoating them with a material with a high specific surface area suitable for accommodating noble metal nano-particles (the main catalyst). CFD simulations which combine heat and mass transfer inside porous medium with the concept of heterogeneous catalytic reactions (chemistry) can give powerful insights for design optimization. Besides, the importance of numerical investigations on the impact of washcoat distribution is an added value. Combining knowledge-based engineering (KBE) and prototyping in a hybrid workflow can bring significant advantages to product development processes. This synergy creates a dynamic and efficient product development process. It leverages the strengths of both approaches, combining automated design with physical validation, leading to improved designs, reduced development cycles, and ultimately, more successful products. Therefore, first we stated a realistic and customizable workflow while defining different levels of learning flexibility for each manufacturing step. An agile collaboration framework was suggested to break manufacturing challenges into sub-goals. This framework can be used as a draft of a software package dealing with multiple aspects of AM of structured catalytic converters. This structured breakdown provides a detailed guidance for researchers aiming to conduct a multidisciplinary design analysis for the wash-coating of additively manufactured metallic monoliths. Each section is designed to contribute to a holistic understanding of the topic, combining theoretical insights, computational analyses and experimental validations.

Acknowledgements

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