Investigation of additive-manufactured catalysts in the decomposition of hydrogen peroxide

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Highlights

- Investigation of additive-manufactured woodpile structures in the H_2O_2 decomposition.
- Relationship between catalytic activity and geometric surface area.
- Influence of the macroporosity on the catalytic activity.

1. Introduction

High-concentration hydrogen peroxide (H_2O_2) is proposed as a low-toxic alternative for generating thrust in the space industry. During its decomposition, oxygen is produced, and a significant volume expansion occurs. Therefore, catalysts with a low pressure drop but a high geometric surface area are preferred. Here, monolithic honeycomb catalysts come into play. They consist of parallel straight channels and fulfill the upper requirements. But since there is no connection between the channels, there is no mass transfer in the radial direction. Woodpile structures are one possibility to overcome this limitation. They consist of layered strands with interconnected channels. In this work, woodpile structures with varying macroporosities and geometric surface areas, both values derived from the macroscopic geometry, are investigated, and their influence on the conversion of H_2O_2 is analyzed.

2. Methods

Woodpile monoliths of aluminum oxide were manufactured using the direct ink writing technique. Impregnation with 0.3 wt-% platinum (Pt) leads to suitable catalysts for the decomposition of H_2O_2. The Pt loading and distribution on the surface were checked using ICP-OES measurements and microscopic imaging. Catalytic tests were performed in a plug flow reactor with a solution of 4 wt-% H_2O_2 in water. The pH of the solution was adjusted to 2 using hydrochloric acid. The monoliths were wrapped with Teflon tape to prevent bypass flow. The conversion was calculated for four volume flows ranging from 7.7 mL min^{-1} to 30.8 mL min^{-1}.

3. Results and discussion

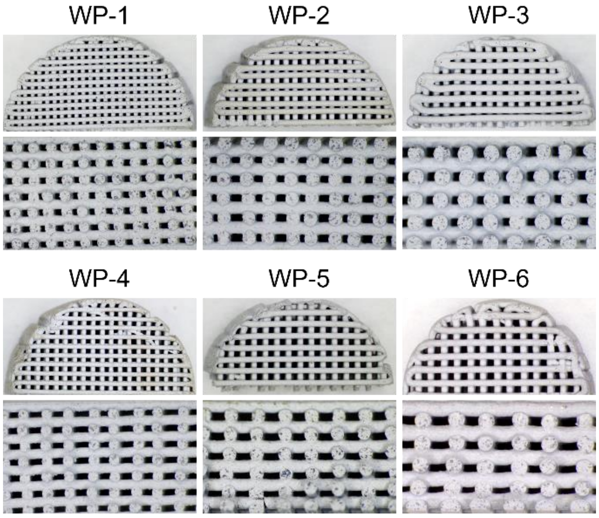

Figure 1. Microscopic images of the monoliths are shown, with top views above and side views below for each sample.

The different printed woodpile structures are depicted in Figure 1. From left to right, the nozzle diameter increases. The top row displays monoliths with 50% macroporosity; the bottom row shows those with 60%. A uniform distribution of platinum is observed on both the outer surface and inside the monolith, but a higher concentration is found inside the strand pores. Moreover, a decreasing number of strands can also be observed with increasing nozzle diameter, as well as with decreasing infill ratio. The influence of this observation on the geometric surface area and the macroporosity is shown in Table 1.

Table 1. Macroporosity, geometric surface area, and Pt loading for the investigated samples.

Sample	WP-1	WP-2	WP-3	WP-4	WP-5	WP-6
Macroporosity / %	50	50	50	60	60	60
Geometric surface area / m^{-1}	5300	4200	3600	4200	3400	2900
Pt loading / wt-%	0.35	0.22	0.29	0.32	0.29	0.34

As expected, a higher number of strains using the same nozzle decreases the macroporosity while the geometric surface area increases. For different nozzles and the same infill ratio, the macroporosity remains nearly constant with an increasing number of strains, as the increase is counteracted by the enlargement of the nozzle size. Thus, the macroporosity is mainly influenced by the infill ratio rather than by the nozzle diameter. Since smaller strains have a higher surface area-to-volume ratio, both an increase in the infill ratio and a smaller nozzle diameter result in a higher surface area. The Pt loading for all monoliths is in the range of the desired loading of 0.3 wt-%. Figure 2 illustrates the corresponding results of the conversion of H_2O_2 for increasing inlet volumetric flow rate.

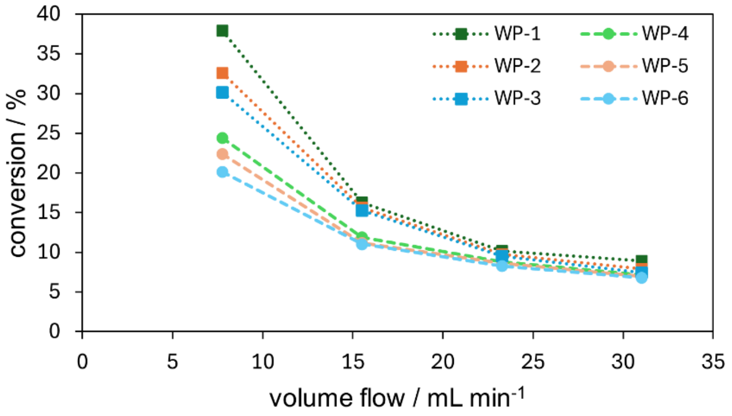

Figure 2. Conversion of H_2O_2 vs. inlet volume flow for samples with different macroporosity and geometric surface area.

As can be seen, the conversion decreases with increasing flow rates. The reason is that higher flow rates result in shorter residence times in the catalyst bed, and the H_2O_2 has less time to react. Furthermore, a higher geometric surface area increases the conversion. This is explained by more Pt being located directly on the surface of the strands, where the H_2O_2 does not need to diffuse into the pores. Moreover, the dispersion of the Pt may be higher due to a greater available surface area for anchoring. An increase in macroporosity results in a decrease in conversion because the infill and the weight of the monoliths decrease. Thus, there is less Pt on the monolith in total, resulting in a decrease in conversion.

4. Conclusions

Woodpile structures with different macroporosities and geometric surface areas were printed, and the conversion of the H_2O_2 decomposition was determined. While higher geometric surface areas lead to higher conversion, higher macroporosities exhibit lower conversion. For the last parameter, the total amount of Pt on the monolith is also an influencing factor. Therefore, it will be investigated in future work, as well as the influence of the specific surface area.

Keywords
Additive-manufacturing; structured catalyst; hydrogen peroxide decomposition.