Hierarchal zeolite supported on cellular foam catalysts for process intensification

Xiaolei Fan,[1,*] Yilai Jiao,[2]

1 School of Chemical Engineering and Analytical Science, The University of Manchester, Oxford Road, Manchester, M13 9PL, United Kingdom

2 Institute of Metal Research (IMR), Chinese Academy of Sciences, 72 Wenhua Road, Shenyang 110016, China
**Corresponding author: xiaolei.fan@manchester.ac.uk*

Highlights

- Novel methods developed for preparing hierarchal MFI zeolites/SiC catalysts.
- Novel MFI zeolites/SiC catalysts for process intensification of MTP process.
- Fe-ZSM-5/SiC catalysts for environmental catalysis.

1. Introduction

Cellular ceramic foams [1-4] have gained increasing popularity in chemical engineering, especially as catalyst supports, due to their irresistible combination of attractive features, such as, low pressure drop (in comparison with fixed beds with the same exchange area), and enhanced mixing (in comparison with monolithic reactors). Specifically, for continuous-flow catalytic processes with liquid-or multi-phases, in addition to the low pressure drop, enhanced transport phenomena such as mass and momentum transfer are also mandatory because the diffusivity of species in the liquid phase is generally five orders of magnitude lower than one in the gas phase. Therefore, cellular foams are attractive candidates for developing new multiphase catalytic processes. Silicon carbide (SiC) is a popular substrate for the preparation of cellular foams due to its unique properties of lightweight, high mechanical strength (hardness = *ca.* 2800 kg/mm^2), high oxidation and chemical resistance, good thermal conductivity (360–490 W/m·K) and possibility of surface modification. Advanced manufacturing process such as the controlled reaction bonding sintering method has been developed [3] which can produce SiC ceramic foams with the excellent mechanical processing property and low cost (comparable to monoliths) making SiC foams promising enablers for process intensification of catalytic reactions. Herein, the recent development of structured MFI zeolites/SiC foam catalysts is presented including (i) the design of structured zeolite on SiC foam catalysts for practical applications; (ii) the advanced characterization of hierarchically structured foam catalysts; (iii) the process intensification of catalytic reactions using MFI zeolites/SiC foam catalysts.

2. Methods

SiC foams were prepared by controlled reaction bonding sintering method, as shown in Figure 1. MFI zeolites/SiC foam catalysts were prepared using different methods including direct hydrothermal synthesis, microwave assisted synthesis and dip coating. The developed catalysts were characterised extensively using various techniques and assessed using the methanol-to-propylene (MTP) and catalytic wet phenol oxidation (CWPO) reactions.

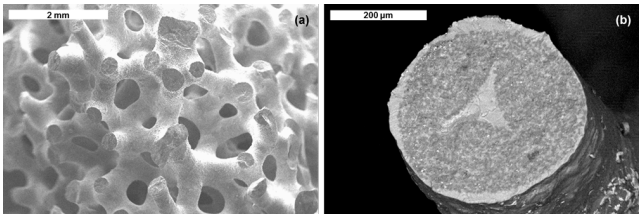

Figure 2. SEM micrographs of open cell SiC foams: (a) microstructure of the open-cell SiC foam and (b) the cross section of the SiC strut.

3. Results and discussion

Different strategies such as post-synthetic TPAOH vapor-phase transport (VPT) modification and creation of intrazeolitic aluminum (acidic) gradient were developed to improve the catalytic performance of MFI zeolites/SiC catalysts in MTP reactions. Combined with the intrinsic property of macroscopic SiC foams, these modified MFI zeolites/SiC catalysts demonstrated an excellent activity in catalytic MTP reaction

(Figure 2), surpassing the state-of-the-art hierarchal ZSM-5 monolith catalyst. Additionally, the coke formation was significantly retarded due to the improved design of the structured catalysts (Figure 3).

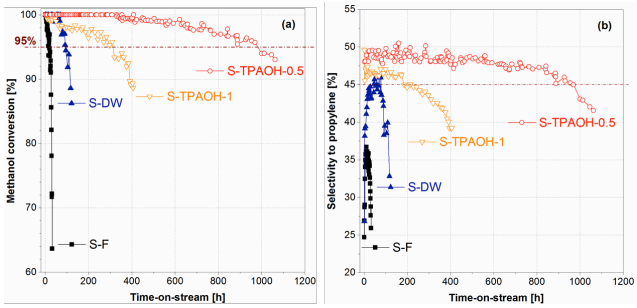

Figure 2. (a) Methanol conversion and (b) selectivity to propylene as a function of time-on-stream over TPAOH modified MFI zeolites/SiC foam catalysts.

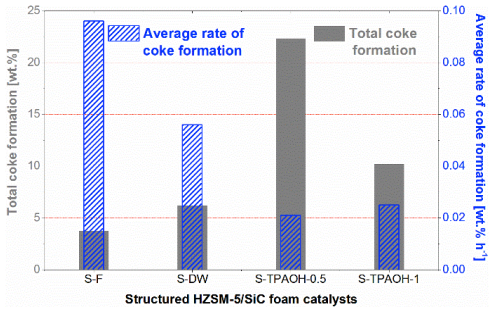

Figure 3. Coke formation of structured MFI zeolites/SiC foam catalysts after MTP reactions.

Intra-framework ferrisilicate/SiC catalysts were also developed using a hydrothermal method, showed good performance in catalytic wet peroxide oxidation of phenolic compounds under flow conditions. The developed ferrisilicate/SiC catalysts showed good stability in the cyclic test of total 24 hours with high conversions of phenol and TOC (Figure 4).

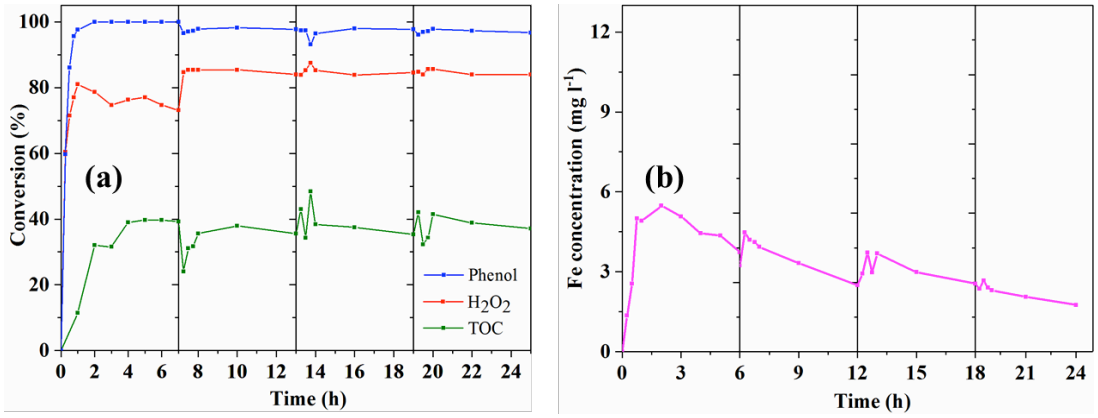

Figure 4. Stability evaluation of the developed ferrisilicate/SiC catalyst using catalytic wet peroxide oxidation of phenol as the model reaction: (a) conversions and (b) iron leaching under the continuous flow condition. Conditions: C_{phenol} = 100 mg/l, T = 80 °C, F = 1 ml/min, cell size of the foam bed = 80 µm.

4. Conclusions

We demonstrate various methods of preparing and optimising MFI zeolites supported on SiC foams as structured catalysts for intensifying catalytic reactions. Good catalytic performance was achieved by the developed hierarchically structured catalysts for the selected model systems, suggesting that SiC foams are promising catalyst carriers for practical applications.

References

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Keywords
Structured catalysts; SiC foams; MFI zeolites; Process intensification.