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Preface

Editorial

The well-established field of zeolites has been revitalized in recent years by a wave of research targeting improved accessibility and transport of molecules in these microporous materials. One major reason for this trend is related to the use of suitably modified zeolite materials as new or improved heterogeneous catalysts in industry. Despite being dressed with a truly unique combination of properties, zeolite catalysts appear under-utilized in many industrial reactions due to diffusion constraints. To circumvent this drawback, hierarchical zeolites have emerged, attracting significant attention by materials and catalysis researchers from academic and industrial environments. These modified zeolites combine the intrinsic catalytic properties of conventional zeolites and the facilitated access and transport brought by an auxiliary mesopore network. Several types of hierarchical zeolites can be prepared by a multitude of methods, which can be broadly classified as templating routes (bottom up) and post-synthesis routes (top down). This has led to an assorted 'materials supermarket' awaiting applications. By now, a representative number of laboratory studies have shown improved activity, selectivity, and/or lifetime on hierarchical zeolites compared to conventional (purely microporous) zeolites. This applies to a wide range of catalyzed reactions, including isomerization, alkylation, acylation, aromatization, cracking, pyrolysis, methanol to olefins/gasoline, etc. Therefore, one is not taking much risk when stating that in the near horizon an increasing number of hierarchical zeolites will be implemented in industry for specific processes. USY (ultra-stable Y) was probably the first zeolite applied industrially (in fluid catalytic cracking) that combined micro- and mesopores. Steam and acid treatments of zeolite Y primarily stabilized the FAU structure by expelling part of lattice Al to extra-framework positions, leading to extra mesoporosity. However, the positive effect of the additional external surface area in terms of improved transport was not unequivocally evidenced. Until early 2000, the introduction of secondary (meso)porosity in zeolites to relieve diffusion limitations did not receive much attention by the scientific community. Research efforts on hierarchical zeolites were intensified once realized that ordered mesoporous materials, despite their excellent accessibility, cannot match key zeolites' properties such as super-acidity and thermal stability.

We considered it timely and of great inspiration to edit this volume, entitled 'Hierarchical Zeolites in Catalysis', containing original contributions from world-leading scientists in the field. The issue opens with a review covering catalysis by hierarchical zeolites and is followed by several excellent original papers devoted to synthesis, characterization, functionalization, diffusion, and catalytic application of hierarchically organized zeolites. The guest editors wish to thank all contributors and reviewers for their support in the preparation of this issue.

On top of the spectacular achievements hitherto, there are numerous remaining challenges and future opportunities for hierarchical zeolites in catalysis. Progress in the following directions can be anticipated within the next few years:

- *Precision design of hierarchical zeolites*: The benefits of hierarchical zeolite catalysts have been often illustrated by comparing them with their purely microporous counterparts. However, the optimal hierarchical zeolite for a given reaction has not been ascertained and hence the improvement margin of these modified zeolites still remains large. The establishment of quantitative synthesis–property–function relationships will enable the optimal design of hierarchical zeolites. Many aspects can contribute to rationalize the design of these materials, *e.g.*, a more detailed characterization of pore inter-connectivity, wider utilization of methods to investigate transport properties, identification and application of descriptors (like the proposed hierarchy factor and accessibility index), etc.
- *New applications*: Entirely new uses of zeolite catalysts can be envisaged, such as processes based on catalysts featuring zeolite-immobilized species that are larger than typical micropore openings (*e.g.*, homogeneous catalysts, enzymes, and crystalline metal particles) and processes that involve very bulky substrates, which are fully excluded from the pores of conventional zeolites. Outside the catalysis field, we also expect detailed studies assessing the properties of hierarchical zeolites as ion exchangers and absorbents *e.g.*, in applications requiring fast uptake or release of ions or molecules.
- *Industrial production*: A crucial next step for practical realization of hierarchical zeolites requires the successful extrapolation of the encouraging catalytic results obtained in laboratory studies on powdered catalyst samples to the industrial scenario, that is, multi-ton preparation of shaped bodies containing relevant binders and additives.
- *Economic analyses*: Clearly, introduction of hierarchical zeolites in industry requires that attractive cost-benefit analyses can be established. Thus, the additional costs involved in manufacturing hierarchical zeolites must obviously be counter-balanced by improved product yields or product properties. Therefore, all measures of catalytic activity and selectivity should be reported also in the industrially relevant units, which is often space-time yield rather than mass-based catalytic activity even though it varies from case to case.

Our firm impression after setting up this volume is that the future of hierarchical zeolites is bright and promising in terms of both fundamental and practical scenarios;

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these materials are true candidates to substitute conventional zeolites in a multitude of catalyzed reactions within oil refining, petrochemical, fine chemical, and/or environmental applications.

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