Chemical Engineering Journal 329 (2017) 56–65

Contents lists available at ScienceDirect

Chemical Engineering Journal

journal homepage: www.elsevier.com/locate/cej

Method for generating pore networks in porous particles of arbitrary shape, and its application to catalytic hydrogenation of benzene

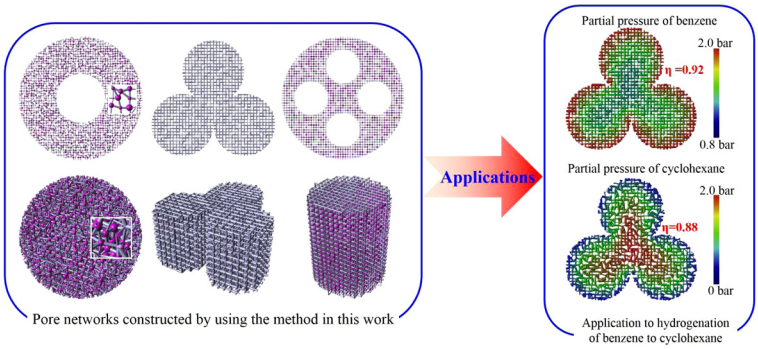
Guanghua Ye [a], Yuanyuan Sun [a], Xinggui Zhou [a,*], Kake Zhu [a], Jinghong Zhou [a], Marc-Olivier Coppens [b,*]

[a] State Key Laboratory of Chemical Engineering, East China University of Science and Technology, Shanghai 200237, China
[b] Department of Chemical Engineering, University College London, London WC1E 7JE, UK

HIGHLIGHTS

- An adaptable, efficient pore network cutting algorithm is proposed.
- Pore networks with 8 archetypical shapes are successfully built as examples.
- Pore networks are applied to simulate diffusion and reaction in porous catalysts.
- Shape and randomness of the pore network could affect catalytic performance.
- A larger effectiveness factor of catalysts is found for a regular pore network.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Available online 16 February 2017

Keywords:
Porous materials
Pore network
Particle shape
Cutting algorithm
Hydrogenation of benzene
Catalysis

ABSTRACT

A method is established to generate pore networks within domains of arbitrary shape, as long as the domain can be mathematically described by a set of inequalities. In this method, a stochastic network algorithm is adopted to construct pore network skeletons, which are then cut into the desired shapes using a new pore network cutting algorithm. The latter can be embedded into other methods to transplant its 'pore network cutting' function. Using this method, pore networks with four archetypical two-dimensional shapes (namely, cross-sections of one-holed rings, trilobes, four-holed rings, and wheels) and four three-dimensional shapes (namely, spheres, cylinders, trilobes, and hollow cylinders) are constructed as examples. Then, some of these pore networks are applied to simulate diffusion and reaction in Pd/γ-alumina catalyst particles for hydrogenation of benzene to cyclohexane. It is shown that the randomness of the pore network and the external particle shape significantly affect the performance of catalysts, because of their impact on effective diffusivity and diffusion length, respectively, indicating that this structural information must be accounted for to achieve a model with high accuracy. The versatile method proposed in this article is ideal to study the effect of particle shape and pore network structure on the performance of porous materials for catalysis and other applications.

1. Introduction

Porous materials are widely used in the chemical industry as catalysts, adsorbents, membranes, etc. Such materials contain a huge number of pores of different geometry and size, which are interconnected to form networks of different topology. In addition,

* Corresponding authors.
E-mail addresses: xgzhou@ecust.edu.cn (X. Zhou), m.coppens@ucl.ac.uk (M.-O. Coppens).

http://dx.doi.org/10.1016/j.cej.2017.02.036
1385-8947/

these materials come in various shapes, depending on their applications. External shape, pore morphology and pore network topology can all strongly affect the overall properties of porous materials [1–4]. Therefore, accurate pore network representations are essential for porous material characterization and design.

Many types of pore models have been conceived to describe porous materials; these have been reviewed by Sahimi et al. [5] and Keil [6]. Early models include the parallel pore model, tortuous pore model, cylindrical pore model, model of Wakao and Smith, and grain model. Although these early models could describe certain features of the pore space (pore size distribution, porosity, and, to some extent, tortuosity) and could be extended to account for morphological features like pore surface roughness, they do not explicitly account for the pore network topology or pore connectivity, or for the spatial distribution of the pores. Compared to these early models, pore network models are more representative of the pore space [7,8]. At present, pore networks are frequently used to represent the pore space of rocks [9,10], adsorbents [11], membranes [12–14], fuel cell electrodes [15–17], and porous catalysts [18–21].

Pore networks can be generated by using regular lattice-based, stochastic, and image-based methods [22–24]. Bethe lattices with a connectivity of 3 or more, square lattices with a connectivity of 4, and cubic lattices with a connectivity of 6 have been employed to build lattice-based pore networks. The connectivity can be altered by removing or adding bonds. The pore size is assigned according to some statistical distribution, such as the Gaussian distribution. Although some irregularities can be introduced in the lattice-based pore networks by varying the position of sites, these pore networks are not adequate to describe irregular pore network structures. Stochastic pore networks are better suited in this case. Normally, sites are randomly or uniformly distributed in a square domain or a cubic domain; then, adjacent sites are interconnected according to connectivity; shapes and sizes of sites and bonds are assigned in the final step. Finally, image-based pore networks are extracted from the three-dimensional (3-D) images that can be obtained by using statistical methods, process-based methods, and X-ray micro-tomographic characterization [25]. Although this image-based method yields pore networks closest to the real porous materials, this method is computationally intensive and

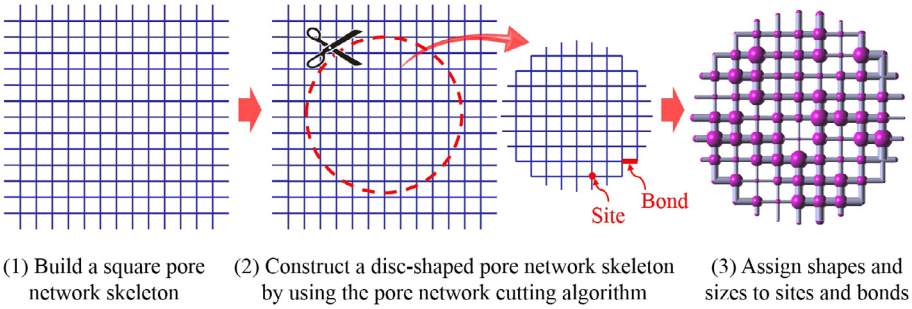


Fig. 1. Schematic illustrating how to generate a disc-shaped pore network. In this illustration, a regular network is given as an example, however, this method is actually applicable to any network, including irregular ones.

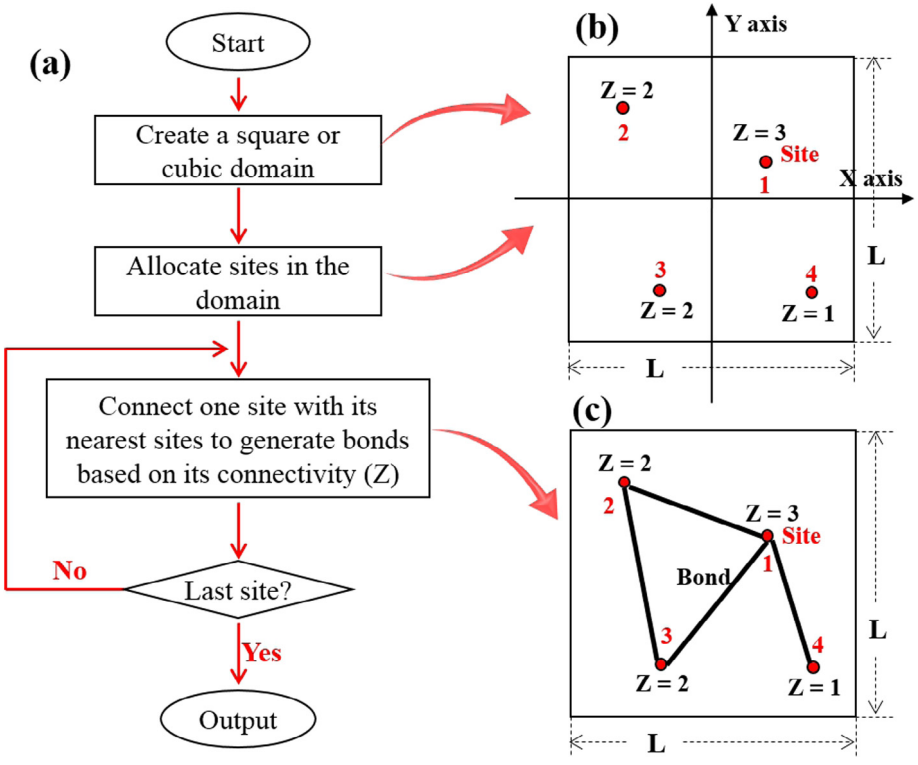


Fig. 2. (a) Flowchart for the stochastic network algorithm; (b)–(c) a 2-D illustration for the algorithm.