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Review paper

A review on sustainable synthesis of zeolite from kaolinite resources via hydrothermal process

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ABSTRACT

Synthesis of zeolite through hydrothermal process has been commonly used for decades. However, it does not satisfy the serious terms of sustainability that leads to reduction of costs, reduction of waste, eliminating negative environmental impacts and improvement of any system efficiency. The bottom line of this review paper is to highlight the current trends in the hydrothermal synthesis of zeolite, attention is paid to the utilization of natural resources and manufacturing wastes as raw materials to synthesize zeolite. Optimum conditions for sustainable hydrothermal synthesis of zeolites from kaolinite natural resources are also studied and discussed in this paper.

Contents

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1. Introduction

Extensive studies have been made on zeolite in terms of the structure as it relates to the properties and synthesis. Gougazeh and Buhl [1] emphasized the importance of synthesizing zeolite in the form that suits to the industrial application. In addition, Meng et al. [2] is particularly concern about green routes for synthesizing zeolites, specifically focusing on significant reduction of organic templates and possible elimination of solvent by employing templates recycling approach with the use of nontoxic templates. Historically, the first synthesis of zeolite was attempted by St. Claire-Deville in 1862 [3]. However, the pioneer work of Barrer on zeolite adsorption and synthesis in 1948 concluded that a wide range of zeolites could be synthesized from aluminosilicate gels [4,5], which has brought to the more exploration and new advancement of zeolite from various sources of alumina and silica.

Mostly, sodium aluminosilicate gel is employed to synthesize zeolites by several techniques ranging from the complex to the very simple techniques i.e. hydrothermal treatment [6–8]. Commercial synthetic zeolites is more often used than natural zeolites due to its higher purity [9] and more uniform particle sizes, and thus make it more suitable for most engineering applications and scientific purposes [9–11]. However, the preparation of synthetic zeolites from chemical source of silica and alumina is relatively expensive [11,12]. Numerous studies have been conducted in searching of affordable raw materials that is suitable for synthesis of zeolite [13,14]. Various ecological materials, agro waste and manufacturing wastes have been studied and used as a starting material in synthesizing zeolite, such as: rice husk ash [15–17], coal fly ash [16,18,19], oil shale ash [20–22], bagasse fly ash [23–25], high silicon fly ash [25,26], paper sludge [27,28], waste sandstone cake [29,30], waste porcelain [31,32] and kaolinite [12,33–36].

The similarity in the composition of most of these waste ash with zeolite has enticed more researchers to intensively study the basic and fundamental of chemical composition of the aluminosilicate waste structure to the zeolite structure. As for example Youssef et al. [37] has claimed that, the similarity of the bulk chemical composition of kaolinite clay makes it suitable for the use as a starting materials for zeolite synthesis. This hypothesis was further supported by the recent works of Jamil and Youssef [38] based on the early investigation by Kovo [39], Barrer [40] confirming the existence of high percentage of aluminosilicate in both composition of kaolin and zeolite [35,40–42].

Hence, researchers are smartly propagating the utilization of geological materials and manufacturing wastes for the synthesis of zeolite as a more cost efficient system. Therefore, this has led to the more advancement and exploration of zeolite synthesis using a simple and less expensive method by controlling processing parameters especially using a simple hydrothermal techniques.

Fortunately, some areas of the topic has been reviewed elsewhere, Cundy and Cox [43] conducted an evolutionary background study highlighting the array of discoveries and the resultant development of ideas in the field of zeolite synthesis from the 1940s to 2002. Their study concentrated on hydrothermal methods of synthesis of aluminosilicate zeolites but has not clearly described the most credible steps through which the amorphous aluminosilicate reagents are transformed to crystalline molecular sieves.

Zimmermann and Haranczyk [44] employed a data-driven approach in reviewing the history and utility of zeolite framework type. The scope of their work covered 229 crystallographic information files from the IZA website, on which they conducted a detailed investigation on the discovery year of such frameworks. Their motivation is to realize the practical usefulness of such materials. Although various discovery phases and the realization of exotic zeolite materials has been highlighted, their work is limited only to exploiting the discovery rate of new zeolite materials and the commercial development of such materials.

The most recent review by Johnson and Arshad [12] briefly discussed the evolution of synthetic zeolite up to the year 2012. The study emphasized the advantages of using cheap natural materials for kaolinite-based zeolite synthesis compared to conventional system. However, the review has not highlighted any effort towards green routes for synthesizing zeolite.

This review presents a brief introduction on zeolite, the hydrothermal synthesis technique, as well as the factors affecting the synthesis of zeolite through simple and low cost hydrothermal technique. Furthermore, kaolinite natural resources that can be originated from the clay was chosen as the main raw materials to synthesize zeolite since it is abundantly available in our country. At the end, the optimal conditions for the sustainable hydrothermal synthesis of zeolites from kaolinite natural resources were summarized.

The target of this review article is to afford researchers and students with a broad but relatively short overview of the current development in synthesis of zeolite from natural resources, and to complement the work of other researchers who reported a major surge in the synthesis of zeolite and characterization of its properties as well in the study of its possible industrial applications. The first section of the study begins with a brief introduction on zeolite and zeolite framework structures, then followed by the advancement in the study of zeolites. The second section discussed the hydrothermal synthesis process as a major synthesis route for many zeolite and zeotype materials. Specifically, synthesis of zeolite from kaolinite and the factors affecting the synthesis of zeolite via hydrothermal process is equally discussed. The third section discussed the various sustainable routes for the synthesis of zeolite, and finally concluded by summarizing the optimum condition for the synthesis of zeolite.

1.1. What is zeolite?

Zeolites are highly crystalline, porous aluminosilicate earth metal minerals based on a three dimensional network of tetrahedral $[SiO_4]^{4-}$ and $[Al_4]^{5-}$ whose frame work are open structures with cations positioned within the material's pores. The cation neutralizes the negative charge on the lattice [10,35]. Xu et al. [45] mentioned that the unique ion-exchange and the catalytic properties of zeolites and other similar materials is basically the result of this cation mobility.

A prosperous study on the relations amid the synthesis, structure and properties of zeolite has been documented. The atlas of zeolite structure type [46] is a publication of the International Zeolite Association (IZA). The IZA structure commission publish and ensured that the atlas of zeolite structure type is frequently updated. The term zeolite framework refers to a corner-sharing network of tetrahedrally coordinated atoms [46]. The IZA structure commission subsequently coded these frame work topologies regardless of its composition for easy management of the frame work database. Examples are: FAU [molecular sieves with a faujasite topology], LTA [Linde zeolite A], MFI [ZSM-5 and silicalite topologies], MOR [mordenite topology], and AFI [aluminophosphate AlPO4-5 topology], see examples in Fig. 1(a–b) [47,48].

Petrov and Michalev [10], Zimmermann and Haranczyk [44] describe the structure formula of zeolite based on the crystallographic unit cell as follows: $M_{x/n}$ $[(AlO_2)_x(SiO_2)_y]$ wH_2O, such that (M) refers to alkaline earth cation, while the valence of the cation is expressed as (n). The number of water molecules per unit cell is represented as (w), x and y connote to the total number of tetrahedra per unit cell.