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A Review of Research Progress on Sustainable Zeolite Synthesis Technology

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Abstract: With the surge in global demand for sustainable materials, zeolite has shown irreplaceable value in the fields of environmental remediation, green catalysis and new energy storage due to its unique pore structure and versatility. However, traditional zeolite synthesis relies on high purity silicon and aluminum raw materials and high energy consumption process, which is not only costly, but also aggravates resource consumption and carbon emissions. To this end, this study focuses on sustainable zeolite synthesis technologies and systematically reviews their innovative processes, industrial applications and contributions to carbon neutrality goals. The results show that the solid waste based molecular sieve has great application value. The transformation of inexpensive source materials and waste can provide lucrative zeolites with qualities akin to those of commercially synthesized zeolites. Currently, zeolite synthesis technology has evolved from the conventional hydrothermal method to include microwave-assisted hydrothermal synthesis, the molten salt method, and alkali fusion hydrothermal method, which markedly enhances efficiency and diminishes energy consumption. However, large-scale production still faces challenges such as high equipment costs, raw material volatility and by-product generation. The future needs to focus on green scale technology to promote the transformation of zeolite from laboratory innovation to industrial grade solutions.

Keywords: Sustainable waste management, zeolite, fly ash, green synthesis, adsorption

1. Introduction

As a multifunctional material with a regular micropore structure, a high specific surface area, and an adjustable acid site, zeolite has attracted extensive research interest in catalysis and separation technology in recent years [1]. Grand View Research reports that the global zeolite market is projected to expand at a compound annual growth rate (CAGR) of 6.2% annually [2]. Its core driving force is the urgent need for green technologies in the chemical, energy and environmental sectors. Although the existing research on zeolites has made remarkable progress, most of the existing research only focuses on the material properties, and the environmental impact on the whole life cycle has not formed a standardized paradigm. The global chemical industry confronts challenges of resource scarcity and environmental pollution, necessitating an effective green mass production strategy and technological innovation to facilitate the industrialization of zeolite catalysts and supply the requisite materials for associated chemical processes [3]. This study focuses on the technical progress and value of zeolite synthesis and application, combined with industry case analysis and technology comparative analysis, to explore its technical bottlenecks and future potential. The

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research results of this paper are conducive to promoting the industrialization of green synthesis technology, formulating industry standards and policies (proposing a zeolite performance index and LCA evaluation framework to provide a basis for global market standardization) and solving the interdisciplinary technology gap. Sustainable zeolite synthesis and application not only promotes cross-cutting innovation between materials science and environmental engineering but also provides technical reference for solving the dual challenges of resource efficiency and pollution control. Accelerate zeolite technology towards the goal of carbon neutrality and strengthen the coordinated development of regional economic benefits and ecological benefits.

2. Sustainable synthesis of zeolite

As the core field of materials science, zeolite synthesis has realized the leap from single micropore structure to multistage pore structure through the innovation of the hydrothermal method, the template method and the green process in the past 60 years [4, 5]. This evolution is not only reflected in the innovation of synthesis methods but also in the strategic transformation of the raw material system. From relying on high-purity silica, kaolin and other traditional minerals, to the efficient use of industrial solid waste such as red mud and fly ash. The diversified application of traditional raw materials (silica, clay minerals) and industrial waste (red sludge, fly ash) not only reduces the cost, but also promotes the resource cycle [6]. The following is a systematic analysis of the mainstream preparation methods of zeolite, and the most mature hydrothermal synthesis method is taken as an example to discuss the regulation mechanism of the key process parameters on the structure and properties of the product.

2.1. Hydrothermal synthesis

Hydrothermal synthesis as the core method of zeolite synthesis, on the basis of simulating the natural metallogenic conditions (high temperature and high pressure hydrothermal environment), through the green process innovation to achieve a double breakthrough of environmental friendliness and efficient use of resources. Aluminum salt slag, a hazardous waste can be used as a suitable Al source for the synthesis of alumina and zeolite. Incorporating additional raw materials allows for the conversion of each kilogram of waste into 2.5 kilograms of zeolite, eliminating residual waste production [7]. Hydrothermal synthesis reaction is carried out under high temperature (90-150°C; Or under-critical conditions (100-240°C)) and pressurized environments (1-15 bar) [5,6]. The reaction time is generally 24-96 hours, depending on the type of target zeolite (e.g., Na-X zeolite takes 18 hours to reach the crystallization peak [6]). By precisely regulating the activation solution (such as a low concentration NaOH system), Si-Al ratio (different Si-Al ratio will produce different types of zeolite), reaction temperature, reaction pressure and reaction time, the hydrothermal method can be used to synthesize zeolite products with specific pore structure and functional sites [8]. This efficient and ecologically compatible technology path provides a low carbon solution for zeolite production.

2.2. Various hydrothermal synthesis techniques

2.2.1. Microwave-enhanced hydrothermal method

Microwave assisted hydrothermal synthesis is an advanced technology that uses microwave radiation energy to accelerate chemical reactions and achieves rapid and uniform heating through direct interaction between electromagnetic waves and materials [9]. Using hydrothermal synthesis of zeolite, this process can take hours to days [10]. Microwave heating can rapidly increase the crystallization temperature, thus speeding up the zeolite synthesis reaction and significantly shortening the production time (for example, the synthesis time of TS-2 zeolite is reduced from 48

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hours to 15 hours; the synthesis rate of ZSM-5 molecular sieve increased by a factor of 4), and the homogeneity, phase purity and morphology control of the product were improved [5, 6]. Although microwave-assisted hydrothermal synthesis can significantly shorten the reaction time and improve energy efficiency, its non-uniform heating characteristics are prone to causing local temperature gradients, affecting the synchronization of nucleation and crystal growth [11]. At present, it mainly stays in the laboratory or pilot stage, and has not yet realized large-scale industrial applications.

2.2.2. Alkaline fusion-assisted hydrothermal process

The alkali fusion hydrothermal method is a high temperature and high-pressure synthesis technology based on a strong alkaline environment, which is suitable for zeolite conversion of silica-alumina raw materials (such as fly ash and kaolin). The core steps include: first, the raw material is mixed with NaOH/KOH and melted at high temperature (500-800°C) to destroy the stable silica-aluminum lattice in the raw material and generate soluble silicate and aluminate [12]; Subsequently, it is aged for several hours at 20-50°C and finally synthesized Na-X, P-type zeolites by hydrothermal crystallization (100°C, 24-48 hours) [6]. This method can efficiently utilize silicon and aluminum resources (fly ash and kaolin) in industrial solid waste, shorten synthesis time and improve product purity (crystallinity of more than 62%), but it requires high corrosion resistance of equipment, and alkaline effluent treatment requires additional treatment to reduce environmental load [13].

2.2.3. Molten salt method

The molten salt method uses a salt mixture ($NaOH\text{-}NaNO_3$ or $NaOH\text{-}KNO_3$rather than water as the reaction medium to achieve a directed synthesis of zeolite at relatively low temperatures (200-400°C) [13]. Its principle is to significantly reduce the reaction activation energy through the ion conduction characteristics of molten salt, accelerate the dissolution and mass transfer process of silica-aluminum precursors (fly ash and kaolin), and promote the formation and growth of zeolite crystal nuclei. In addition, compared with the alkaline melting hydrothermal method, the reaction temperature of this method can be reduced by more than 50%, and the environmental problems caused by the traditional alkali smelting process are solved.

2.3. Synthesizing zeolite from fly ash

Coal fly ash is a combustion product obtained from the combustion of coal, biomass or a mixture of two materials, and its main components include $SiO_2(36.40wt\%)$, $Al_2O_3(35.00wt\%)$, $CaO(3.95wt\%)$, $Fe_2O_3(2.11wt\%)$, $K_2O(0.34wt\%)$, $MgO(0.18wt\%)$, $SO_3(0.47wt\%)$, and others(18.30wt%) [7, 13, 14]. In the United States, the coal industry produces 1.143 billion tons of fly ash annually, and much of it is not effectively reused [15]. The most common process for the synthesis of zeolite from coal fly ash is to pulverize coal fly ash and mix it with NaOH, destroy its crystal structure through various chemical reactions, release the active components of silicon and aluminum, and then carry out a hydrothermal conversion process to finally obtain zeolite A, X, Y and other high value-added products [16]. The CO_2 adsorption capacity of coal-ash zeolite 13X is 225 mg/g, with a specific surface area of 643 m^2/g, demonstrating performance comparable to commercial goods [16].

Therefore, the use of coal fly ash as a raw material to synthesize zeolite not only reduces the treatment cost of coal fly ash, but also greatly reduces the adverse impact of landfill on the environment, thus obtaining economic and environmental advantages [7, 12]. However, the chemical composition of coal fly ash (such as Si-Al ratio, and impurity content) is significantly affected by coal sources, combustion conditions and other factors, resulting in unstable crystal form and properties of synthetic zeolite. For example, aluminum-rich ash tends to form zeolite A, while silica-rich fly ash

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tends to form zeolite X or zeolite Y [11]. In addition, after repeated use, zeolite performance may be reduced due to skeleton collapse or inactivation of the active site. At present, there is a lack of unified performance standards (such as purity and pore size distribution) for industrial grade fly ash zeolite, and the lack of collaborative research between material science and environmental engineering and chemical process restricts its transformation from laboratory to industrial end.

2.4. Discussion on synthesis technology and application of zeolite

The selection of materials and methods for zeolite synthesis significantly influences the structure, characteristics, and environment of the finished product. The traditional hydrothermal method is still the mainstream of zeolite synthesis, but its shortcomings such as long reaction time, high energy consumption, high raw material cost and high pollution have promoted the development of green alternative technologies. Zeolite synthesis is shifting from the traditional process of high energy consumption and high pollution to the sustainable path with resource recycling and green innovation as the core. Various hydrothermal synthesis technologies, such as microwave-assisted hydrothermal synthesis of alkali, melting, and salt melting, break through the bottleneck of raw materials and energy consumption, although facing challenges such as purity and equipment cost, but provide a sustainable zeolite synthesis path for the petrochemical and environmental protection industries. In the future, sustainable zeolite synthesis needs to take into account high efficiency, low consumption and ecological friendliness, and promote zeolite synthesis from "laboratory environmental protection" to "sustainable industry chain".

3. Application of zeolite

3.1. Application in the water treatment

The expansion of urban areas and industrial sectors has resulted in significant water contamination globally. Wastewater from industrial, agricultural, and home sources comprises many pollutants, including heavy metals and both organic and inorganic contaminants [12]. These contaminants can negatively impact the environment and human health. Over 80% of wastewater in developing nations is released untreated, leading to suspended particles and detrimental compounds that induce eutrophication or contamination of rivers, lakes, oceans, and soils [17]. Due to its porous structure and surface properties, zeolite may effectively absorb detrimental contaminants and heavy metals in industrial effluent. The excessive release of ammonia, nitrogen, and phosphate has been demonstrated to be a primary contributor to water eutrophication, disrupting the equilibrium of the ecological environment [18].

According to Kumar et al., mordenite and clinoptilolite have been shown to be effective adsorbents for ammonia removal [19]. Seyed et al. HZSM-5 nano-zeolite synthesized from fly ash showed good performance in the removal of aromatic pollutants [20]. In addition, in the dosage (8 g/100 ml) of nano-zeolite synthesized from fly ash (ZFA), the removal efficiency of nano-zeolite NaP1 synthesized with fly ash is 95% and 98% respectively for nitrogen and phosphorus [11]. Heavy metal pollution in wastewater is a major global challenge because of the far-reaching harm of heavy metal ions even at very low concentrations [12]. According to Moniem et al., the zeolite synthesized from blast furnace slag has a removal rate of more than 99% of Cu and Cd metals within 10 minutes, and reaches complete removal within 20 minutes [21].

3.2. Gas adsorption and catalysis

Due to the increasing levels of gaseous pollutants in the atmosphere, air pollution has become a serious problem affecting human health and the environment. Due to its unique microporous structure, high specific surface area and controllable surface chemical properties, zeolite shows significant advantages in the adsorption and catalytic removal of atmospheric pollutants (such as VOCs, CO, methane, etc.). According to Dabbawala et al., ZSM-5, zeolite-β, zeolite-A and zeolite-Y are widely used in gas separation, adsorption, ion exchange and catalysis due to their high efficiency and cost-effectiveness [22]. Even zeolite Y synthesized from fly ash can absorb twice as much SO_2 in gaseous pollutants as its commercial counterpart [12]. In addition, according to Wang et al., CHA type SSZ-13 molecular sieve synthesized with activated coal fly ash as primary zeolite provided a practical and energy-saving method for removing NO from diesel exhaust [23].

4. Discussion

The zeolite business has established a comprehensive industrial chain, particularly vibrant in deep processing, producing items such as molecular sieves, catalysts, and adsorbents, which are extensively utilized in environmental protection, the chemical sector, medicine, and other domains. Among them, global environmental protection policies have promoted the use of zeolite in flue gas desulfurization and sewage treatment. The chemical industry relies on zeolite as a catalyst carrier to improve production efficiency. In the pharmaceutical and food industries, zeolite is used for drug delivery and bone/tooth tissue engineering due to its unique mechanical, biological and antimicrobial properties. New energy demand is also strong, such as hydrogen energy catalysts, lithium-ion battery materials and other emerging directions that will promote zeolite demand growth. In the future, the zeolite industry will focus on the research and development of high-performance materials, such as nano-zeolite, composite zeolite, etc., to improve adsorption efficiency and catalytic activity. The optimization of synthesis technologies (such as hydrothermal and solvothermal methods) will reduce production costs and promote the development of high-end products. In addition, green production processes, such as low-energy purification technologies, will become mainstream and meet the requirements of the "dual carbon" target.

5. Conclusion

Zeolite as a strategic material with both environmental remediation and green catalytic functions, the innovation of its synthesis technology and the expansion of its application boundary provide a key material basis for the low-carbon transformation of the chemical industry. This paper reviews the latest progress of sustainable synthesis technology of zeolite and its core contributions in water treatment, gas adsorption and catalysis, and reveals its potential and challenges from laboratory innovation to industrial application. At present, most of the research on solid waste-based zeolite is only carried out on a laboratory scale, and there is a lack of systematic evaluation of energy consumption and impurity interference in large-scale production. It is suggested to further study the long-term properties of zeolite in a complex environment. There is room for improvement in the scope of literature coverage in this study, and more interdisciplinary literature (such as energy storage and carbon capture) should be included in the future to enhance the foresight of the review.

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