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УДК 666.3

工业生态与节能——粉煤灰基 CO_2 吸附沸石绿色制备

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Summary. *CCUS carbon capture, utilization and storage is one of the key technologies to deal with global climate change, which has been highly valued by countries all over the world and has increased research and development efforts, but there are still difficulties in industrialization. This paper introduces a green preparation process of zeolite for carbon dioxide adsorption.*

Human beings release too much greenhouse gases such as CO_2 in production and life, and the anthropogenic increase in atmospheric greenhouse gas concentrations is the main factor of current and future climate change, and if measures are not taken, CO_2 concentrations will continue to increase for many years in the future [1]. As a major CO_2 emitter, China is facing arduous CO_2 emission reduction tasks, and carbon capture and utilization technology is one of the important technologies to solve this problem. At present, CO_2 capture technology has been industrially applied, and the preparation of high-efficiency CO_2 adsorbents is the most critical part. Based on the above problems, the research on finding better technologies to prepare high catchability for capturing and storing CO_2 and optimizing them to achieve the ideal level has been gradually emphasized. At present, there are many mainstream porous adsorption materials in the world,Such as type A zeolite, type X zeolite, ZSM-5, MOFs, etc. [2]. Peter G. Boyd et al. analyzed some porous materials with optimal adsorption for CO_2 adsorption separation through comparative studies.Patrick Nugent et al. [3] synthesized metal-organic framework materials with strong carbon capture performance. However, in the existing zeolite synthesis process, it is impossible to avoid the generation of waste liquid, and the treatment process of waste liquid will cause secondary pollution to the environment.

In addition, coal, as one of the most stable and effective energy sources in our country, will produce a large amount of fly ash in the process of its combustion. To 2000, China's coal ash emissions about 160 million tons. The treatment and utilization of fly ash has become a major environmental protection problem in today's world.Many teams have explored the influence of different synthesis methods on the types and properties of zeolite prepared from fly ash for the problem of how to rationally recycle and apply fly ash, a solid waste with huge emissions [4]. Wu Xuecheng's team [5] of Zhejiang University processed fly ash by acid leaching and alkali melting, synthesized various types of zeolite, and studied its adsorption performance, verifying the feasibility of zeolite synthesis from fly ash.

According to current theoretical analysis and previous experiments, four reaction parameters can be determined: alkali-ash mass ratio (referred to as alkali-ash ratio for short), crystallization time, liquid-solid mass ratio (referred to as liquid-solid ratio for short) and crystallization temperature.Among them, the ratio of alkali to ash affects the concentration of alkali in raw materials, and the addition of NaOH can dissolve the amorphous phase in raw materials, destroy mullite and Shi Ying phases, decompose them, and promote the activation of SiO_2 and Al_2O_3. Liquid-solid ratio plays an important role in the synthesis of zeolite. Too large or too small liquid-solid ratio directly affects the alkalinity of solution and the crystallinity of zeolite products. Li Bo [6] explored the role of liquid-solid ratio in the synthesis of zeolite A. The results showed that when the liquid-solid ratio was 5:1, the mass fraction of zeolite A was 59 %, which was the highest compared with that of zeolite A synthesized under other liquid-solid ratios. Crystallization time affects the nucleation and crystal nucleus growth process of zeolite.At the same time, it will affect the crystal particle size of synthetic zeolite. Generally speaking, the higher the crystallization temperature, the larger the particle size of zeolite. The crystallization time will be prolonged if the crystallization temperature is too low; High crystallization temperature will produce hydroxysodalite, resulting in low purity of zeolite. Therefore, too high or too low crystallization temperature is not conducive to the synthesis of zeolite A. Lian Xianjin et al. [7] considered the time required for crystallization and the quality of zeolite, and considered that the ideal crystallization temperature was 80~100 ℃.

By studying the influence of crystallization time, solid-liquid ratio, silicon aluminum ratio and other conditions on the synthesis of zeolite, as well as the recycling of waste liquid, we analyze how to prepare zeolite with the best performance. The characteristic peaks, crystallinity and cell parameters were analyzed and determined by X-ray diffractometer (D8 ADVANCE, Brooke, Germany); Use a physical adsorption instrument (ASAP 2460, McMurray Teck Instrument Co., Ltd.) to test the nitrogen adsorption desorption isotherm of the sample at liquid nitrogen temperature; Fourier infrared spectrum analyzer (Cary-660-FTIR, Agilent Technology Co., Ltd.) was used to analyze the functional group structure of the adsorbent molecule, and the characterization and performance test of the sample were completed.

Based on the previous exploration and the above experimental exploration, it is proved that the preparation of fly ash based zeolite A by alkali melting hydrothermal synthesis has extremely high carbon dioxide adsorption performance, and there is still a large amount of Al_2O_3 and SiO_2 in the waste liquid that do not participate in the reaction, so the reaction can continue.Considering the reuse of pollutants and no secondary pollution of waste liquid, a green preparation scheme of zeolite A was designed and optimized.The use of cheap raw materials and template free synthesis methods will contribute to the large-scale application of green synthesis processes in the future.

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