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Nanoparticles used for CO2 capture by adsorption: A review

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Rapid industrialization, technological advancement, and innovation have led to a significant rise in carbon emissions globally, resulting in the growing problem of climate change. With the advancement of nanotechnology, the adsorption is becoming an effective strategy to directly capture CO2 with nanomaterials. This mini-review deals with the investigation for physical adsorption, amine-modified nanomaterials for chemisorption, and moisture-swing nanomaterials for chemisorption. The purpose is to highlight the current technologies available for a simple, environmentally safe, non-toxic, low-cost CO2 capture. In detail, this study examines several CO2 capture nanomaterials with an emphasis on economical and environmentally safe low to high temperature solid adsorbents.

* 1. Introduction

The increase in atmospheric CO2 levels is mainly due to the burning of fossil fuels such as gasoline and diesel. Reducing CO2 emissions is challenging due to the country's economic growth and accompanying development such as industrialization, urbanization, and transportation. Global fuel combustion released approximately 32.8 billion tons of CO2 (Grim et al., 2020). Additionally, the increased use of personal vehicles, driven by overpopulation, is also contributing to the rise in CO2 levels.

At the 2015 UN Climate Change Conference, participants reached an agreement to create a global framework to prevent severe climate change by limiting global warming below 2 ◦C and striving to keep it below 1.5 ◦C (Streck et al., 2016). According to the IPCC (Intergovernmental Panel on Climate Change), reducing CO2 emissions is essential if we are to keep the concentration of carbon dioxide in the atmosphere below 450 ppm by the end of 2100 and prevent climate change from becoming irreversible.

A variety of methods have been developed for removing CO2 from gas streams, including adsorption, absorption, cryogenics, and membranes (Mancini et al., 2022). Membrane separation stands out for its ability to produce high-purity CO2 streams, especially for power plants. It has several advantages, such as energy efficiency and continuous operation, compared to other methods like adsorption and absorption (Momeni et al., 2019). Cryogenic separation of CO2 involves liquid CO2 flow under high pressure, but it is expensive due to the need for refrigeration and water removal. This method is mainly suitable for highly concentrated CO2 streams. Adsorption is a relatively new approach to CO2 removal. To date, adsorption, together with absorption, is the most economical way to separate CO2 from large flows (Zeini Isfahani et al., 1994). This study focuses on enhancing the effectiveness of adsorption technology. In detail, the effect of nanoparticles on adsorption has been studied.

* 1. Nano particles effect for CO2 Capture

CO2 capture can be achieved through physical and chemical adsorption at room temperature and atmospheric pressure, and the captured CO2 can be easily released through gas flow and heating methods. CO2 can also be captured and released through moisture-swing adsorption by controlling moisture levels.

**2.1. Nanomaterials for physical adsorption**

The effectiveness of physical adsorption using nanomaterials depends largely on the exposed surface area, porosity, diffusion path, and availability of adsorption sites on the material, which result from physical interactions between nanomaterials and CO2 molecules (Oschatz and Antonietti, 2018). After extensive scientific research, various nanomaterials with strong CO2 adsorption capabilities have been developed, including activated carbon, zeolite, metal-organic frameworks, boron nitride, and MXene (class of two-dimensional inorganic compounds). For example, activated carbon is an inexpensive and readily available material that has a high surface area, large porosity, and good thermal and chemical stability, making it ideal for physical adsorption of CO2 under normal conditions (Karimi et al., 2018). The porosity and surface area of activated carbon can be easily adjusted through pyrolysis and etching methods to meet the structural requirements of the adsorption process (Sreńscek-Nazzal and Kiełbasa, 2019). The capacity and selectivity of activated carbon materials in terms of adsorption efficiency are limited by the weak physical interactions between CO2 molecules and the material (Coromina et al., 2015). To enhance the efficiency of CO2 physical adsorption, zeolite materials are utilized due to the stronger physical interactions between CO2 molecules and the negative charges on the surface of zeolite. In addition to the high surface area and abundant porosity in the zeolite structure, the intensity and distribution of surface charge can be controlled through chemical methods, which is crucial to the CO2 adsorption efficiency. The aluminum and silicon ratios in the material can be adjusted through different synthesis methods, and this ratio can greatly impact porosity and chemical activity. Optimizing this ratio can enhance the material's ability to capture CO2. Zeolite material's adsorption properties can be regenerated through heating (Santori et al., 2018). A major drawback of using zeolites is their strong water sorption capacity, as they undergo a similar absorption and desorption cycle as CO2. This makes it difficult to use zeolites for DAC (Direct Air Capture) in humid environments.

MOFs (Metal Organic Frameworks) have gained significant attention and have been widely used in various fields due to their versatility. MOFs are inorganic-organic hybrid crystalline materials formed through the self-assembly of inorganic metal cations or clusters and organic ligands. With their diverse composition, customizable pore size, high surface area, and tunable internal structures and properties, MOFs have become a promising alternative to traditional liquid and solid carbon capture sorbents. ZIF-8, a well-known type of MOFs, is a suitable candidate for CO2 adsorption due to its high nitrogen content, large surface area, and porous structure (Liu et al., 2014). ZIF-8 is famous for its chemical and thermal stability and can accumulate CO2 either in its molecular cage or in interspatial areas. The CO2 sorption capacity of ZIF-8 nanoparticles (ca. 30 nm in size) material was reported to be around 29 mg/g at atmospheric pressure and room temperature, with high-content nitrogen atoms providing basic sites for CO2 adsorption with higher capacity and selectivity.

Conjugated microporous polymers (CMPs) with optimized pore size distribution and high surface area are synthesized (Liu et al., 2014), resulting in a CO2 sorption capacity of 76.5 mg/g, 2.6 times higher than that of pure ZIF-8 materials. The CO2 adsorption enhancement is attributed to the pore enrichment effect of the nanomaterials.

A bioinspired MOFs (Bien et al., 2018) are developed with nucleophilic Zn-OH groups for trace CO2 adsorption at mild temperatures. A Zn benzotriazolate metal–organic framework has been subjected to a mild CH3CO2–/HCO3 ligand exchange procedure followed by thermal activation to generate nucleophilic Zn–OH groups that resemble the active site of α-carbonic anhydrase. The post synthetically modified MOF exhibits excellent performance for trace CO2 capture and can be regenerated at mild temperatures. IR spectroscopic data and density functional theory (DFT) calculations reveal that intercluster hydrogen bonding interactions augment a Zn–OH/Zn–O2COH fixation mechanism (Cai et al., 2020).

Sun et al. (2013) report a study of the adsorption of CO2 on boron nitride (BN) nanosheets and nanotubes (NTs) with different charge states. The results show that the process of CO2 capture/release can be simply controlled by switching on/off the charges carried by BN nanomaterials. CO2 molecules form weak interactions with uncharged BN nanomaterials and are weakly adsorbed (Sun et al., 2013b). The study shows that introducing excess electrons to BN nanomaterials results in strong adsorption of CO2 molecules, making these nanomaterials highly effective absorbents. The negatively charged BN nanosorbents selectively capture CO2 from its mixtures with CH4 and/or H2. Removing the electrons causes the CO2 molecules to spontaneously desorb. BN nanomaterials are therefore excellent for controlling, highly selective, and reversible CO2 capture and release.

Owuor et al. (2017) developed a 3D BN composite foam chemically connected with poly(vinyl alcohol) materials, which showed high CO2 uptake due to its improved porosity, surface area, and mechanical properties. Creating interconnections between inert nanosheets can remarkably enhance their mechanical properties . However, controlled design of such interconnections remains a fundamental problem for many applications of hexagonal-BN foams.

Li et al. (2017) reports that MXene is a promising candidate for CO2 physical adsorption. Two-dimensional (2D) transition-metal (groups IV, V, VI) carbides (MXenes) with formulas M3C2 have been investigated as CO2 conversion catalysts with well-resolved density functional theory calculations. While MXenes from the group IV to VI series have demonstrated an active behavior for the capture of CO2, the Cr3C2 and Mo3C2 MXenes exhibit the most promising CO2 to CH4 selective conversion capabilities. Results predicted the formation of OCHO• and HOCO• radical species in the early hydrogenation steps through spontaneous reactions. This provides atomic level insights into the computer-aided screening for high-performance catalysts and the understanding of electrochemical mechanisms for CO2 reduction to energy-rich hydrocarbon fuels, which is of fundamental significance to elucidate the elementary steps for CO2 fixation.

**2.2. Amine-modified nanomaterials for physical adsorption and chemisorption**

Compared to physical adsorption, chemical adsorption using nanomaterials has a higher sorption capacity and stronger interaction with CO2 molecules. Amine-modified nanomaterials have proven to be an effective solution for CO2 chemical adsorption, due to their low energy consumption, chemical stability, and reversible nature . When in contact with CO2 molecules, amine groups on the nanomaterials react to form carbamate or carbamic acid under dry conditions, or bicarbonate under moist conditions. Nanomaterials with porous structures, large surface areas, and abundant gas diffusion channels, such as nanosilica, carbon nanotubes, nano cellulose, MOFs, and porous alumina, are ideal support materials for the amine groups (Deng and Park, 2019).

For example, Xu et al. (2002) impregnated mesoporous silica with polyethylenimines containing amine groups, resulting in a nanoporous solid adsorbent which can serve as a “molecular basket” for CO2 in the condensed form with twice the CO2 adsorption capacity of pristine polyethylenimines. The novel “molecular basket” material can effectively adsorb CO2 at very low CO2 concentration, e.g., 0.5%, and it is stable in cyclical operations at relatively high temperatures.

Belmabkhout et al. (2010) chemically grafted amine-bearing mesoporous silica, which had a high CO2 uptake, fast adsorption rate, and stability at low pressure. The increased CO2 adsorption effectiveness is largely due to the increased accessibility of amine groups made possible by the porous and hierarchical structures of the supporting materials. In addition to unprecedentedly high CO2 uptake at low pressure, high adsorption rate and stability, this material exhibited very high selectivity for CO2 over N2 and O2, regardless of the presence of moisture. It is thus highly suitable for use in CO2 removal from air, for example in air purification and in closed-circuit breathing systems (Belmabkhout et al., 2010b). Nanomaterials produced through physical loading are cost-effective and easy to prepare, while those produced through chemical binding have a higher CO2 adsorption capacity and better structural stability. Primary amines react with CO2 to form ammonium carbamate, which is strongly adsorbed on the amine sites and forms networks of hydrogen bonds. This can lead to diffusion barriers during CO2 release.

On the other hand, secondary amines react with CO2 to form weakly adsorbed carbamic acid species, which can be released by gas flow at 35°C (Zhai and Chuang, 2017). High CO2 concentration is required to overcome the diffusion barriers of primary amines. Secondary amines are more expensive to produce, but they are more suitable for capturing low-concentration CO2. On the other hand, primary amines are better suited for high-concentration CO2 environments.

Hoshino et al. (2012) found that CO2 can be adsorbed and desorbed through temperature-responsive phase transition of micro and nanogel particles containing amine groups. This differs from simply relying on the porous structure of nanomaterials for CO2 adsorption. Amines in shrunken materials form ion pairs with absorbed bicarbonate ions at temperatures below the phase-transition temperature. Above the phase-transition temperature, the amines in shrunken materials release CO2. This highlights the potential of temperature-responsive nanomaterials for efficient and low-energy CO2 absorption. Metal-organic frameworks (MOFs) are a promising option for amine modification due to their well-defined adsorptive sites, adjustable pore sizes, and functionalities.

Z. Shi et al. (2020) created MOFs that were both thermally and chemically robust by appending amine groups to triazolate linkers. They developed a strategy based on appending amino groups to the triazolate linkers of MOFs to achieve exceptional chemical stability against aqueous, acidic, and basic conditions. These MOFs exhibit not only CO2/N2 thermodynamic adsorption selectivity as high as 120 but also CO2/H2O kinetic adsorption selectivity up to 70%, featuring distinct adsorptive sites at the channel center for CO2 and at the corner for H2O, respectively. This material had high adsorption selectivity, with distinct adsorptive sites at channel centers for CO2 and corners for other gases.

Ionic liquids (ILs) have garnered attention for CO2 adsorption due to their structure tunability, CO2 affinity, and low volatility. Nanomaterials with high levels of amine-modified ionic liquids have potential for CO2 chemisorption. For example, Yu et al. (2014) created a "ship-in-a-bottle" system of amine-functionalized ionic liquids in zeolite materials for CO2 chemical adsorption. The genuine host-guest systems were thoroughly characterized and tested in CO2 capture from simulated flue gas. It was evidenced the encapsulated ILs are more stable than the bulk ILs. This host-guest system showed high sorption capacity and rapid reversibility, even after multiple cycles under harsh conditions. The chemisorbed CO2 can be desorbed rapidly by flushing with N2 gas at 50 °C. The excellent physicochemical properties and the CO2 capture performance of the host-guest systems offer them great promise for the future practice in the industrial CO2 capture.

García-Gutiérrez et al. (2019) developed cellulose-supported solid ionic liquids (SoILs) for CO2 adsorption with increased sustainability. They have been demonstrated recently to be effective and low-cost sorbents for CO2 capture. The results of this study can be used to target the hotspots and improve the environmental performance of cellulose-supported SoILs through sustainable design. Hierarchical amine-modified solid ionic liquid-based nanomaterials could be a promising research direction for CO2 chemical adsorption in the future.

**2.3. Moisture-swing nanomaterials for chemisorption**

A novel moisture-swing technique has been put forward to further reduce energy consumption in CO2 adsorption, with the ability to capture CO2 under dry conditions and release CO2 under moisture conditions. The adsorption and desorption processes are directly controlled by humidity variations in the ambient environment and are not dependent on outside high energy consumption (X. Shi et al., 2020). Moisture-swing offers cost-effective CO2 capture using water without high energy consumption and simplifies the design of CO2 adsorption systems by eliminating the need for heating and cooling units. This flexible technique can also be combined with other applications and can continuously provide suitable CO2 concentrations for plants in greenhouse facilities. However, the water used in moisture-swing should be clean to avoid substrate contamination, and the technique may be affected by weather. The released CO2 concentration is comparatively low and needs to be enriched for further applications. The mechanism of the moisture-swing technique is based on the reversible conversion of carbonate and bicarbonate ions under dry and wet conditions (Shi et al., 2016).

For instance, T. Wang et al. (2011) presented a resin that utilizes moisture-swing technology for capturing CO2 from air in regular conditions. An amine-based anion exchange resin was prepared in an alkaline form and dispersed on a flat sheet of polypropylene to capture CO2 from air. The resin, featuring quaternary ammonium cations attached to the polymer and hydroxide or carbonate groups as mobile counterions, absorbs CO2 when dry and releases it when wet. In the presence of ambient air, the moist resin naturally dries and then absorbs CO2.

Improving the CO2 adsorption ability of moisture-swing materials by enhancing their porosity and surface area is a promising approach. For example, Song et al. (2019) introduced a simple grounding/sieving method to improve the porosity and surface area of an ion-exchange resin for the moisture-swing technique. The resulting hierarchical porous structures greatly improved the accessible surface area and facilitated the diffusion of CO2 and H2O molecules. The CO2 sorption capacity of the porous resin was much higher than that of the dense and smooth counterpart. Song et al. (2018) presented quaternized chitosan/polyvinyl alcohol aerogels with hierarchical nanostructures and large surface area for moisture-swing CO2 adsorption. The porous structures in the aerogels provided ordered nano-channels for highly efficient diffusion of CO2 and H2O molecules, and the moisture-swing aerogels delivered 38% higher CO2 capacity than commercially available polymer resins. The modified pseudo-first-order kinetic model they proposed fit the experimental results well and provides a theoretical method for designing moisture-swing materials. In conclusion, the moisture-swing technique is still in its early stages and has many challenges and opportunities that need to be addressed in future work. The construction of nanoscale ordered channels in moisture-swing materials is crucial for improving material exchange efficiency during CO2 adsorption/desorption processes. Advanced in-situ characterization techniques can be used to gain a deeper understanding of moisture-swing processes, which is crucial for designing high-performance CO2 sorbents.

* 1. Conclusion

The escalating issue of global climate change caused by excessive carbon dioxide emissions is one of the most pressing challenges facing modern society. This review outlines recent advancements in the use of nanomaterials for the adsorption of CO2. Various technologies and nanomaterials for CO2 adsorption are discussed, including physical sorbents, chemical sorbents, and moisture-swing sorbents. Physical sorbents are cost-effective and chemically stable but have low adsorption capacity and poor selectivity with other gases. Chemical sorbents based on amine-modified nanomaterials have higher CO2 sorption capacity and better selectivity, but their chemical stability depends on the adhesion strength of the amino groups to the substrate. Moisture-swing sorbents allow for CO2 adsorption and desorption through ambient moisture control, however, the materials currently available are limited to a few polymer resins and more functional nanomaterials need to be developed in the future.

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