

Special Issue on Carbon Capture and Utilization

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1. Introduction

Carbon dioxide (CO₂) emissions to the atmosphere have drastically increased in recent decades, with the energy and transport sectors representing major fractions of total greenhouse gas (GHG) emissions. This increase, which can be translated to a 50% increase in atmospheric CO₂ concentration since pre-industrial levels, has been associated with several negative environmental impacts, such as the greenhouse effect increase, global warming, and ocean acidification. Therefore, it has become urgent for world economies to reduce their CO₂ emissions, reduce carbon intensity associated with the energetic and transport sectors, and adopt effective CO₂-capture techniques.

Carbon capture and utilization (CCU) combines CO₂ capture and recycling. There are several sustainable CCU options: (i) biological CO₂ capture with biomass valorization; (ii) use of CO₂ as geothermal working fluid; and (iii) electrochemical reduction of CO₂. This Special Issue, entitled Carbon Capture and Utilization, aims to present an overview of currently applied techniques for CO₂ capture, transport, and applications, focusing on their advantages and disadvantages and the main challenges which are faced in their large-scale application. Additionally, new methodologies for CO₂ emissions inventory and environmental impact assessment are presented.

2. Contributions to This Special Issue

Methane (CH₄) and carbon dioxide (CO₂) often coexist in nature, forming several gas mixtures (e.g., natural gas, syngas, and landfill gas). The separation of CO₂ is essential in enhancing the purity and energetic value of these gas streams. In this context, Awadallah-F and Al-Muhtaseb [1] evaluated the adsorption of CO₂ and CH₄ with a mixture of activated-carbon (AC) nanoparticles with multiwall carbon nanotubes (MWCNTs) with several ratios. It was expected that the combination of different features of the selected materials could improve the gas-adsorption and -separation processes. The single adsorption capacity (adsorption/desorption isotherms of pure CO₂ and CH₄) and binary adsorption equilibria and selectivity were assessed. The adsorption capacities for both components were measured at different temperatures, and the dual-site Langmuir (DSL) model was applied to describe experimental data. The adsorption capacity of MWCNTs was found to be significantly lower than that of AC. Future research should be performed on different forms of carbon-carbon nanostructures. The study of interactions between them will enable researchers to track the associated mechanisms and enhance the potential gas-adsorption applications.

CO₂ capture can be performed following three technological concepts: post-combustion capture systems, pre-combustion capture systems, and oxy-fuel capture systems. For the latter, Hou et al. [2] investigated the effect of flow rates of oxygen and recirculated flue gas (which modifies the combustion atmosphere) on the heating performance and pollutant emissions (oxy-combustion characteristics) of Australian coal in a 0.3 MW furnace. For low oxygen flow rate, the flue gas contained residual oxygen and NO concentrations. On the



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other hand, the increased recirculated flue gas flow rate decreases the temperature of the radiative section and increases this parameter in the convective one. A high CO₂ concentration (up to 94.8%) can be achieved with a recirculated flue gas in a range of 3–5% (*v/v*), reducing the energy requirements (and CO₂ emissions) by carbon capture and storage. Compared with air combustion, a reduction of up to 74% of NO emissions was achieved. Besides these advantages, the investment in oxygen plant and flue gas recirculation system (CAPEX) and the energy consumption of these systems (OPEX) limits the application of oxy-fuel combustion to the conventional coal power plant. For post-combustion capture systems, Cruz-Navarro et al. [3] assessed the CO₂ capture and conversion of low volumetric flow rates into carbonates using Sr and Ba alkaline solutions with a simple system to implement in small industries. The study of the reaction showed that a 20-min reaction time is required to maximize the carbonate production. CO₂ conversion of 40% was achieved for the best reaction conditions with Sr(OH)₂. The formed solids were characterized by XRD, SEM/EDS, FTIR, and TGA. The synthesized carbonates have commercial value in several sectors, being an alternative and sustainable solution for recycling CO₂ and reducing its emission to the atmosphere.

CO₂ capture from large emission sources cannot be enough to fulfil the established climate targets. About 50% of CO₂ emissions are from diffuse sources (e.g., automobiles, shipping, and aircraft), and mitigation actions have been delayed due to economic interests (high fossil fuel dependence among world power economies). Therefore, considering the long residence time of CO₂ in the atmosphere, atmospheric CO₂ capture may be required. Yu and Brillman [4] evaluated the performance of a new radial-flow reactor for CO₂ adsorption from ambient air. The adsorption is achieved with 2 kg of supported amine sorbent, and the reactor can operate in two modes (batch or semi-continuous). Experiments showed that this unit is characterized by a low-pressure drop and contacting energy (0.7–1.5 GJ per ton of CO₂), uniform flow distribution, and short adsorption time (24–43 min); this is a viable solution for scaled-up applications.

The Intergovernmental Panel on Climate Change (IPCC) published guidelines for national greenhouse gas inventories in 2006 which provide a procedure for calculating emissions. However, it does not consider the net CO₂ emissions to the atmosphere (released in clinker fabrication minus those due to concrete carbonation) by the Portland cement clinker. Sanjuán et al. [5] applied a quick and easy method to quantify net CO₂ emissions of the cement industry. This method considers CO₂ uptake with concrete and mortar carbonation (cement-based materials), and it was applied to calculate the Portuguese net emissions of cement produced from 2005 to 2015. The results showed that 8.7 million tons of CO₂ were taken up in carbonation processes in the analyzed period; 37.8 million tons were released due to the calcination process. Short carbonation periods delivered a low CO₂ uptake. This methodology may improve the developed IPCC climate models. Sanjuán et al. [6] applied a similar method to the Spanish cement industry, evaluating the CO₂ emissions which were avoided by the carbonation process.

After CO₂ capture, the gaseous stream can be transported by pipeline or ship. The last option may be advantageous if the distance between the capture unit and the destination is large. In this case, the energy consumption for the stream liquefaction may be significant, being important to define an energy-efficient process. Jackson and Brodal [7] modelled several CO₂ liquefaction processes for a range of ambient temperatures, thus optimizing process operating parameters. Different scenarios were defined and modelled. The lowest energy consumption was achieved for the cooling temperature of 20 °C. A temperature change to 50 °C increased energy consumption by around 40%.

CO₂ storage at high concentrations is not presently acceptable due to the risk of leakage and its associated impacts. Thus, the development of materials for CO₂ storage is important. Satar et al. [8] synthesized and characterized (structure and physicochemical properties) three novel heteroatom-doped porous organic polymers (POPs) containing phosphate units. They have the following characteristics: (i) high surface area ($S_{\text{BET}} = 82.7\text{--}213.5 \text{ m}^2/\text{g}$); (ii) small pore-size distribution in terms of pore volume ($0.11\text{--}0.32 \text{ cm}^3/\text{g}$); (iii) small pore

diameter (1.96–2.43 nm); and (iv) high CO₂-storage capacity, in particular for high pressures (1.42 mmol/g (6.00 wt%) at 323 K and 40 bar). This high performance was mainly attributed to their geometry, since it provides a highly distorted and extended surface area network compared with other POPs.

To evaluate the sustainability of a process, the life cycle assessment (LCA) method is commonly used. However, integrated sustainable analysis is not possible within this method, as LCA does not quantify the biophysical capacity of the ecosystems to supply ecosystem services. Mora et al. [9] propose a methodology for connecting a process– or system–product flow demand from the technosphere and the feasibility of the ecosystem being able to supply ecosystem services based on sink capacity. The case study was a combined heat and power (CHP) plant with and without post-combustion carbon-capture technology in Mexico. Three scenarios were tested, and the CHP plant requires between 323.4 and 516 ha to supply the required oil as stock flow and 46–134 ha to supply the required freshwater. Concerning CO₂ sink, 52–5,096,511 ha is necessary to sequester the total emissions.

3. Conclusions

The capture of CO₂ from flue gases or directly from the atmosphere is a relevant process to decrease its emissions in the short term, while world economies remain highly dependent on fossil fuels. In this Special Issue, different CO₂-capture processes were tested with different gaseous streams: CO₂ and CH₄ mixture, flue gases, and atmospheric CO₂. In the adsorption processes, the applied materials were characterized and isotherms were determined. In terms of process, the operating parameters were optimized to increase the feasibility of CO₂ capture. CO₂ transport and storage were also covered, with studies aiming to surpass some difficulties of implementing CCU. New methodologies were presented to quantify net CO₂ emissions of an important industrial sector (the cement industry) and to perform an integrated sustainable analysis, using a combined heat and power plant as a case study.

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