


CO₂ Capture and Utilization Editorial

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Climate change is likely the most relevant challenge that humanity will face this century. An IPCC special report entitled “Global Warming of 1.5 °C” detailed the impacts of global warming of 1.5 °C above preindustrial levels. It also highlighted the relevance of limiting the global warming to this value in order to avoid the most severe effects of climate change, achieving net-zero CO₂ emissions by 2050.¹ This is a very demanding scenario, despite the striking development of renewable energy, since more than 80% of global primary energy is still fossil-fuel-based.

Most decarbonizing scenarios consider a relevant role of carbon capture, utilization, and storage (CCUS) technologies. Namely, all mitigation approaches will need to offset between 100 and 1000 Gt CO₂ during the 21st century. Besides, not implementing CCUS would increase the mitigation costs by 29%–297% to limit the atmospheric CO₂ concentration at 450 ppm by the end of the century.¹

In this context, CCUS will very likely play a relevant role in the next decades. Not surprisingly, carbon capture is one of the fastest growing topics in sustainable chemistry and separations science. Enormous efforts within the scientific community have widely focused on CO₂ capture from large stationary sources such as electricity-generating power plants to minimize emissions. In recent years, the capture of CO₂ from other industrial sources (e.g., cement, steel, refineries, etc.), and also from ambient air, has attracted rapidly increasing attention.

The use of the captured CO₂ as feedstock to produce value-added products beyond a compressed gas for enhanced oil recovery or geological storage has also been an attractive topic for research. Many catalytic applications such as artificial photosynthesis, photocatalysis, and the synthesis of chemistry building blocks, fuels, and pharmaceutical compounds have been proposed.

Here, we present a special issue on CO₂ capture and utilization, compiling more than 40 selected publications submitted by internationally renowned researchers on this multidisciplinary research field, covering novel advances in process optimization, modeling, reactor design, and high-performance materials. In the following pages, we will summarize the most relevant features of the main research lines and the findings presented by the contributions to this special issue.

■ CO₂ CAPTURE

Amine scrubbing is the most developed technology for post-combustion CO₂ capture. The typical flowsheet of this technology is similar to an industrial acid gas removal system using a chemical solvent. Since amine scrubbing relies on

thermal swing regeneration, a greater heat of CO₂ absorption reduces energy consumption. This effect of thermal swing is dependent on the temperature of the reboiler, which is limited by the thermal degradation of the solvent. Degradation must be minimized not only to moderate the cost of the process but also to reduce the emissions of toxic byproducts. Degradation behavior and related toxicity must be well understood before amine absorption is used commercially. Piperazine derivatives show faster CO₂ absorption rates and greater resistance to thermal degradation, compared to other second-generation amines, although further investigation is required to assess the corrosion tendency of these advanced amine-based solvents.^{2,3} In addition, less time-consuming and reliable techniques for solvent monitoring are essential to ensure efficient and safe operation. Infrared spectroscopy, combined with principle component regression (PCR), allows for a rapid measure of solvent composition, compared to traditional approaches such as chromatography.⁴ Amines can also absorb aromatics during gas washing processes. A deep knowledge of CO₂–hydrocarbon mixtures is essential for the design and operation of absorption systems. Binary parameters can be obtained from empirical models that can satisfactorily describe the thermodynamics of this type of amine solutions for wide ranges of temperatures and pressures.⁵

The electrochemically mediated amine regeneration (EMAR) is an alternative method to the conventional thermal regeneration of amines aimed at reducing the energy demand of the CO₂ capture process. The EMAR process does not require steam integration, because only electrical energy is used for the regeneration of the solvent. This is a great advantage in the retrofitting of CO₂ emitter plants, because the capture unit can perform independently of the main plant.⁶ A large amount of heat is consumed during the regeneration of aqueous amine solutions, not only to increase the temperature of the solution until the stripping temperature but also to provide the inevitable energy for water evaporation. A significant reduction in energy demand is possible by using organic solvents instead of water. Mixtures of pentane with monoethanolamine (MEA) show good solubility and high selectivity to CO₂. Moreover, a considerably lower amount of energy must be supplied, because this system requires a lower

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regeneration temperature than the conventional MEA-based process.⁷

Apart from post-combustion applications in large stationary CO₂ emitters, chemical solvents are also suitable for capturing the CO₂ generated in other sources. Marine transport accounts for 3% of global CO₂ emissions. The utilization of amine absorption systems to reduce carbon emissions in ships has gained interest recently. The models of marine diesel and ship energy systems have been developed and validated, showing the potential of solvent-based systems to significantly reduce the carbon footprint at a reasonable cost. The recirculation of exhaust gas greatly reduces energy demand, because the sensible heat contained in the recycle is suitable for the reboiler.⁸

Intensive research over the past years has also shown that adsorbent-based capture technologies have the potential to significantly reduce the energy penalty (OPEX) and/or equipment costs (CAPEX), when compared with state-of-the-art amine-based solvent capture processes.⁹ Adsorption-based capture is based on a cyclic process where CO₂ molecules interact and bond with the adsorbent material either through weak intermolecular forces (physisorption), or through strong covalent bonding (chemisorption). By swinging the thermodynamics of the system, for instance, pressure and temperature, either the adsorption or the desorption step is favored.

The modification of porous materials via the loading of amines is a well-known technique to obtain solid chemisorbents. These hybrid adsorbents aim to reduce amine corrosivity, evaporation loss, and heat-consuming regeneration, while maintaining their selectivity toward CO₂.

Amine functionalization by grafting involves the chemical reaction between a silane and the surface of a porous material. Conventional (dry) and wet grafting of silanes on silica gels are reported by Yang.¹⁰ Large pores and the wet method were found to enhance CO₂ adsorption performance. By grafting linear and branched silanes onto mesoporous silica, it was found that branched compounds yield higher CO₂ uptakes.¹¹ The thermal and oxidative stabilities of the materials were also assessed.

Impregnation is another major functionalization route. In this case, there is no specific reaction with the pore surface, so a higher N load can be achieved compared to grafting. Large amines or even polymers are used for this purpose. Nevertheless, obtained adsorbents usually present a poor cycling or stability performance. The most commonly impregnated compound is polyethylenimine (PEI). The combined effect of PEI and a silane on silica gel was explored, reaching better results than PEI itself.¹² Furthermore, the group of Prof. Sayari has covalently immobilized PEI to porous Al-silica to overcome the disadvantages of the impregnation technique, directing his efforts to investigate the oxidative stability of the materials.¹³

Measuring adsorption kinetics on amine-loaded sorbents is crucial not only for material development purposes but also for reactor design and process development. New methods are currently being explored¹⁴ to measure fast adsorption kinetics in novel microfluidized bed thermogravimetric analyzers. Obtained apparent adsorption kinetics were described by a two-stage model of the linear driving force model.

Non-aminated adsorbents are also used, provided they present a sufficient selectivity toward CO₂. Activated carbons obtained from agricultural wastes are also considered. Date

seeds¹⁵ and hazelnut shells¹⁶ were used as raw materials in a laudable effort to achieve adsorbents within the circular economy system.

In parallel to the above-mentioned development of adsorbent materials, significant breakthroughs in separation science have been made through the discovery of novel nanoporous materials (e.g., metal organic frameworks, MOFs) for low-temperature separations.¹⁷ These breakthroughs have afforded the synthesis of innumerable different nanoporous structures. By combining organic ligands and metal-ion nodes in MOFs, over 70 000 different MOFs have been synthesized, and even more structures have been proposed computationally.¹⁸ Hence, we can now tailor-make materials with well-defined properties.

However, promising materials must be assessed and modeled at a process level as process integration plays a key role in the energy efficiency and economics of capture processes. The evaluation and screening of adsorbent materials is frequently done by making use of basic material properties metrics (i.e., working capacity, selectivity, ...) or by metrics guided by simplified process models. More-complex process models would provide more insights into the required performance of the materials for cyclic separation processes, but they are much more time-consuming to develop and computationally expensive. Park et al. investigated how a simplified pressure swing adsorption (PSA) model would differ from a detailed model if both were to be used for the screening of MOFs at subambient temperatures for post-combustion CO₂ capture.¹⁹ They identified the CO₂ swing capacity and the adsorbent performance score as being useful in predicting the ranking of materials for their studied process. Also, in an effort to avoid computational time in solving nonequilibrium adsorption-based models, the concept of the Thiele modulus-effectiveness factor from heterogeneous catalysis was adopted into describing mass transfer in an adsorbing nonequilibrium adsorption system.²⁰ This approximation was proofed to provide a simplified and accurate description of intraparticle mass transfer in adsorptive systems.

In addition, novel advanced materials are normally synthesized in powder form, and they are difficult to handle when implemented in the process. For instance, large pressure drops in fixed-bed contactors is a well-known problem when using powder capture adsorbent materials. Those process-derived issues can be addressed by shaping the adsorbents into different structures (e.g., foams, monoliths, polymer beads, etc.). Understanding the optimum structured form of the material for a particular capture process is a key aspect to advance these novel promising technologies, and this has been the focus of many ongoing research activities. For instance, a three-dimensional (3D) printing technique based on liquid printing dopes was applied by the group of Rezaei to develop 3D-printed MOF monoliths, specifically MOF-74 and HKUST-1, with the potential for possible scaling-up of the monoliths.²¹ At the process level, the performance of NaY monoliths was investigated in an intensified vacuum pressure swing adsorption (VPSA) unit to capture CO₂ from a high partial pressure stream in a small-scale steam methane-reforming process.²² Also, spherical MOF beads (CPO-27-Ni) were tested in a moving-bed TSA process and compared to zeolite 13X.²³ Because of the wide variety of adsorbents and working conditions available for CO₂ capture, Prof. Sofía Calero has proposed an algorithm to select the optimal

combination by combining molecular simulation and machine learning.²⁴

CO₂ capture technologies based on high-temperature solid looping cycles have also undergone great progress in the past decade, showing their potential to capture CO₂ at moderate cost and low energy penalty. Calcium looping uses CaO-based materials as CO₂ sorbents. Limestone is a widely available, inexpensive, and environmentally benign sorbent. However, this natural material typically shows fast deactivation after multiple carbonation/calcination cycles, because of solid sintering, giving, as a result, residual activities of <10% after 25–30 cycles. The development of high-performance synthetic sorbents is also a matter of great interest for calcium looping applications, especially in those operated with clean fuels (e.g., natural gas, syngas). The production of these upgraded materials is significantly more expensive; therefore, a compromise between durability and cost is required. In an attempt to reduce costs, several techniques are being developed, such as sol–gel synthesis, precipitation, impregnation, granulation, dry mixing, and coating. Particles of well-distributed nanosized CaCO₃ coated with alumina result in stable morphological structures that highly improve the CO₂ uptake capacity of the material.²⁵ Although calcium looping has been mainly developed so far for post-combustion applications, many investigations are focused on combining this technology with other processes. The separation of CO₂ using CaO in steam methane reforming or gasification processes is an attractive option for the sustainable conversion of fossil fuels to high-purity H₂. The combination of CO₂ sorbent and catalyst in all-in-one materials should improve mass and heat transfer in these processes. The development of advanced particle models that accurately describe the kinetics of these combined materials should facilitate the scale-up of this technology once they have been implemented and validated in adequate reactor models.²⁶ Another interesting possibility is combining calcium looping with thermal storage. The integration with concentrated solar plants allows the energy required for sorbent regeneration to be obtained from a renewable source, giving, as a result, a great reduction in fuel consumption associated with the CO₂ capture process.²⁷

Chemical looping combustion (CLC) is another promising high-temperature CO₂ capture technology, in which fuel is oxidized by a solid oxygen carrier that is reduced. This avoids the dilution of the combustion products with N₂ from air, thereby generating virtually pure CO₂. Common oxygen materials are transition metals, such as oxides of Ni, Cu, Mn, or Fe. Most investigations on CLC have been focused on arrangements of interconnected fluidized-bed reactors. Because of that fact, oxygen carriers must show not only high and stable reactivity to ensure complete fuel combustion during operation, but also high mechanical strength to minimize the loss of active material by attrition. The use of suitable supports, such as zirconia²⁸ or MgO–kaolin,²⁹ allows the activity of the O₂ carriers to be maintained in the long term, while reducing the particle attrition rate measured in laboratory-scale fluidized-bed reactors. CLC with solid fuels has recently gained great interest. The use of biomass is particularly interesting as the CO₂ that is captured can be considered as a negative emission. Detailed simulations of this process allow a deeper knowledge of the optimal operating conditions to be acquired, which is essential to facilitate the scale-up of this technology.³⁰ Although the control of temperature and the replacement of spent materials are much easier in fluidized

beds, the generation of power by means of CLC integrated in a combined cycle (i.e., at high pressure) is only feasible using fixed-bed reactors. A suitable reactor configuration, accompanied by appropriate heat management strategies, improves the energy efficiency of the process.³¹ Chemical looping technology can also be adapted for the production of H₂ or syngas with inherent CO₂ capture. A cocurrent moving-bed reactor loaded with an iron–titanium oxygen carrier is able to partially oxidize methane to syngas containing H₂/CO ratios of ~2, while almost complete fuel conversion and negligible carbon deposition are achieved.³² CO-rich gas can be produced by means of reverse water–gas shift of mixtures of CO₂/H₂ using spinel-structured ternary ferrites.³³ Pure H₂ can be obtained by means of water-splitting CLC of natural gas. An excess of heat can be stored in reduced O₂ carrier particles, and their subsequent oxidation generates high-temperature heat suitable for iron ore sintering plants.³⁴

Alternative novel technologies for capture applications include membrane-based ones, where CO₂ separation is achieved by permeating one or more constituents of the gas stream through a selective membrane that acts as a barrier, while excluding other components of the stream. Membranes benefit from the lack of the regeneration step, a waste-free character, and compact size. However, the molecular-level understanding of mass transport phenomena and the development of scalable methodologies for their manufacture are some of the main hurdles to overcome in order to advance next-generation membranes to industrial application. A review study on the current state-of-the-art for CO₂ absorption using membrane contactors is provided by Chuah et al.³⁵ The paper focuses on membrane materials, liquid absorbents, process design, and pilot-scale demonstration of membrane contactor processes. Critical challenges, such as membrane fouling and pore wetting, are also discussed. Membrane separation properties are strongly affected by the presence of gases other than CO₂ in the gas feed to the membrane system. Single-gas and mixed-gas permeation in DDR-type zeolite membranes is also reported.³⁶ The effect of temperature and CO₂ concentration on permeance and selectivity was systematically investigated.

When the process of capturing CO₂ from a wide range of emission sources, such as power generation sources, industrial sources, and even directly from the air, is coupled with new CO₂ conversion routes, CO₂ accumulation in the atmosphere could be reduced. If CO₂ capture is coupled with permanent storage solutions, even a shift to negative emissions could be possible. For instance, the direct CO₂ capture from air (DAC) combined with permanent storage is considered a promising negative emission technology.³⁷ This technology requires vast contact surfaces with a great affinity and selectivity toward CO₂. Absorption methods generally use a strong alkaline aqueous solvent solution, such as Ca(OH)₂, NaOH, or KOH, that reacts with CO₂ to yield carbonated salts. The use of membrane electrodialysis for the regeneration of the solvent and CO₂ recovery is shown to be less energy-intensive than other methods, but the high cost of these bipolar membranes hinders, for the moment, the scaling up of this technology.³⁸ Potassium carbonate is also a chemical solvent that can be used in direct air capture. This material typically shows high stability and CO₂ absorption capacity, slow corrosiveness, little toxicity, and low cost. The regeneration of the saturated solvent demands low temperatures and the CO₂ recovered is suitable

for subsequent chemical synthesis applications, such as the production of CH₄ by catalytic methanation with H₂.³⁹

■ CO₂ UTILIZATION AND THE CHEMICAL INDUSTRY

The climate crisis poses a huge challenge to the chemical industry, because they produce carbon-based products, mostly derived from fossil carbon, and they will ultimately need to be decarbonized. Three different routes—CCS, CCU, and a bio route—were qualitatively compared by Gabrielli and co-workers for the production of a generic chemical product and quantitatively compared for the case of methanol production.⁴⁰ A clear winner was not identified and all routes proved to be feasible and had different pros and cons, with key hurdles being identified for each route. A scenario analysis of CCUS supply chains was specifically performed for Italy and Germany.⁴¹ They minimized an objective function that consisted of remission of an element of carbon tax, economic incentives, and revenues subtracted from total supply chain costs, to determine optimal designs for CCUS systems.

Currently, main CO₂ utilization research areas include (1) thermochemical conversion and hydrogenation routes, (2) electrochemical and photochemical conversion, (3) CO₂ conversion to solid carbonates, (4) biological conversion routes, and (5) enhanced hydrocarbon recovery with carbon storage. The development of these technologies to ensure net CO₂ emissions is a challenge, and there are still many fundamental and technological aspects to be solved. For instance, the development of effective thermal and (photo)-electrochemical catalytic reaction pathways, the understanding of the kinetic mechanism for minerals and industrial waste systems for inorganic carbonate formation, or the acceleration of biological CO₂ conversion routes, among others.

In routes (1) and (2) above, active research is currently being conducted to develop green solid heterogeneous catalysts that can contain multiple active sites and efficiently convert CO₂ to high-value-added products. Zhang et al. contributed to these efforts by reporting a green bifunctional heterogeneous catalyst that can be used for the formation of cyclic carbonates through the CO₂ cycloaddition reaction.⁴² Similarly, efficient CO₂ electrochemical reduction under mild conditions also requires high-performance catalysts. Different catalyst electrodes (Sn/Cu, BiSn/Cu, Bi₂Sn/Cu, Bi₃Sn/Cu, Bi₄Sn/Cu, and Bi/Cu) were fabricated by electrodeposition, and their activity, stability, and selectivity toward CO₂ reduction to formic acid was measured. In addition, insights into the proposed mechanism that is catalyzed by the electrodes were provided by density functional theory (DFT) calculations.⁴³ DFT modeling was also the tool used to understand the reaction mechanisms of the direct catalytic conversion of CO₂ and methanol to dimethyl carbonate (DMC).⁴⁴

The conversion of CO₂ from exhaust gas marine emissions via a hydrogenation route was investigated by Iliuta and Larachi.⁴⁵ They explored, by process modeling, the combination of CO₂ reduction with hydrogen under microwave-induced plasma conditions and catalytic methanol synthesis in an integrated process. CO₂ hydrogenation to methanol was also investigated by Geng et al.,⁴⁶ but their focus was the promotional effect associated with bimetallic phosphide catalysts. They showed that the combination of Ru and Mo in a bimetallic catalyst provides a favorable interaction with CO₂ through electronic effects to promote hydrogenation toward methanol. Nickel-based catalysts for methane dry

reforming (MDR) are the focus of the work of Li et al. They aimed to suppress the well-known problems of carbon formation and metal sintering, due to the high temperature of the process, by using microwaves as an energy source to drive the MDR reactions under mild conditions (below 220 °C).⁴⁷

The CO₂ utilization route based on CO₂ conversion to solid carbonates involves a complex set of reactions by which CO₂ reacts with Ca, Mg, and/or Fe oxide-bearing phases to yield the corresponding solid carbonate phase. Both gas–solid and aqueous conversion processes have been extensively studied to perform the carbonation reaction. Direct aqueous mineral carbonation of heat-activated lizardite is the focus of the paper by Benhelal and co-workers, where they employed different analytical techniques to characterize carbonation products and to provide insights into the mechanism of formation and the structure of silica-rich byproducts.⁴⁸ A refining slag feedstock with calcium-containing wastewater was considered by Chen et al.,⁴⁹ where they proposed a high-gravity carbonation process. Various liquid agents and their effect on calcium ion leaching behavior was investigated, and reaction kinetics were identified by using a surface coverage model.

As an example of biological conversion routes, photoautotrophic microorganisms such as microalgae have the ability to convert CO₂ to biomass with sunlight via photosynthesis. They simultaneously assimilate CO₂ and organic and inorganic pollutants and produce a range of useful intracellular metabolites. Baroni et al. investigated the role of nitrogen availability in the production and release of extracellular matter in three commercially relevant microalgae.⁵⁰ Variations of extracellular substances, in relation to microalgae species and cultivation conditions, were reported, with important implications for species selection and optimization of microalgae production, harvesting, and dewatering processes.

Regardless of the CO₂ utilization route that is followed, in order to be able to successfully implement CCU, several other factors, such as sustainability of materials and energy processes, economics, and permanence of the CO₂ in the carbon-neutral or low-carbon materials, must be carefully evaluated, following a standardized approach. The upcycling of CO₂ also offers the opportunity for making use of local available renewable energy resources. Ultimately, the development of CCU technologies will also impact the deployment of CCS technologies, and all the knowledge that can be gained from research into those areas will certainly help us in achieving our carbon targets by 2050.

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Notes

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