PERSPECTIVE

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Organic photochemistry for direct light-driven separations

Ariel Y. Wang, [a] Bayu I. Z. Ahmad, [b] Carolyn Ma,[b] Phillip J. Milner [*b] and Richard Y. Liu [*a]

Compared to conventional methods, direct light-driven separations are promising strategies to achieve high selectivity while lowering energy cost. This perspective examines recent progress in photoswitch-enabled separations with a focus on concentration of CO_2 and selective anion recovery from water. We highlight key design strategies for photo-pH-swing capture and identify current practical limitations toward translating solar-driven separations into technologies with meaningful impact.

Introduction

Efficient chemical separations are indispensable to the functioning of modern society. Beyond commodity chemical production, separation science underpins technologies that sustain and improve quality of life, from the purification of essential resources like air and water to the recycling of nutrients such as nitrogen and phosphorus. Despite their necessity, industrial scale separation operations remain wasteful and energy intensive. Separations account for more than half of the total industrial energy expenditure[1] and up to *ca.* 15% of global energy consumption.[2,3] In addition, up to 80% of these processes involve thermally driven steps (*e.g.*, distillation, evaporation, drying, *etc.*), which depend heavily on heat from fossil fuel sources,[3–5] further contributing to the current climate crisis. Recent efforts to integrate renewable energy sources into industrial systems, in the form of green electricity,[6–9] have produced some environmental benefits. However, apart from technological challenges of integrating green energy with traditional separation methods, there are additional issues with the storage and distribution of clean energy.[10] With rising energy demands, renewable electricity alone cannot meet long-term sustainability and emissions goals without also reducing energy requirements at the process level. Recognizing this challenge, several national and international agencies have identified the development of next-generation, low-energy separation technologies as a cornerstone of sustainable industrial practice and decarbonization strategy.[4,5,11–13]

Unlike traditional thermal methods, non-thermal separations can operate without macroscopic phase transitions, instead exploiting molecular differences in size and/or chemical affinity to achieve selective transport or adsorption. Among these, membrane-based separations have emerged as a dominant platform,[14,15] particularly in water desalination,[16–20] gas purification,[21,22] and bioproduct recovery.[23–26] This expansion has been driven largely by progress in polymer synthesis and fabrication, which has led to improvements in membrane performance, lifetime, and applications.[27,28] The impact of non-thermal separations is exemplified by reverse-osmosis desalination membranes, which have lowered energy demand from 16 kWh m^{-3} to *ca.* 2 kWh m^{-3}.[16] Apart from polymer-based porous membranes, the rise of porous crystalline materials, such as metal–organic frameworks (MOFs), further revolutionized sorbent design by offering unprecedented tunability in terms of pore size and chemistry.[29–31]

Although bulk purifications have advanced significantly, many modern challenges, such as carbon capture, uranium recovery from seawater, and rare earth metal purification from ores, require isolating targets from extremely dilute input streams. Concentrating from dilute sources not only requires high selectivity but also the ability to overcome a significant entropic penalty. One strategy to achieve this is the use of chemical sorbents that can form very strong bonds with the target species, thereby driving chemisorption. While this allows for selective capture from dilute sources, significant energy input is then required to drive the desorption process and regeneration of the sorbent. For amine-based systems, the prototypical chemisorbent, regeneration can account for up to 80% of the cost of operation.[32,33] The trade-off between affinity, selectivity, and energetic cost continues to be challenging for both thermal and non-thermal separation systems.

Aiming to achieve a more favorable energetic profile, researchers have become interested in metastable systems that, upon activation of an external stimulus, can generate non-equilibrium states that participate in separations. Known approaches harness external changes in pressure,[34] voltage,[35] or electromagnetic fields,[36] to generate reversible chemical

aDepartment of Chemistry and Chemical Biology, Harvard University, 12 Oxford St., Cambridge, MA 02138, USA. E-mail: richardliu@chemistry.harvard.edu

bDepartment of Chemistry and Chemical Biology, Cornell University, Ithaca, NY, USA. E-mail: pjm347@cornell.edu

Table 1 Summary of CO_2 capture methods reviewed

Ref	Method	System	pH range	Operating temperature	Capture capacity	Quantum yield	Reversibility
52	pH swing	Indazole mPAH and glycine in water	8–10	Room temperature	15.6 μmol CO_2 mL^{-1} h^{-1} (20 mM solution under DAC conditions)	—	3 cycles with diminishing yields
53	pH and temperature swing	Pyranine and K_2CO_3 in water	7.55–7.65	~40 °C	26.7 μmol CO_2 mL^{-1} h^{-1} (0.1 M solution from 15% CO_2 in N_2)	0.125 photon utilization	No degradation over 168 cycles (7 days)
59	pH swing	Merocyanine in 3:1 DMSO/water	6.9–9.9	Room temperature	~6 μmol CO_2 mL^{-1} h^{-1} (20 mM solution from 10% CO_2 in N_2)	1.3%	No difference in activity over 3 capture and release cycles
60	pH swing	Fluorenol photobase in 2:1 ACN/water	8.5–11.6	Room temperature	0.56 μmol CO_2 mL^{-1} h^{-1} (10 mM solution under DAC conditions)	20.1%	No difference in activity over 3 capture and release cycles
61	pH swing	PyDIG and GlyGly in water	6.88–7.81	23–27 °C	1.46–1.81 μmol CO_2 mL^{-1} h^{-1} (1 mM solution under DAC conditions)	4.01%	No difference in activity over 5 capture and release cycles
72	Photo-isomerization	Azobenzene-guanidine in ethanol	—	~40 °C	2.4 μmol CO_2 mL^{-1} h^{-1} (0.2 M solution under DAC conditions)	—	No degradation in activity over 3 capture and release cycles
74	Photo-tautomerization	2MeBP and BzOK in DMSO	—	60–65 °C	26 μmol CO_2 mL^{-1} h^{-1} (0.1 M solution from 10% CO_2 + 3% O_2 in N_2)	—	No degradation over 5 capture and release cycles

changes that facilitate selective transport or desorption. This perspective highlights an emerging frontier: light-driven separation processes.[37] Because sunlight is the most abundant renewable resource, directly coupling photon energy to separation steps bypasses the inefficiencies of generating, storing, and distributing electricity. Photoresponsive materials, especially those incorporating molecular photoswitches, offer a direct way to translate photon energy into chemical or structural transformations, generating out-of-equilibrium states that are capable of selective and efficient capture/release. In the following sections, we review recent advances in light-driven separations, with an emphasis on their applications in CO_2 capture and water purification. We will discuss key design strategies, material considerations, and future directions for enabling scalable and low-energy photoresponsive separation technologies (Table 1).

Light-driven CO_2 concentration

Capturing CO_2 from point sources or directly from the atmosphere is an important component of broader climate-mitigation strategies. CO_2 also serves as a precursor to key commodities such as methanol and urea, giving it additional value as a chemical feedstock. Over the past 15 years, a multitude of new carbon-capture technologies have emerged.[38–40] Most sorbents rely on highly exothermic interactions that overcome the entropy loss during concentration. Common functional groups such as amines and hydroxides form interactions on the order of tens of kJ mol^{-1} with CO_2, which, while beneficial for capture, make regeneration a major hurdle.[41,42] Thermal CO_2 desorption is not only energy-intensive[43] but also introduces secondary issues, such as water evaporation and sorbent decomposition.[44,45] Thus, an important objective in CO_2-capture system design is to enable low-energy, on-demand desorption. Light is an appealing trigger for this purpose because it is abundant, spatially addressable, and can be coupled selectively to specific molecular events. The following section outlines recent advances in light-driven strategies for separation of CO_2, particularly highlighting methods involving pH swing and photoisomerization.

Light-induced pH swing

Capture-and-release using a photoinduced pH-swing has emerged as a leading strategy for light-driven CO_2 separation.[37,46] Because the solubility of CO_2-derived inorganic species in water is strongly pH-dependent, photoresponsive chemistries capable of modulating pH upon irradiation offer a means to drive reversible CO_2 absorption and release (Fig. 1a). Among these systems, metastable photoacids, molecules that induce a substantial, reversible decreases in pH upon excitation, have gained prominence. Photoacids have had a long history of applications in lithography,[47] catalysis,[48] and polymer synthesis,[49] but more recently have been adapted as light-responsive agents for regenerating CO_2 sorbents. Once excited, photoacids provide a transient reservoir of protons that
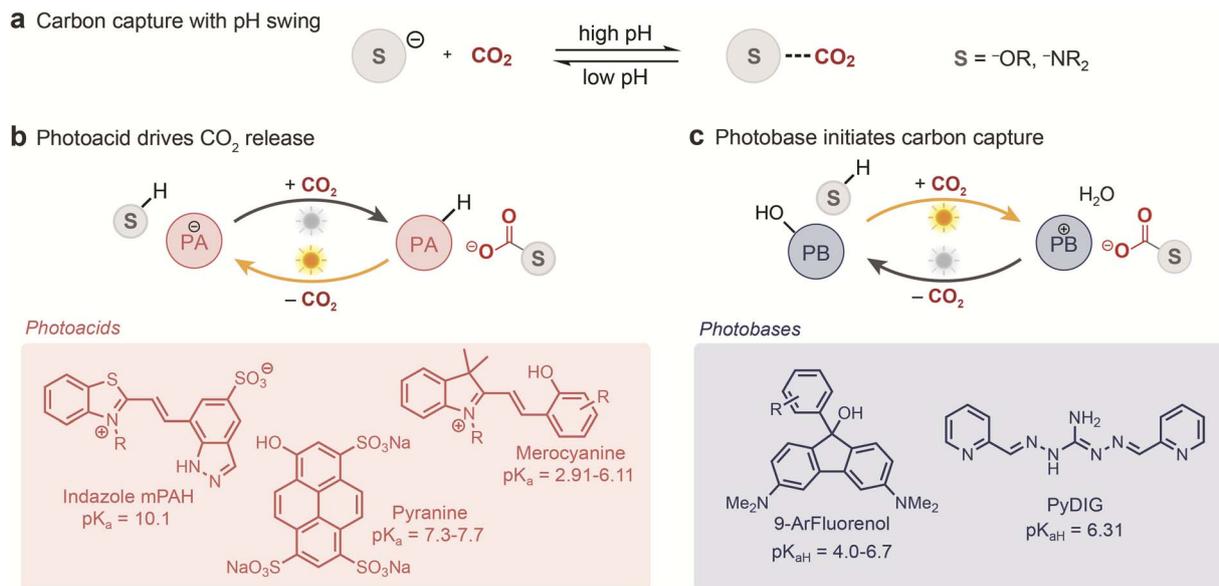
Fig. 1 (a) Carbon capture and release using pH swing. Grey S stands for sorbent. (b) CO_2 is captured by sorbent in the absence of light and released by a proton from an irradiated photoacid. (c) Irradiated photobase initiates carbon capture and CO_2 is released as the photobase relaxes in the dark.

can react with the CO_2–sorbent adduct, thereby promoting CO_2 release and sorbent regeneration (Fig. 1b).

Although the formation of carbonic acid using an excited-state photoacid had been observed spectroscopically,[50] the use of photoacids for triggering CO_2 release in bulk solution was not explored until recently by Bennett *et al.*[51] Using a merocyanine photoacid, the authors showed that an already acidic bicarbonate-containing solution could release CO_2 in an accelerated fashion under irradiation. Although not integrated into a full capture-and-release cycle, this study provided a crucial proof of concept that photoacids can generate sufficiently large pH shifts to promote gaseous CO_2 release. Building on this foundation, Ma and co-workers reported in 2023 that an indazole-based photoacid could drive CO_2 release from glycine solutions saturated with ambient CO_2, achieving up to 78% release after 1 h of irradiation.[52] Although cyclic CO_2 uptake and release was not performed, the results indicate that photoacids may offer a viable route for regenerating amine-based DAC sorbents.

Hatton and co-workers have shown that glycine-based aqueous capture systems can be photochemically regenerated using the water-soluble, excited-state photoacid pyranine.[53] Pyranine's combination of photostability, high quantum yield and excellent water solubility makes it an ideal target for carbon capture applications. Mixtures of pyranine and up to 0.8 M glycine (a *ca.* 800-fold increase compared to previous studies)[52] were able to sequester and release CO_2 from an input stream of 15% v/v CO_2 in N_2 (simulating an industrial flue gas mixture). Control experiments indicated that the mechanism for bulk CO_2 release was dominated by photothermal heating and water-vapor formation. In fact, photoacid-induced pH changes near the solution surface contributed significantly only during the first ~30 seconds of simulated release, as calculations showed

that the population and lifetime of excited states are too low to support significant bulk pH changes. To study photoacid-induced CO_2 desorption in isolation, the authors constructed a continuous-flow photochemistry setup and demonstrated sustained CO_2 release from solutions preloaded in the dark with a 15% v/v CO_2 in N_2 feed. Continuous operation of this flow system over 1 week yielded 60 mmol of CO_2, with a photon utilization of 0.125 CO_2 molecules liberated per photon. Under simulated DAC conditions (*ca.* 420 ppm CO_2 in ambient air), continuous operation sequestered 3 μmol over 2 h. These results demonstrate that light-swing operation is feasible in practical flow architectures and highlight the strong dependence of photorelease efficiency on reactor design.

To bypass limitations such as water solubility and photoacid hydrolysis, Dawlaty and co-workers attempted to directly release CO_2 from monoethanolamine (MEA) with a merocyanine photoacid.[54] As in aqueous systems, CO_2–MEA adducts can be reversibly driven toward CO_2 release in the presence of added protons, and a total of *ca.* 30 mg (0.3%) mass loss from bulk solution was observed over 14 h. Relative to dark controls, irradiated MEA–photoacid solutions exhibited roughly twice the mass loss, attributed primarily to CO_2 release. This study demonstrated the feasibility of photoacid-induced CO_2 release in amines, but it also highlighted several practical constraints. Pure MEA is highly viscous at rt, over 20 times more viscous than water,[55,56] requiring heating to 80 °C to permit efficient CO_2 outgassing. Although this is below the temperature required for conventional thermal release (*ca.* 120 °C for amine sorbents),[57] it still imposes a substantial energy burden for continuous operation. Furthermore, CO_2 release is slower in MEA than in water-containing mixtures (*e.g.*, a 10:1 H_2O/MEA mixture), consistent with slower proton-transfer dynamics. These limitations pose significant barriers for scaling such systems, and

alternative strategies to make aqueous-based capture systems more efficient and robust may be preferable.

Changes to the solvent system have been found to strongly impact both long term stability and release capacity of photoacids in water. To systematically probe the role of solvation, Lukatskaya and co-workers conducted an in-depth study of merocyanine photoacids in DMSO/water mixtures.[58] Their results revealed that optimizing the solvent composition could profoundly enhance both photoacid performance and stability. At an optimal DMSO mole fraction of 0.15, the merocyanine photoacids exhibited a fivefold increase in CO_2 release and more than doubled their lifetime relative to pure water. DMSO served as a stabilizing cosolvent, improving solubility and suppressing hydrolytic degradation, but at the expense of diminished pH swing and slower relaxation back to the ground state. To rationalize the observed trends, the authors used molecular dynamics simulations, which revealed a distinctive microsolvation pattern: DMSO primarily coordinated to the nonpolar regions of the photoacid, while water is localized near the polar phenolic and carbonyl sites. This selective solvation pattern explains the disproportionate impact of DMSO on stability. For instance, DMSO coordination at the alkene, the site most susceptible to hydrolytic attack, can impede decomposition even at relatively low concentrations.

The preceding examples have involved two separate components: a traditional sorbent to perform CO_2 capture (e.g., amine and hydroxide), and a photoacid to regulate release. This raises a natural question: can a single compound mediate both capture and release steps? In principle, the merocyanine formed upon ring opening of spiropyran contains a phenoxide that may be sufficiently basic to form salts with carbonic acid. Leveraging this property, Liu and co-workers developed a one-component merocyanine photoacid system that enabled CO_2 separation under simulated flue-gas conditions (10% v/v CO_2 in N_2).[59] To achieve efficient and cyclic CO_2 separation processes, they showed that the pK_a (dark) of the photoacid must be carefully matched to the effective pK_a of CO_2 (ca. 6.25). If the photoacid is too basic, not enough protons are available for effective release; if it is too acidic, the dark pH becomes too low to enable effective capture. Although developed in the context of a single organic component, this principle can be extended naturally to multicomponent systems. During capture and release, protons are shuffled between the sorbent and the photoacid, so optimal performance requires matching the pK_a values of the key components of the system.

Light can also be used not only to trigger CO_2 release but also to drive CO_2 capture itself. As systems achieving higher pH have better potential and greater capacity for fast CO_2 uptake, photobases, which transiently raise solution pH upon irradiation, offer a more promising strategy for capturing CO_2 from dilute streams (Fig. 1c). Recent work from the Liu group demonstrates that newly developed fluorenol photobases can indeed drive CO_2 capture from dilute streams, including air.[60] These Arrhenius photobases transiently release hydroxide ions upon photoexcitation, driving CO_2 capture through bicarbonate formation. Once the light is removed, the photobase relaxes and the pH returns toward its initial value, promoting CO_2 release.

Liu and co-workers achieved efficient capture under simulated DAC conditions (400 ppm CO_2) and enabled subsequent release at concentrations more than sevenfold higher than that of the input stream.

In a complementary approach, Custelcean and coworkers recently reported the use of a diiminoguanidine photobase (pyDIG) to initiate capture with peptide sorbents.[61] Upon irradiation with UV light, the pH increase leads to deprotonation of the zwitterionic peptide, initiating carbon capture. Under this system, a 120 mL solution of 1 mM pyDIG/peptide captured up to 0.0467 mmol of CO_2 within a 24 h period under irradiation, maintaining similar performance over four cycles. The thermal ("dark") release process has a weaker driving force, taking up to 5 days to complete at rt, but the process can be sped up to 2 d by heating to 80 °C. Although photobase-driven capture remains in early stages of development, these results highlight a compelling complement to photoacid strategies – providing a light-capture, dark-release pathway particularly suited to low-temperature CO_2 separations.

Guiding principles for pH-swing-driven carbon capture

Given the number of carbon capture systems based on photoacids and photobases that have been reported to date, several fundamental principles governing their operation can be gleaned. Light-driven carbon capture systems that operate through a pH-swing mechanism involve proton transfer between dissolved CO_2 and the photoswitch. In photoacid systems, the solutions typically begin at high pH, which decreases during CO_2 uptake from the gas phase (Fig. 2b). When exposed to light, the photoacid transitions to a metastable state that favors releasing its proton to the surrounding medium. This proton transfer shifts the equilibrium of bicarbonate and/or carbamate species back toward molecular CO_2, reversing the capture process. In comparison, photobases operate via the opposite pH trajectory. Upon photoexcitation, a photobase transiently increases the solution's pH by abstracting protons, thereby promoting the capture of CO_2 by hydroxides. When the photobase relaxes to its ground state, its acidity is restored, resulting in proton donation and subsequent CO_2 release (Fig. 2c). Thus, both photoacids and photobases rely on reversible, light-mediated pH modulation to toggle between CO_2 capture and release.

The efficiency of these proton relays is intimately tied to the acid–base equilibria governing CO_2 speciation, from carbonic acid to its various basic forms (Fig. 2a). Although the true pK_a of carbonic acid is closer to 3.4,[50,62] it is a minor species under relevant conditions. Accounting for the hydration and solubility equilibria of CO_2 results in the useful effective value of $pK_a = 6.35$ for CO_2 at 25 °C. Liu's work has shown that photoacids with ground-state pK_a values near this threshold value are best suited for efficient CO_2 release. Photoacids with lower pK_a will have a decreased capacity for CO_2 capture. In contrast, photoacids with higher pK_a cannot efficiently release CO_2, as the equilibrium always favors carbon capture over release. Similarly, in

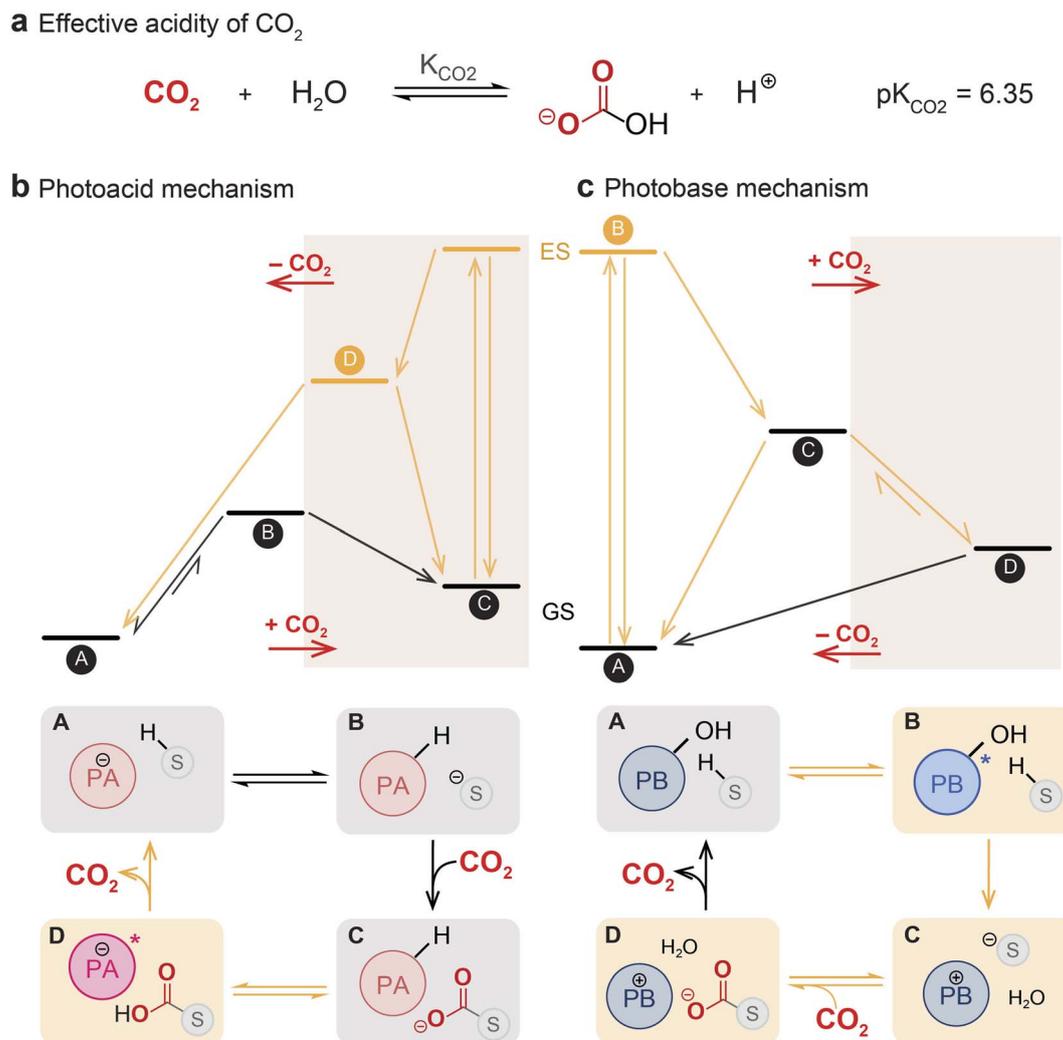**Fig. 2** (a) Effective acidity of dissolved CO_2 in water. (b and c) Summarized energy diagram of photoacid and photobase driven DAC, respectively. Yellow and black arrows indicate light-dependent or independent equilibria.

photobase systems, the optimal fluorenyl photobase used by Liu and co-workers has a conjugate acid pK_a of 6.1, which allows for effective CO_2 release in the dark. Some exceptions exist to this pH-matching principle: for example, pyranine, with a ground-state pK_a of *ca.* 7.3, was found to release CO_2, although this is largely through bulk photothermal heating rather than through a pH swing.

In general, a well-designed photoswitch should have a large pK_a difference between ground and metastable states. In addition, the ground-state pK_a of the photoacid (or conjugate pK_a of the photobase) should be near the effective pK_a of CO_2 (6.35). Tuning the initial pH of the system with basic additives can improve the overall capture ability of the system, which was reflected in the increased capturing capacity observed by Ma and co-workers upon initial pH adjustment of their system with KOH.[52] However, such pH modifications can also alter the speciation and photochemical behavior of the photoswitch, potentially reducing its capacity to donate or accept protons. Added sorbents that act as buffers (for example, amines) should

be considered in combination with the photoactive agent, and the behavior of the composite system is not easily predicted from the pK_a values of the photoswitch alone.

CO_2 release using light-induced isomerization

Another strategy for inducing CO_2 release relies on light-driven structural changes within the sorbent compound. Light-induced geometric isomerization is a hallmark of photoswitches and has been leveraged in a number of materials applications, including biomacromolecules,[63] polymers,[64] and molecular machines.[65] Many photoresponsive materials are based on azobenzene chemistry, which undergoes *trans*-to-*cis* isomerization upon irradiation with light. For CO_2 sorbents, isomerization can introduce unfavorable steric clashes around adsorption sites that destabilize bound CO_2 and shift the equilibrium towards release (Fig. 3). This approach has been applied largely in the solid state, including porous metal–
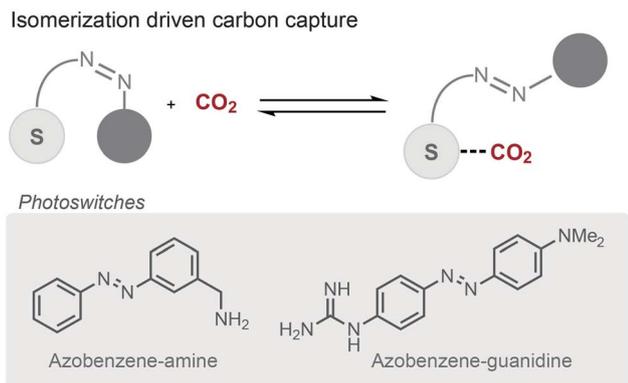
Fig. 3 Photoswitch isomerization driven carbon capture.

organic and covalent organic frameworks.[66–70] Although these studies establish the potential of isomerization-induced release for sorbent regeneration, they have not yet demonstrated separation from dilute streams such as ambient air.

In principle, solid-state adsorption has several advantages over solution-state sorption, including bypassing limitations with solubility, offering improved long-term stability and gravimetric capacities, and reducing energy losses from solvent evaporation.[71] However, such solid-state systems suffer from limitations of their own, including poor thermal transport, slow adsorption kinetics, and the need for additional mechanical processing. Recent work by Inagaki and co-workers illustrates several challenges with solid-state DAC.[72] The authors developed an amine-based sorbent that incorporates an azobenzene switch to promote CO_2 release. From elevated CO_2 concentrations, the sorbent was able to achieve the theoretical capacity and release limit of the system (1 CO_2 per 2 amine sites). However, the carbon-capture efficiency was highly dependent

on the CO_2 input concentration, and light-triggered release under DAC conditions yielded a lower loading ratio of 2 CO_2 molecules per 5 amine sites. In addition, solid-state systems appear extremely sensitive to changes in the adsorption site: substituting the amine with guanidine eliminated CO_2 capture entirely, even though the more basic guanidine was expected to enhance uptake (Fig. 3). By contrast, transferring the system from the solid state to ethanolic solution restored the activity of the guanidine-based sorbent, achieving up to a 1:1 CO_2 per amine capture ratio. The resulting sorbent–CO_2 adduct could be isolated as a precipitate and the bound CO_2 released over a 2 h period. The ability to sequester CO_2 as a solid adduct can be valuable, improving the ease of handling and transportation, with release occurring at centralized hubs or points of use, such as greenhouses or chemical manufacturing facilities.[73]

Photenol capture

In all of the photoswing CO_2 separations outlined above, one of the two steps—capture or release—was carried out in the dark. This invites the question: is a fully light-driven process for CO_2 capture and release possible? Such a process would benefit from potential isothermal operation as well as the distribution of the energetic requirements across both steps. Milner and co-workers recently reported such a process using the inexpensive and photoresponsive organic molecule 2-methylbenzophenone (2MeBP, Fig. 4a).[74] Previous work had shown that, upon UV light irradiation, 2-methylbenzophenone analogs can photo-tautomerize to the corresponding photoenols, which can then undergo cycloaddition with dienophiles such as CO_2.[75–78] Subsequent ring-opening (in the case of CO_2) leads to the corresponding arylacetic acid derivative. While previous work had shown that these molecules are capable of CO_2 release, UV light
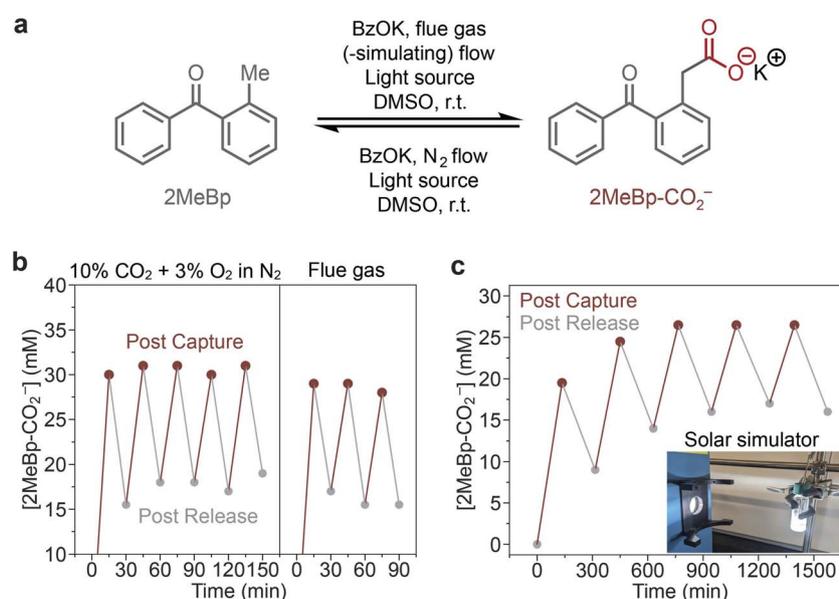

Fig. 4 (a) 2MeBp captures and releases CO_2 in the presence of visible light. (b) Simulated carbon capture from different gas mixtures. (c) 2MeBp carbon capture with a solar simulator instead of 390 nm light.

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and long reaction times were required.[79] Because the photodecarboxylation of 2-arylacetate derivatives is a common process in photoredox catalysis,[80] the authors hypothesized that treatment of the obtained arylacetic acid product with base to generate the corresponding carboxylate ($2MeBP–CO_2^-$, Fig. 4a) would greatly accelerate CO_2 photorelease. The successful combination of these two processes enabled the first light-driven process for CO_2 capture and release.[74]

Milner and co-workers commenced by demonstrating that CO_2 capture with the photoenol derived from 2MeBP can be conducted under more realistic conditions than in previous studies, including with visible light irradiation (390 nm), dilute CO_2 streams (10% v/v CO_2 in N_2), short reaction times (30 min), and in the presence of O_2 as a contaminant. Next, the feasibility of CO_2 release was evaluated. As expected, CO_2 release from the carboxylic acid product was negligible upon light irradiation or heating to 60 °C. In contrast, irradiation of $2MeBP–CO_2^-$ with light (390 nm) led to rapid CO_2 release and reformation of 2-MeBP upon back proton transfer. Remarkably, the presence of base during the capture step led to enhanced CO_2 capture *via* the corresponding photoenolate. This is advantageous because it means that the base can remain in the reaction mixture during both the capture and release steps. Transient absorption (TA) spectroscopy measurements and density functional theory (DFT) calculations support that the decarboxylation step proceeds *via* a unique intramolecular photodecarboxylation pathway on the ps timescale.

To assess the ability of the combined CO_2 capture and release system, the authors performed cycling experiments using more realistic mixtures. 5 cycles of capture and release under constant light irradiation were achieved, simply by switching between 10% v/v CO_2 in N_2 flow (capture) and pure N_2 flow (release) to simulate a light-driven, pressure-swing process (Fig. 4b, left). CO_2 removal was also achieved from a sample of flue gas from Cornell University's natural gas-fired power plant (Fig. 4b, right) and using a solar simulator instead of 390 nm light (Fig. 4c). Capture and release were also performed in the window of the authors' laboratory in Ithaca, NY, demonstrating the potential for this system to mediate CO_2 removal using natural sunlight as the primary energy input. Future studies should focus on increasing the kinetics of the capture and release steps and improving the absorbance overlap of 2-methylbenzophenone variants with the solar spectrum.

Anion capture and water purification

Contamination of drinking water with certain anions represents a significant challenge for water potability. These include inorganic anions such as sulfate, arsenate, nitrate, perchlorate, and selenate, as well as organic anions such as per- and polyfluoroalkyl substances (PFAS). Previously overlooked anions such as chloronitramide have also recently been identified as hazardous contaminants of municipal water supplies.[81] Numerous sorbents have been developed to remove these ions from water and make it fit for human consumption.[82–85] However, the high binding affinities of these complexes hinder their subsequent regeneration. To address this issue,

supramolecular complexes that change their anion-binding affinities upon light irradiation have been identified.[86–95] However, most of these compounds have not been employed to achieve the effective removal of anions from water, but were merely shown to modulate the concentration of free anions in aqueous solution.

Recently, Flood and co-workers reported a pioneering example of photoswitch-based anion removal from water.[96] Specifically, they demonstrated that the cyanostilbene macrocycle cyanostar[97] is capable of extracting the hexafluorophosphate (a byproduct of Li-ion battery manufacturing) and perrhenate (a surrogate for radioactive pertechnetate-99)[98] anions from aqueous solution. Irradiation with light causes some of the *trans*-stilbene units to isomerize to their *cis* conformers, dramatically decreasing the anion-binding affinity of the macrocycle and triggering release (Fig. 5a and b). In its all-*trans* form, this macrocycle displays high affinities for larger anions (*e.g.*, hexafluorophosphate) over common anion contaminants (*e.g.*, chloride, nitrate, and sulfate), making it a suitable platform for selective ion extraction from water (Fig. 5c). The cyanostar binds large anions through the formation of a 2 : 1 complex, with the anion trapped between two molecules of the macrocycle, as confirmed by single-crystal X-ray diffraction.[97]

The ability of cyanostar to mediate selective anion removal from water was evaluated using a liquid–liquid extraction (LLE) setup. First, cyanostar was shown to facilitate the partitioning of tetraethylammonium hexafluorophosphate from the aqueous phase into the organic phase, even in the presence of competing chloride. Next, the release of free anions was evaluated by irradiating the anion-bound complex with 365 nm light. Release of the bound anion was supported by NMR spectroscopy, which confirmed both the *trans* → *cis* isomerization of the stilbene units in cyanostar as well as the recombination of released hexafluorophosphate anions with tetraethylammonium cations in solution. The authors then designed an LLE experiment by simply using anion-binding to extract hexafluorophosphate from the aqueous phase to the organic phase using cyanostar in the dark, and then releasing it back into fresh water upon light irradiation. Remarkably, the extraction of the anion was quantitative even in the presence of large excesses of competing chloride (up to 4 M), supporting the potential viability of this system for real-world applications.

The authors next turned their attention to extracting perrhenate from water. Both sodium and ammonium salts were employed, and the exchange of perrhenate was validated using UV-vis spectroscopy. Under similar LLE conditions as employed for hexafluorophosphate capture, quantitative perrhenate extraction from the aqueous phase to the organic phase was observed in the dark. Light irradiation allowed for 87% release of perrhenate back into the aqueous phase. A similar LLE for pertechnetate-99 was also demonstrated down to a concentration of 400 ppb, using the radioactivity of each phase to track anion extraction and release (Fig. 5d). Tethering two cyanostar macrocycles into a "clamshell" preorganizes the requisite 2 : 1 host–guest architecture and enabled extraction of pertechnetate-99 from solutions as dilute as 4 ppb, reflecting its
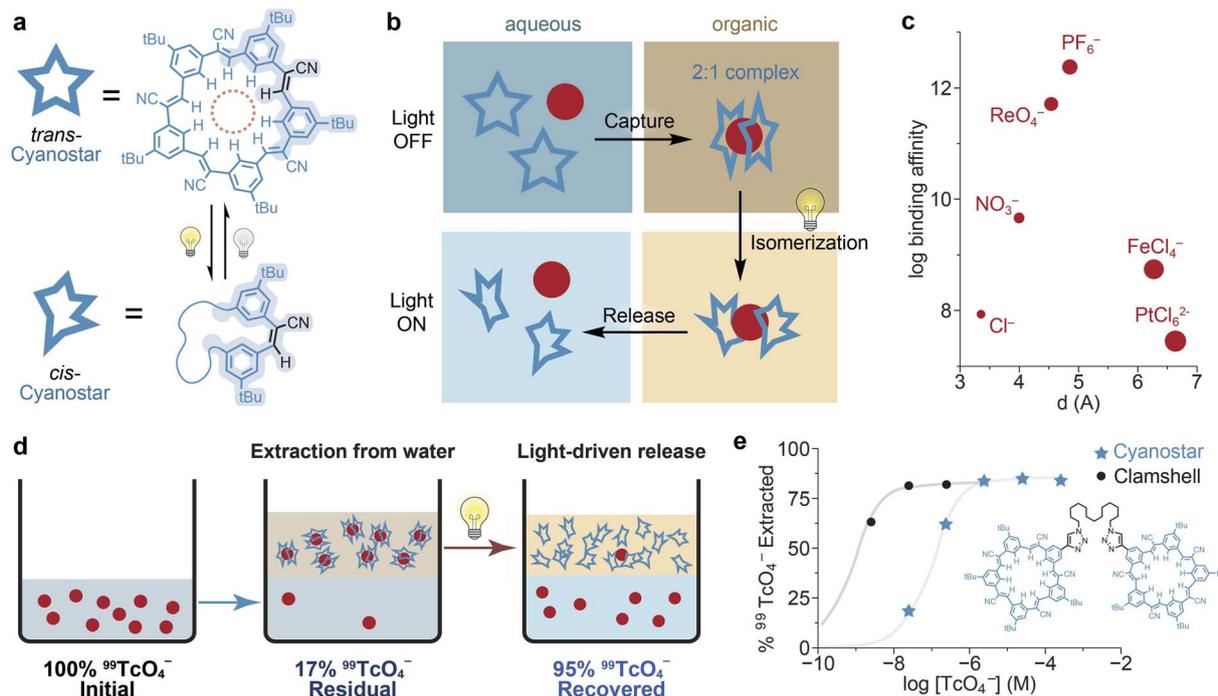
Fig. 5 (a) Structure and photoisomerization of cyanostar macrocycles. (b) Illustrative scheme of liquid–liquid extraction of anions through selective binding by cyanostar and their subsequent light-driven release. (c) Preference of such capture for size-matched anions. (d) Application of such system for the extraction and recovery of radioactive TcO_4^-. (e) Clamshell cyanostar allows effective anion capture at lower concentrations.

exceptional binding affinity (Fig. 5e). Overall, Flood's work represents a significant step forward for the use of photo-responsive cages to mediate anion capture from aqueous solutions in an LLE setup. The reliance on a 2 : 1 binding mode (reducing the sorption capacity) and the need for UV-light irradiation are practical limitations that may be addressed in future work. The labor and resource intensity of LLE[99] could also be overcome by adopting such a reactive motif within a porous material to enable solid-phase extraction.

Outlook

One of the central challenges in advancing light-driven separations is understanding and controlling how external factors, notably the solvation environment and additives, influence the behavior of photoresponsive molecules. For instance, subtle variations in solvent composition, initial pH, or irradiation wavelength have been shown to produce unexpectedly large shifts in photoacid performance, defying simple mechanistic interpretation.[52,54,58] These sensitivities highlight that photoacid performance is not an intrinsic molecular property, but emerges from complex solvent–solute interactions that modulate proton transfer, stability, and switching dynamics. Beyond solvent composition, additives such as amino acids, bases, and electrolytes further complicate the chemical environment, and it is currently difficult to predict their effect on photoswitching from first principles. Progress in both experimental photophysics[100,101] and computational modeling[102] are crucial to

improving our understanding of key intermediates and equilibria governing photoacid performance and long-term stability.

For light-driven separations to advance beyond proof-of-concept demonstrations and mature into deployable technologies, laboratory discoveries must translate into systems that are scalable, robust, and process-relevant. To date, most experimental studies have emphasized fundamental aspects, such as photoacid stability, pH-swing dynamics, and mechanistic pathways for CO_2 release, studied in small-scale, idealized environments. While such controlled investigations are essential for elucidating underlying mechanisms, the next stage of development demands integrated systems capable of operating under realistic process conditions and at meaningful throughput. This includes the use of realistic (sun)light fluxes and inclusion of realistic contaminants, including water, oxygen, SO_x, and NO_x, in input streams. In addition, long-term cycling stability (e.g., over thousands of cycles) has yet to be demonstrated for any of the systems discussed herein.

Process modeling can play a central role in identifying bottlenecks, guiding design trade-offs, and prioritizing the parameters critical for large-scale optimization. For example, a recent preprint by Lukatskaya and co-workers modeled a fully light-driven carbon-capture workflow based on spiropyran-mediated pH swings.[103] This analysis explored the operational window required to achieve high CO_2 recovery while minimizing energy input. The authors found that attaining >90% CO_2 recovery with substantially reduced electricity demand would require photoacid concentrations exceeding 0.3 M.

Reaching near-molar photoacid concentrations in aqueous media presents a formidable solubility challenge, and additional molecular or system-level modifications may be needed to maintain photochemical reversibility at such high loadings. Additional modeling of viable process and scrubber designs is needed to help push photoswing separations towards realistic operation.

Scaling present-day photoswing separations systems will require concurrent optimization across multiple dimensions, from the molecular design of the photoswitch to the architecture of realistic photoreactor platforms. Meeting these challenges will necessitate interdisciplinary collaboration, bridging synthetic chemistry, photophysics, and process engineering to establish a pathway toward practical, light-driven separations.

Author contributions

All authors contributed to writing of this manuscript.

Conflicts of interest

P.J.M. and B.I.Z.A. are listed as inventors on a patent that includes a fully light-driven carbon capture process using photoenols (Cornell University, international patent application no. PCT/US24/42589, filed 15 August 2024). Harvard University is in the process of applying for patent(s) on the use of photobases for CO_2 capture, on which R.Y.L. is listed as an inventor.

Data availability

No primary research results, software or code have been included and no new data were generated or analysed as part of this review.

Acknowledgements

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