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Challenges and Opportunities in Electrification of Adsorptive Separation Processes

Abdullah Al Moinee, Ali A. Rownaghi,* and Fateme Rezaei*

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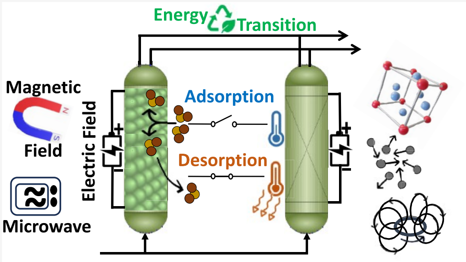
ABSTRACT: **A global energy transition from fossil-based to renewable-based systems requires advancements in various energy sectors. Current chemical separation processes are quite energy-intensive and require significant advancement to make this transition happen. In the energy transition era, reducing the dependency of separation processes on fossil-based thermal energy seems inevitable for transitioning smoothly and meeting the stringent timelines. In particular, adsorption-based separation processes have the potential to be fully electrified through innovative strategies in developing stimuli-responsive adsorbents and swing processes. In this review, we discuss the recent efforts in electrification of adsorption processes and provide an overview of emerging materials and processes. The challenges associated with electrified adsorption-based separations are discussed in detail, and opportunities to expedite the transition from traditional practices to advanced energy-efficient systems are provided in the end.**



STATE-OF-THE-ART ADSORPTIVE SEPARATIONS

In the United States, separation processes account for 10−15% of the total national energy usage. Most of these processes rely on energy-intensive thermal methods like distillation and evaporation, which demand substantial energy, escalating overall energy consumption and environmental impact.[1] Compared to legacy thermally-driven separations, adsorption offers a more efficient path to a sustainable future by lowering the energy demand and environmental footprints.[2,3] The current state-of-the-art adsorption-based separations rely essentially on pressure or temperature changes within the column to facilitate adsorption and desorption of targeted adsorbates and include pressure or vacuum swing adsorption (PSA or VSA), temperature swing adsorption (TSA), or a combination of pressure, temperature, and vacuum swing adsorption (e.g., TVSA, PTSA, VPSA).[4−7] Amidst ongoing research on conventional processes, the exploration of cutting-edge emerging mechanisms, such as electric (electrothermal) swing adsorption (ESA),[8] electrochemical swing adsorption,[9] supercapacitive swing adsorption (SSA),[10] electric field swing adsorption (EFSA),[11] magnetic induction swing adsorption (MISA),[12] and microwave swing adsorption (MSA),[13] as shown in Figure 1, offers promise and potential toward advancements in adsorptive separation field.

Adsorptive separation, with a 60-year track record in gas and liquid purification, holds potential in various separation and purification applications, including air separation,[8] toxic gases abatement, H_2 purification,[14] wastewater treatment, protein adsorption,[15] olefin/paraffin separation, C_5−C_6 alkynes and olefins separation,[16] gas drying,[17] CO_2 capture,[9] etc., as shown in Figure 2. Unlike steady-state conventional processes, adsorption operates inherently in unsteady states, thereby allowing for process optimization and simplification of complex designs for specific separations.

THE NEED FOR ELECTRIFICATION OF ADSORPTIVE SEPARATIONS

The field of adsorption has witnessed significant progress in the design of high-performance adsorbents and efficient adsorption swing processes over the past few decades. In that regard, combined theoretical and experimental efforts have enabled a better understanding of the energy requirements of such systems (Figure 3),[18] yet complexity of the process and regeneration energy demand still hamper the scale-up of

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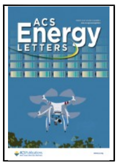

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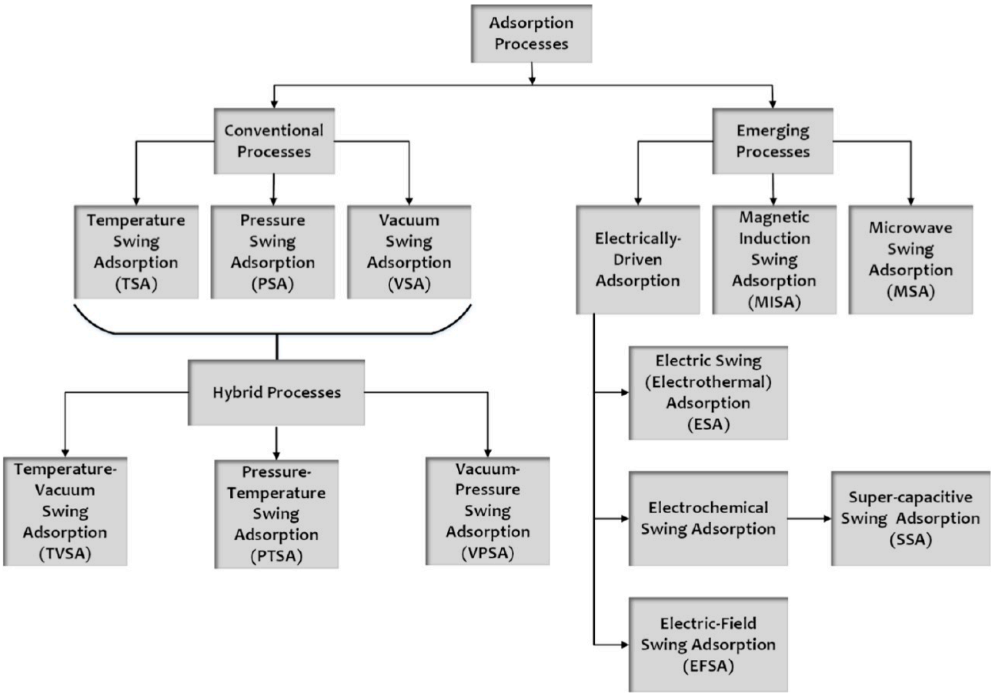

Figure 1. Different adsorptive separation processes.

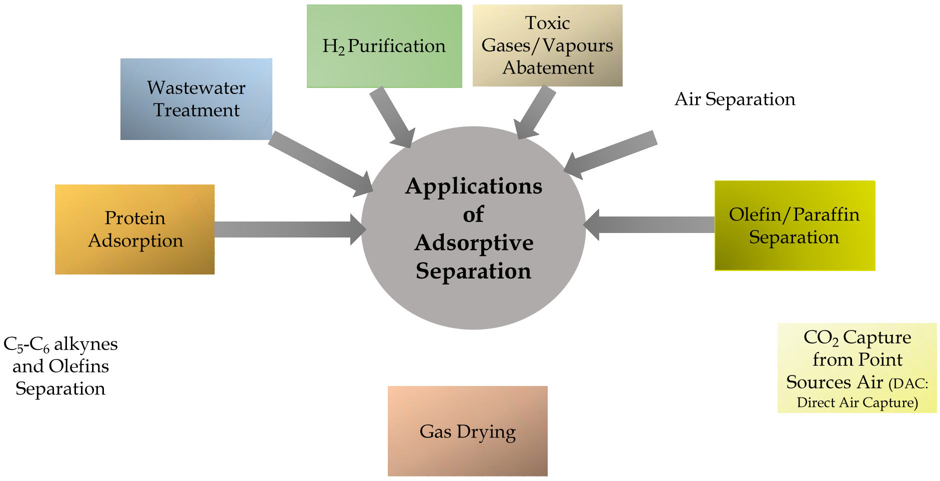

Figure 2. .

adsorption-based separations in many fields.[19] To further advance energy efficiency and sustainability, the electrification of adsorptive separation processes presents promising potential utilizing green electricity.[20]

Conventional PSA is a proven technology that was commercially deployed for air separation in the 1960s,[21] yet it often suffers from high pressure drop across the adsorption column, making the separation process energy-intensive.[22,23] Similarly, industrial deployment of TSA processes is usually hampered by lengthy cycles (on the order of hours) due to long heating and cooling steps.[24]

The opportunities inherent in electrified adsorption processes are underpinned by their potential to accelerate desorption or cooling rate, significantly boosting the overall throughput. Unlike conventional methods, direct electrification for regeneration eliminates the necessity for simultaneous heating and cooling during purging. In conventional approaches, the purge gas often coincides with the heating or cooling process, impacting efficiency and energy requirement. However, with direct electrification, the decoupling of purging from the heating or cooling phase reduces energy demand significantly, thereby increasing operation flexibility.[25] This will in turn lead to shorter cycles and hence higher throughput, less adsorbent inventory, and compact separation units. Electrochemical and electrothermal systems represent promising avenues for minimizing parasitic energy losses, in that they can operate under near-isothermal conditions,

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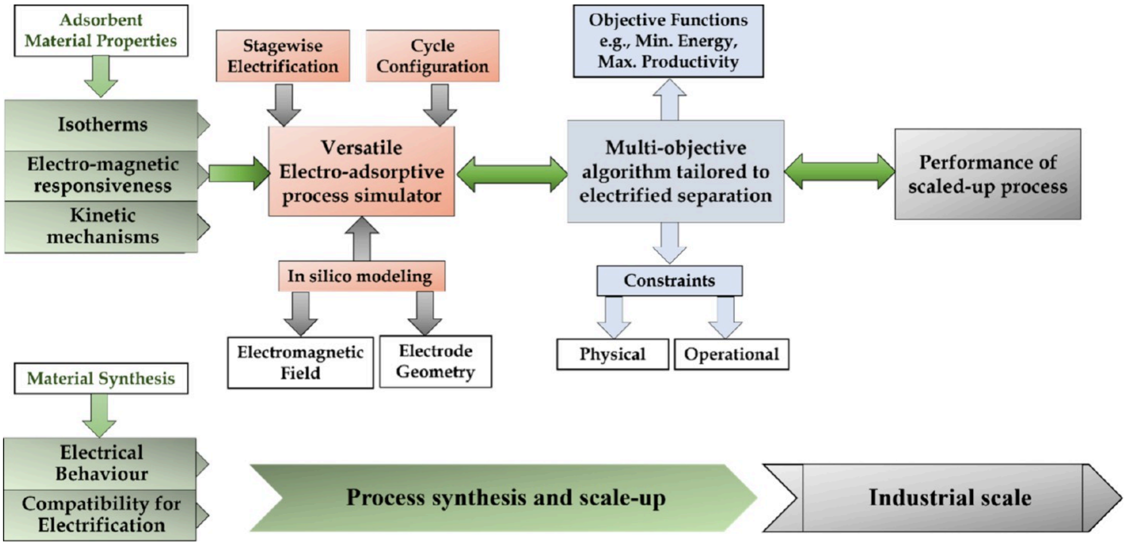

Figure 3. Framework for electrification design of an optimal adsorptive separation process. Image reproduced with permission from ref 18. Copyright 2012 Elsevier.

influencing diffusion processes and offering significantly higher efficiencies compared to conventional TSA and PSA methods.[9]

■ ELECTRICALLY DRIVEN ADSORPTIVE SEPARATION PROCESSES

Electrically driven adsorptive separation processes include separation techniques that utilize electrothermal heating, electrochemical heating, or electric field-induced heating to regenerate the adsorbent during desorption step. In this section, we delve into the myriad opportunities that electrification brings to the forefront, with a focus on electro-swing adsorption processes. We explore cutting-edge advancements and novel strategies in developing electric-responsive adsorbents. Additionally, we discuss the intricate challenges associated with electrified adsorption-based separations, providing a comprehensive perspective on the roadblocks that must be overcome.

Electrothermally Driven Adsorptive Separation Processes. Electrothermally driven adsorptive separation refers to well-known ESA processes that employ electrical heating to trigger adsorbate desorption and adsorbent regeneration. This involves controlled temperature changes in the adsorbent to facilitate the desorption of the target adsorbate molecules. Basically, this method relies on internal heat generation within the adsorbent through Joule heating upon passing the electric current, thereby enabling a much faster desorption rate compared to TSA. Compared to traditional TSA processes, electrothermally driven processes offer potential advantages:

- Higher heat efficiency by directly delivering heat to the adsorbent.[26]
- Heating rate is independent of heating capacity of heat source and heat transfer between adsorbent and source.[8]
- Faster heating allows the designs of smaller systems.[8]
- Mass and heat fluxes align in the same direction, potentially enhancing the desorption performance by leveraging the effects of energy and mass diffusions.[8]
- Independent control of purge gas flow regardless of heating rate makes it possible to achieve higher recovery.[27,28]

Sustaining the inspirational aspect of these advantages, ESA processes have emerged as one of the promising contenders at the forefront in the drive toward electrification. This technology that harnesses electric field to directly deliver energy to the adsorbent for regeneration has been already demonstrated at industrial scale for volatile organic compounds (VOCs) abatement and recovery.[7,29] In a review (2014) by Grande and co-workers,[8] the fundamentals and applications of ESA processes, which rely on a conceptual framework, were discussed; Figure 4 provides a history of efforts that have contributed to the development of methodologies for electrified adsorption and separation systems. As depicted in this figure, the increasing trend of prospect and progress in the research and development of ESA per year shows that the pursuit of electrification in adsorptive separation is not only a contemporary research avenue but also a thriving domain ripe with challenges to be addressed and opportunities to be harnessed simultaneously.

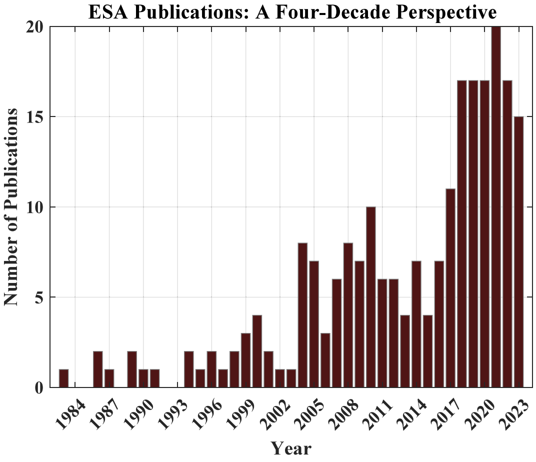

Figure 4. Exploring opportunities in ESA research from 1983 to 2023 (based on Scopus).

The concept of ESA has been investigated for various applications, with a history spanning over 40 years since its patent in 1971 by Fabuss and Dubois.[30] For instance, Mallouk

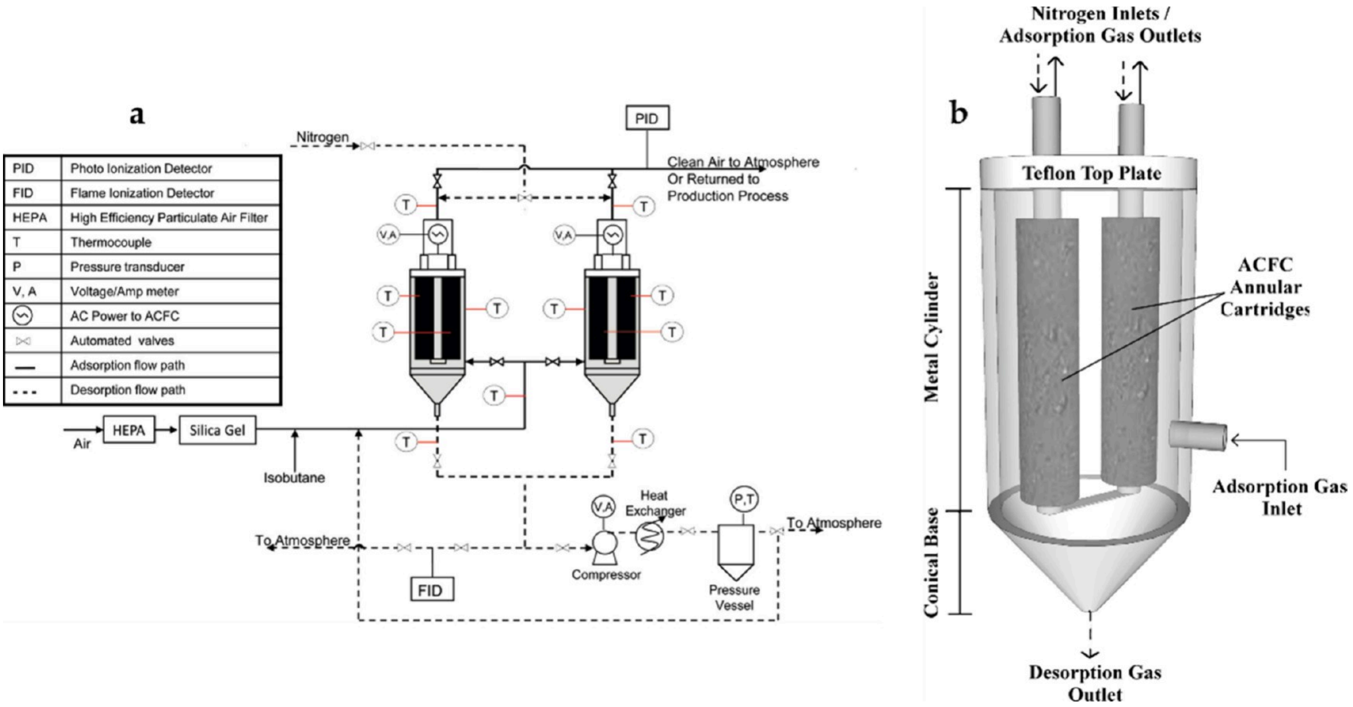

Figure 5. (a) Experimental setup of an ESA process for the adsorption of isobutane on ACFC and (b) adsorption/desorption vessel with two annular ACFC cartridges. Flow paths for adsorption and desorption cycles are represented by solid and dashed lines, respectively. Image reproduced with permission from ref 27. Copyright 2010 American Chemical Society.

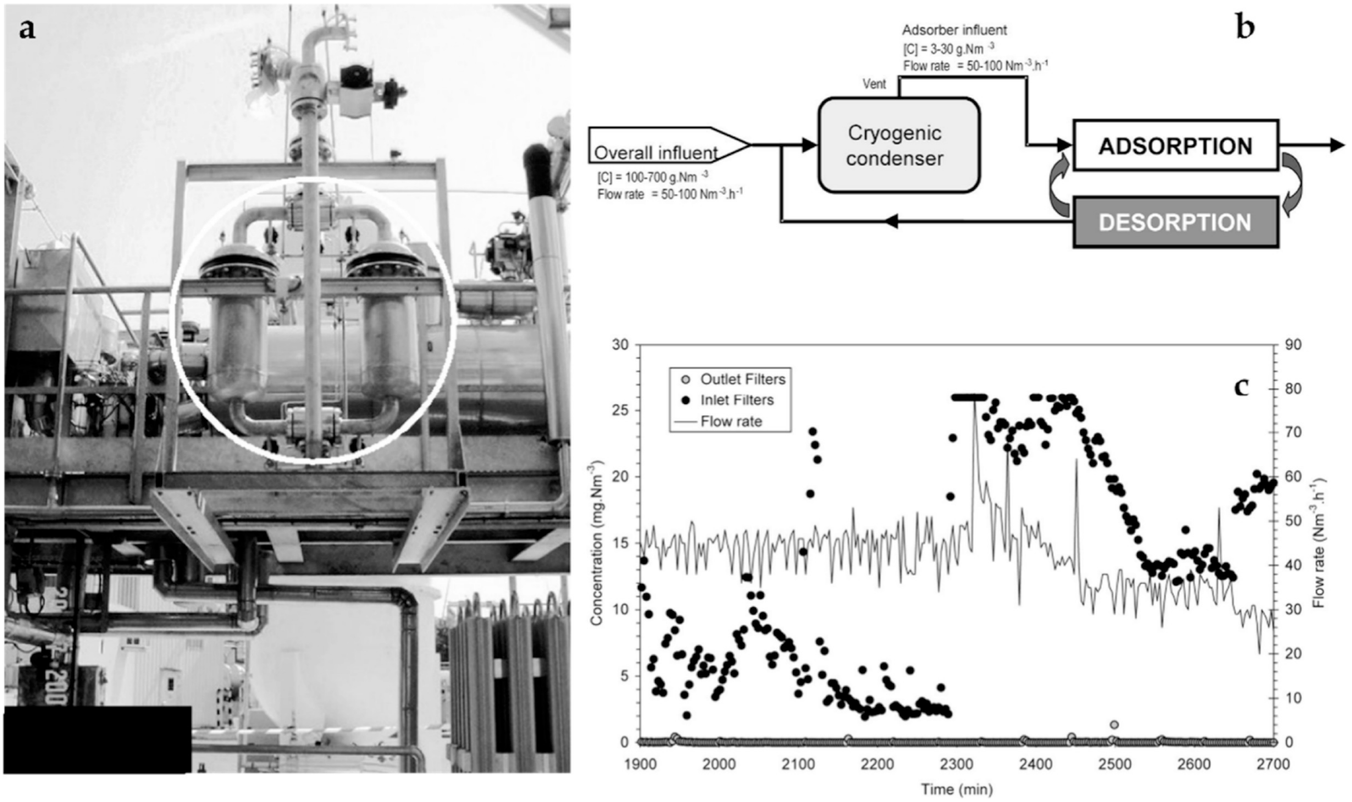

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Figure 6. (a) Electrothermal swing system for VOCs removal by Air Liquide, (b) simplified flow diagram of the process, and (c) concentration and flow rate in the adsorber influent and effluent. Image reproduced with permission from ref 29. Copyright 2010 Taylor & Francis LLC.

et al.[27] demonstrated isobutane adsorption over an activated carbon fiber cloth (ACFC), followed by electrothermal desorption and subsequent liquefaction, as depicted in Figure 5. The process achieved over 98% capture efficiency, highlighting its effectiveness in retaining isobutane molecules. The mixture was introduced into one vessel, passing through

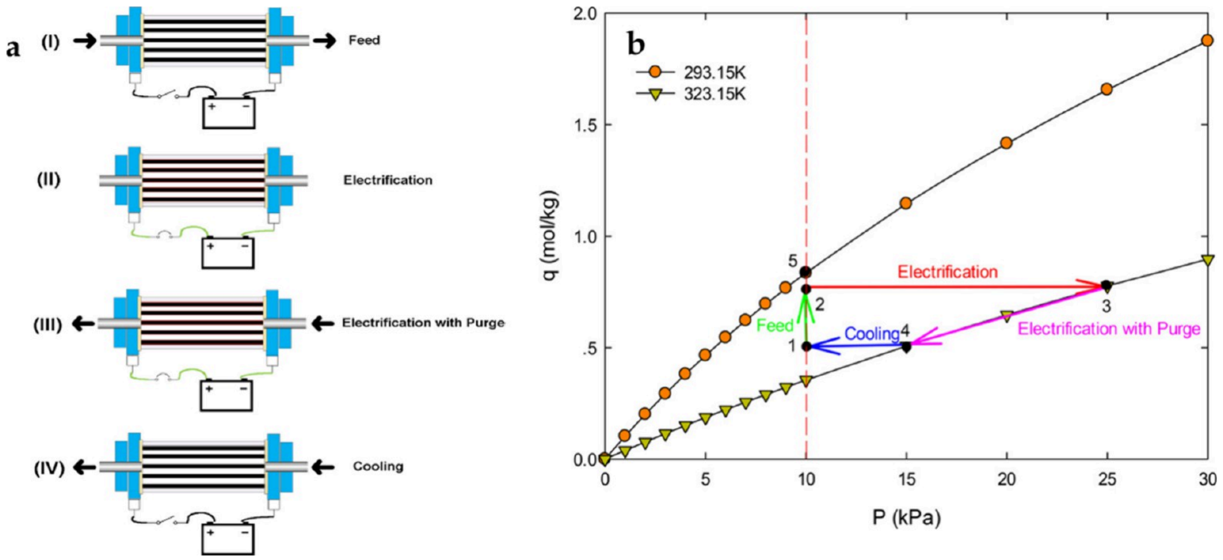

Figure 7. (a) Schematic diagram of a four-step ESA cycle and (b) quantitative representation of the ESA cycle in a *q*-*P*-*T* diagram. Image reproduced with permission from ref 30. Copyright 2018 Elsevier.

the ACFC cartridges, where isobutane was adsorbed. Simultaneously, the desorption process took place in the other vessel. N_2, introduced at the top of the desorbing vessel, carried desorbed isobutane through the heated annular ACFC and exited at the outlet. The direct electrothermal heating utilized a silicon-controlled rectifier (SCR) to control the root-mean-square voltage applied to the cloth. This study focused on the ACFC-ESA system, where ACFC was exceptionally capable of electrothermal desorption, and the system was proven for recovering ultradilute organic gases (e.g., toluene, methyl ethyl ketone, and acetone). It is also worth mentioning here that electrothermal desorption offers distinct advantages compared to vacuum or steam regeneration in terms of energy efficiency and independently controlling adsorbent heating and purge gas flow rate.

In the past two decades, ESA has offered potential applications in controlling VOCs emissions from diverse industrial processes.[31−33] Effectively controlling VOCs emissions, as mandated by the Kyoto Protocol, is crucial in industrial implementations. Existing methods, such as adsorption, thermal treatment, and catalytic oxidation, offer lower costs but become economically demanding for low-level organic emissions (<5000 ppm).[34] To address this concern, Air Liquide implemented an ESA process to abate the VOCs emissions from a chemical process, focusing on methylene chloride in air. The design involved cryogenic condensation, adsorption onto an activated carbon (AC) cloth, and electrothermal regeneration using ACFC for organic recovery, as shown in Figure 6.[29] The continuous process comprised two compact reactors (Figure 6a) and was configured for gas emission treatment (Figure 6b,c), containing methylene chloride ([C] in Figure 6b), aiming to reduce mass flow below 1 g/m^3.[29] The adsorbers, downstream of the cryogenic condenser, utilized homogeneous direct electrical heating by the Joule effect. Electrical current was applied after the adsorbent reached saturation to elevate the ACFC's surface temperature to approximately 60 °C, with an applied electric power of around 200 W. A relatively short cycle time of approximately 1 h enabled a compact filter design, minimizing the reactor volume (less than 100 L).

In a recent study by Zhao et al.,[30] a four-step ESA cycle was used in CO_2/N_2 separation (Figure 7), in which an electric current was supplied to the closed adsorption chamber, initiating an increase in the temperature of the adsorbent due to the Joule effect. By the end of the electrification step, the maximum cycle temperature was attained. The adsorption chamber underwent depressurization following the attainment of a higher temperature, signifying the shift in adsorption. Subsequent to CO_2 separation, the adsorption chamber's temperature was intentionally reduced to recondition the adsorbent and electrodes for the commencement of a new cycle.

In 2003, Pfister et al.[35] introduced an innovative ESA application named an electro-desorption compressor that revolved around a closed-cycle adsorption system where an adsorbate undergoes alternate adsorption onto and desorption from a adsorbent. In contrast to traditional thermally driven desorption methods, this ESA-based system employs electrical energy, particularly electromagnetic waves, to selectively dissociate the adsorbate from the adsorbent, avoiding the need for extensive heating. This approach significantly enhances the overall system efficiency by eliminating the heat-induced cooling phase.

ESA presented in the above studies offers as an aspiring avenue for harnessing electrically driven adsorptive separation processes in various fields of adsorption with promising applications. While TSA involves heating and cooling the entire adsorption matrix, ESA efficiently targets specific adsorption sites, thereby eliminating/minimizing thermal inefficiencies.[9] On the other hand, in PSA, releasing captured material relies on pressure reduction, often involving vacuum or low-pressure steam, resulting in a dilute product that requires additional separation and compression steps, unlike ESA.[36] The simplicity in the ESA design and minimal need for additional equipment, mainly a regulated power source and load, facilitate seamless integration into various processes. However, one specific challenge lies in optimizing the heating of adsorbent materials with homogeneous electrical conductivity and system design for broader applicability, which

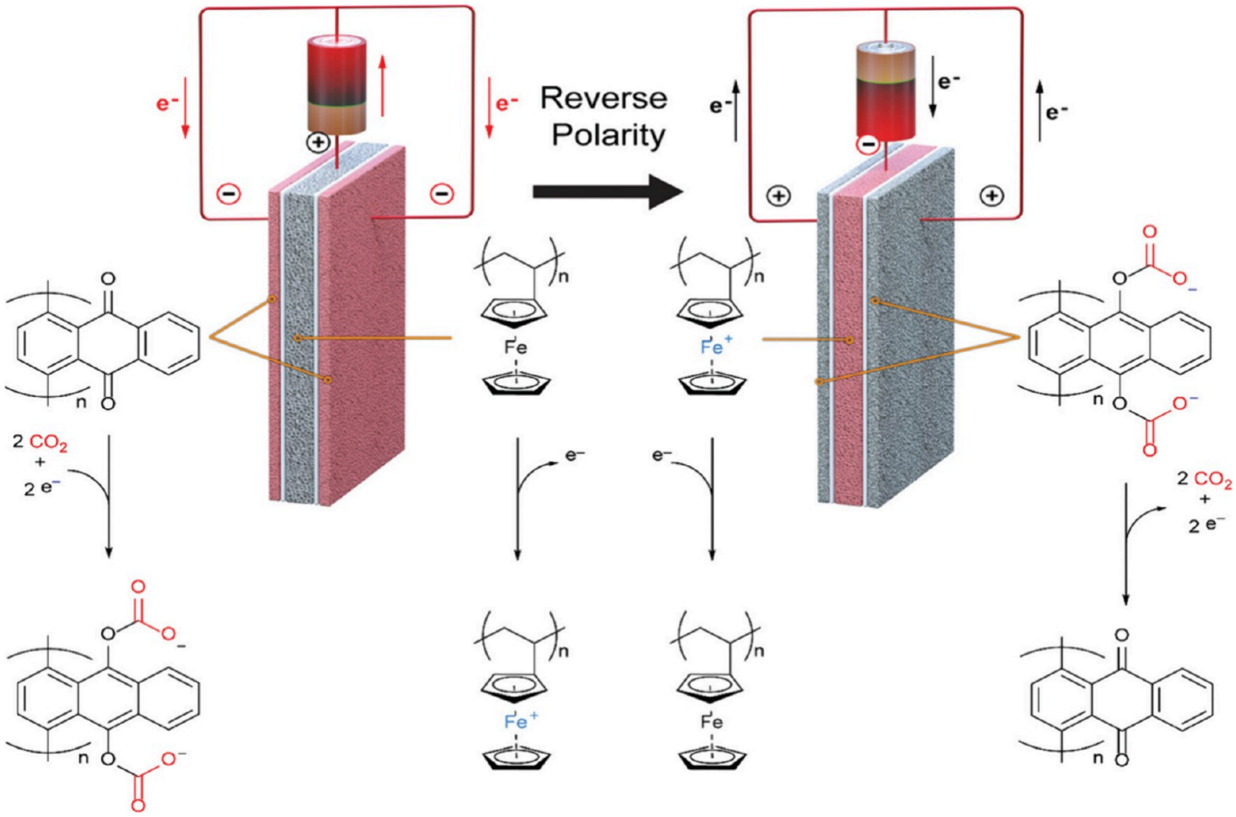

Figure 8. Electrochemical swing adsorption cell for CO_2 capture and release using P14AQ-coated porous electrodes. Image reproduced with permission from ref 9. Copyright 2019 Royal Society of Chemistry.

necessitates a high-current, low-voltage power, potentially resulting in thermal issues.[37]

Electrochemically Driven Adsorptive Separation Processes. Electrochemically driven adsorptive processes represent an innovative approach to a wide range of separation technologies. In these methods, upon application of an electric potential or current to the adsorption column, electrochemical reactions occur at the adsorbent's surface, leading to changes in its chemical and physical properties and, thereby, desorption of the adsorbate molecules and regeneration of the adsorbent. Here, the desorption efficiency is influenced by factors such as the polarity and charge of the surface and target molecules, as well as van der Waals, electrostatic, and hydration forces.[38] Additional mechanisms such as complexation, ion exchange, and microprecipitation can also play crucial roles. Compared to traditional TSA processes, electrochemically driven swing adsorption offers potential advantages:

- Electrochemical forces facilitate selective desorption of target components, enhancing separation efficiency and purity.[39]
- Precise control over adsorption/desorption kinetics.[40]
- Desorption relies on electrical potential rather than temperature changes, which implies minimum energy losses and hence low cost of energy.[9]
- Low environmental impact by reducing emissions with a smaller carbon footprint.[41]
- Suitable for both high- and low-concentration feed streams.[9]

An illustrative electrochemical swing adsorption process involves quinone-based compounds, initially reported by Mizen et al.[42] in 1989 and further developed by Hatton and colleagues in 2019, as shown in Figure 8. In this process, a single electrochemical swing adsorption cell comprising two cathode electrode substrates coated with a CO_2-binding quinone−carbon nanotube (Q-CNT) composite was developed for CO_2 capture. This sandwich structure encloses an anode electrode substrate coated with a ferrocene−CNT (Fc-CNT) composite and utilizes separator membranes situated between the electrodes. The primary objective of this cell architecture was to maximize the surface area available for CO_2 binding, which was accomplished by exposing the cell to gas flow in a parallel passage contactor, where multiple cells were arranged to form parallel gas channels. The configuration employed porous electrodes and electrolyte separators to facilitate CO_2 capture and release during the electrochemical activation and deactivation of quinones within a sealed chamber under standard conditions. Notably, the electrodes exhibited a strong affinity for CO_2 even at low concentrations (e.g., for the DAC process). Specifically, the outer electrodes, coated with a poly-1,4-anthraquinone (P14AQ) composite, played an important role in CO_2 capture during the application of a reducing potential as they helped prevent self-discharge of the cell, enhancing cycling performance with an electroconductive high surface area that increases efficient polymer exposure to the electrolyte. Conversely, when the polarity was reversed, CO_2 was released. The inner electrode, containing polyvinylferrocene (PVF), served as an electron source and sink for quinone reduction and oxidation, respectively. The discharge of the battery initiated the reverse reaction, enabling the device to contribute power while releasing pure CO_2 via the electrochemical swing process.

Most recently, SSA, which relies on the electric double-layer capacitor principle, has emerged as a new electrochemically driven swing adsorption process.[43−46] This technique offers a novel approach in achieving selective adsorption/desorption of adsorbates through charging and discharging of supercapacitive electrodes. In 2014, Landskron and colleagues introduced this concept for CO_2 capture in a gas adsorption cell, as illustrated in Figure 9.[47] This design leveraged porous carbon electrodes submerged in a NaCl electrolyte. The cell pressure dropped during charging in cyclovoltammetry experiments but increased by a similar amount in discharging, indicating reversible gas adsorption. The Fermi level of infiltrated pores is influenced through electron delocalization, affecting adsorptivity. This innovation represents a considerable departure from the more complex conventional electrochemical adsorption processes, offering promising advantages such as: (i) unlike TSA, PSA, or redox-active and pH-swing methods, the SSA operates without substantial fluctuations in pressure or temperature; (ii) it utilizes low-cost AC and neutral aqueous electrolytes, thereby eliminating corrosive or toxic chemicals; (iii) the supercapacitors feature a long lifespan (>100,000 cycles); and (iv) it has a high cycling stability with an energy efficiency exceeding 90% for charge−discharge cycles.[43,45]

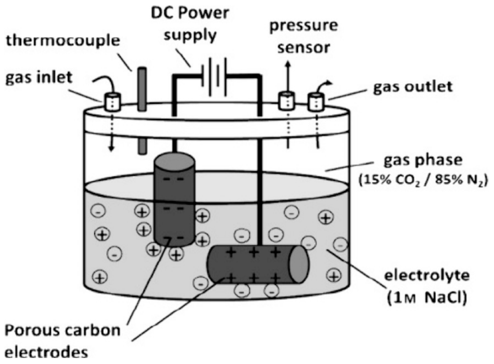

Figure 9. Experimental setup for an SSA experiment with aqueous electrolytes. Image reproduced with permission from ref 47. Copyright 2023 Wiley Online Library.

Although SSA processes appear to be appealing from an energy standpoint, they exhibit slower adsorption kinetics compared to traditional physisorption on porous carbons, potentially due to diffusion within the electrolyte. Figure 10 presents an SSA gas separation module where CO_2 adsorption reaches saturation between 15 and 100 min, similar to liquid amines or amine-impregnated solid adsorbents (20−250 min). Such slow kinetics and CO_2 adsorption may be improved by decreasing the electrode thickness and by switching to a solvent with a higher solubility for CO_2.[44] Moreover, with the continuous improvement of the modules' design, their energy consumption and adsorption capacity are also continuously improving.[45]

It is also worth noting here that although the SSA method has been shown to be effective for CO_2 capture, it has not yet been demonstrated for other separation/purification applications. A constraint of this method lies in its relatively modest/low adsorption capacity, while fundamental molecular mechanisms of SSA remain inadequately comprehended, impeding optimization efforts.[46] The adsorbed amounts in SSA are correlated with the number of electric charges in the electrodes, but variations are observed depending on the specific cation and anion combinations.[45] Quantitative analysis of the relative contributions of the different mechanisms (gas−solid, molecular liquid−solid, and ionic liquid−solid) requires further research to unravel the intricacies of adsorption and desorption. Understanding the intricate interactions among gases, anions, and the electrodes remains a challenge.

Electric Field Swing Adsorption Process. EFSA utilizes an external electric field during adsorption, which results in altering the energy state of the adsorbent and hence adsorption, while upon swtiching off the electric field, the regeneration takes place. This technique differs from electro-

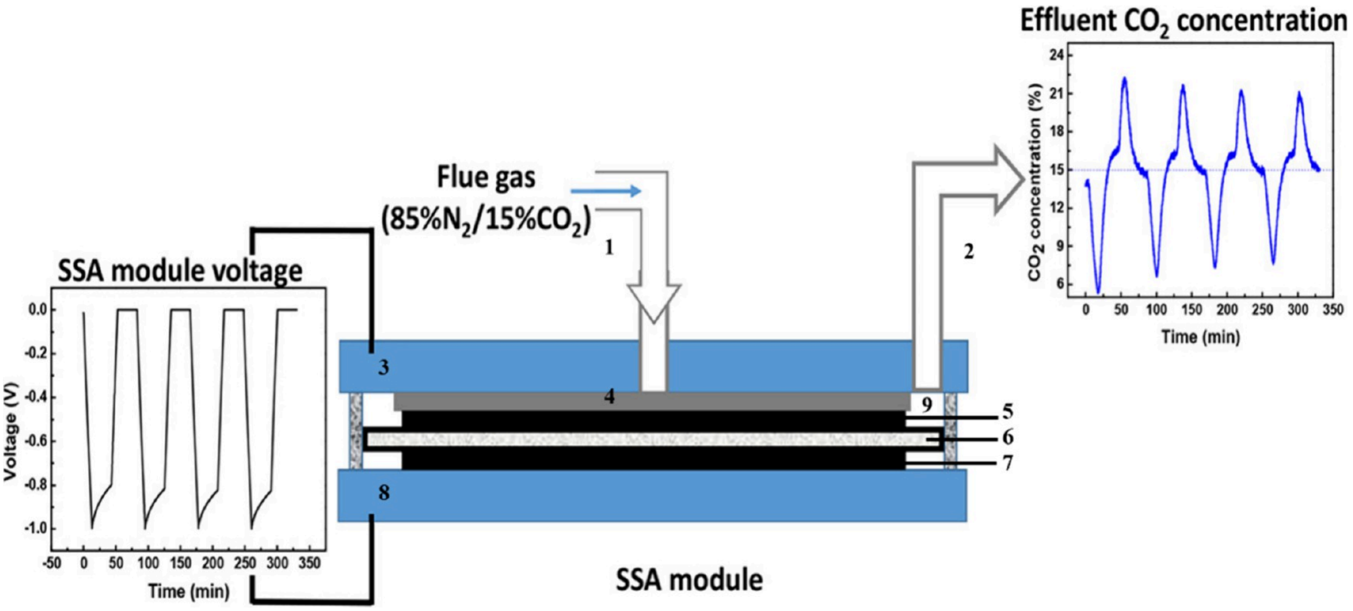

Figure 10. An SSA gas separation module: 1, gas inlet; 2, gas outlet; 3, upper plate; 4, carbon cloth; 5, negative electrode; 6, separator; 7, positive electrode; 8, bottom plate; 9, gasket. Voltage response (left) and CO_2 concentration in the effluent gas (right) of an SSA module were tested using the galvanostatic charge−discharge (GCD) method at constant current. Image reproduced with permission from ref 44. Copyright 2018 American Chemical Society.

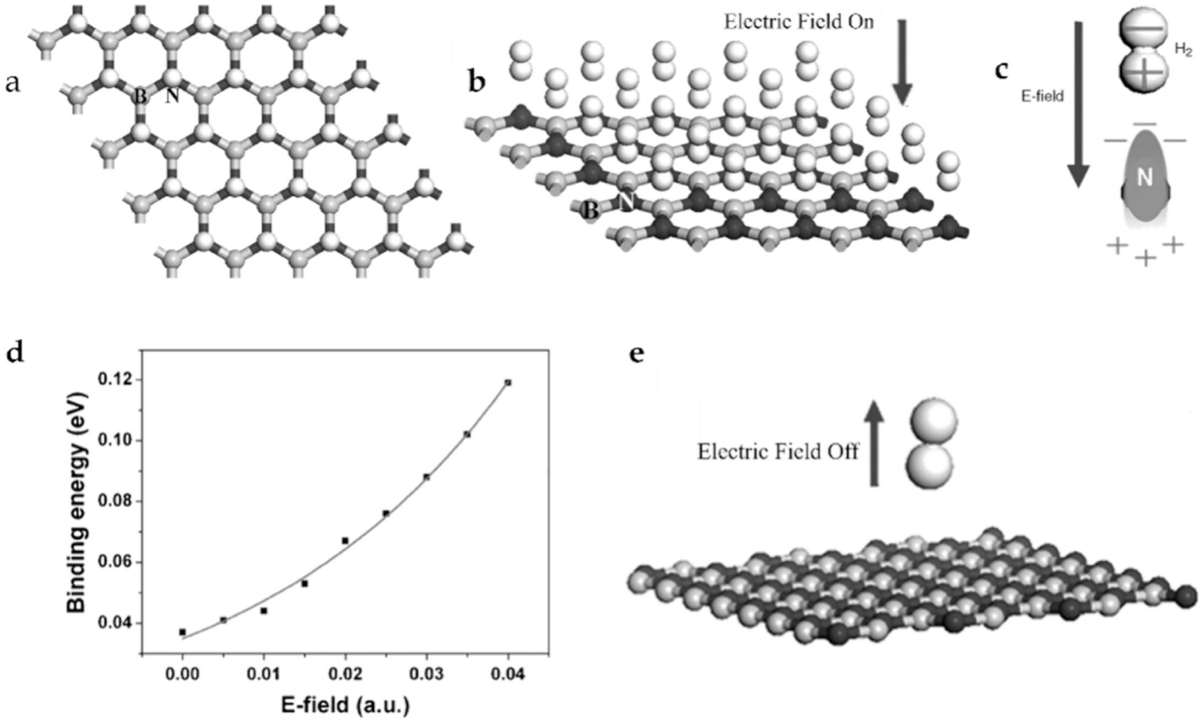

Figure 11. H_2 molecules adsorbed on a BN sheet via switching "on" external electric field: (a) top view and (b) side view; vertical electric field is applied in the *z* direction. (c) Polarized H_2 molecule and the polarized electrons at the N-site on the BN sheet. (d) Changes of binding energy with the swing of electric field. (e) H_2 molecules desorbed from the BN sheet via switching "off" the electric field. Image reproduced with permission from ref 48. Copyright 2010 National Academy of Science (U.S.).

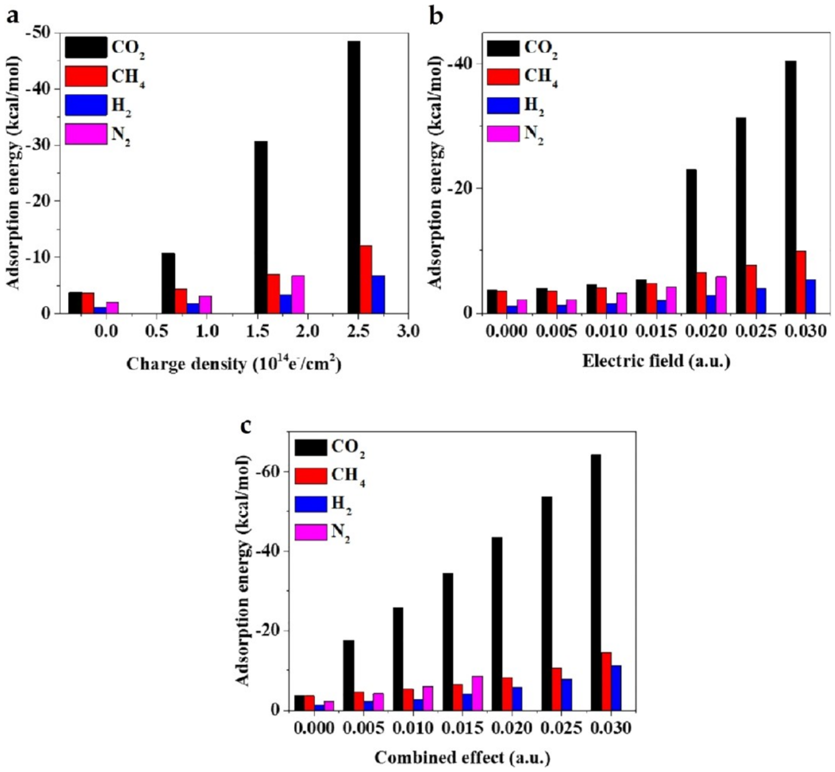

Figure 12. Adsorption energies of CO_2/CH_2/H_2/N_2 on (a) different negative charge densities from 0.91×10^{14} to 2.75×10^{14} e^-/cm^2, (b) applied external electric field from 0.005 to 0.030 au, and (c) combined effect (charge density of 0.91×10^{14} e^-/cm^2 at different electric fields) on the penta-C_2N sheet. Image reproduced with permission from ref 50. Copyright 2021 Elsevier.

thermal and electrochemical swing processes in that it enables adsorption using induced/external electric fields to change the binding energy of the material, which can induce the electrification, as shown in Figure 11, where the adsorption/

desorption of a monolayer of H_2 molecules on a boron nitride (BN) sheet occurs via switching on/off an external electric field.[48] EFSA has been suggested for achieving geometries necessary for molecular adsorption using electric fields to change the thermodynamics of the adsorbent. High-surface-area carbon (e.g., CMK, BPL) provides an electrically conductive adsorbent with large capacity for adsorption in the presence of an active electric field for the EFSA process. Adsorption sites extend throughout the networked micro-porous−mesoporous structure, and the electric field creates favored sites by pumping charge into the adsorbent.[48] Then desorption is run by removing the field, weakening chemisorption interactions between the adsorbate and the adsorbent.[48,49] The EFSA process offers several advantages relative to traditional TSA process:

- Operates at ambient conditions, eliminating the need for significant changes in temperature, pressure, or concentration.[49]
- Enables selective adsorption where desorption occurs by removing electric field, weakening the partial chemisorption between adsorbate and adsorbent.[49]
- The process is reversible and switching between adsorption/desorption is as simple as switching electric field on and off, which in turn alleviates the need to transport or heat the adsorbent.[50,51]
- Electric current used during charging is partially regenerated in discharging, which contributes to minimal parasitic load for EFSA-based gas separation.[49]

Recent advancements have introduced a pioneering approach employing charge regulation and electric fields for gas capture and storage, offering distinct advantages such as precise kinetic control and reversibility. In 2021, Sathishkumar et al.[50] theoretically explored gas adsorption on penta-C_2N (penta-graphene containing C and N atoms). They reported enhanced H_2 storage with electrochemical techniques like negative charge introduction and external electric fields. Consequently, they calculated the average H_2 adsorption energy, as shown in Figure 12, at an external electric field of 0.03 atomic unit, which increased from −1.16 kcal/mol per H_2 on the neutral penta-C_2N to −4.43 kcal/mol per H_2 for the external electric field and −7.22 kcal/mol per H_2 for the combined effect. This example clearly demonstrates the significantly improved adsorption facilitated by external electric fields.

Despite its promise, EFSA faces several specific challenges in its practical implementation. As not all materials exhibit the same degree of polarizability, the challenge necessitates the search for highly polarizable candidates.[48,50] It is necessary to apply an external force to stimulate the adsorption performance and selectivity. Studies on the effect of such novel approaches to charge regulation/electric field demonstrate that the capture-and-release processes of gases are reversible, with fast kinetics,[48] and can be regulated by switching on/off the charge state,[51] excess electrons, and electric field.[50] Though the charge-regulated/electric field gas capture and/or storage approach is advantageous, more research is required to fully understand the fundamental mechanisms pertaining to the charge injection/release processes for electric-field-regulated switchable adsorption/desorption technology. The challenge is to implement an effective way to introduce a charge into adsorbents while certain methods, such as electrospray, electron beam, or gate voltage control, are proposed to alter the charge/electric field state of adsorbent materials.[52] Future research is still needed to find more adsorbents for gas capture and storage with good electrical conductivity, high electron mobility, and a wide surface area for a sufficient number of active sites to achieve high capture capacity.

Electrically Conductive Adsorbent Materials. Electric conductivity is a vital material characteristic that measures the efficiency of electrical charge transport and depends on the direct heating temperature. It is a complex interplay between two key factors: the charge carrier concentration and the carrier mobility.[53] Precise control over these factors demands a profound understanding of the material's electronic structure and the underlying mechanisms governing charge transport and electrical conductivity.[54] The selection of suitable conductive materials in the adsorption processes is a critical and challenging task. Owing to their excellent electrical conductivity, carbon-based adsorbents are often the initial choice in electrified processes.[55] Nevertheless, diverse morphologies and compositions can dramatically influence conductive properties of these materials, which is pertinent to their applications for electrothermal regeneration. For example, in a study by Luo et al.,[56] the dependence of the electric resistivity of activated carbon in monolith (ACM), bead (ACB), and fiber cloth (ACFC) forms on the morphology of the material during electrothermal regeneration was investigated. This dependency is clearly demonstrated in Figure 13, where the ACM form exhibited the highest resistivity relative to the other two forms across the 250−500 K temperature range.

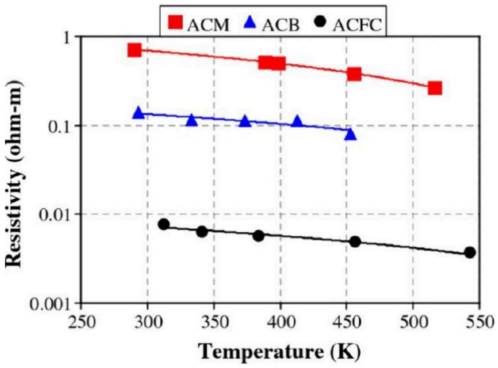

Figure 13. Electrical resistivity dependence on temperature for conductive carbonaceous adsorbents: activated carbon in monolith (ACM), bead (ACB), and fiber cloth (ACFC) forms. Image reproduced with permission from ref 56. Copyright 2006 Elsevier.

Recent years have witnessed extensive testing of various porous solids such as zeolites and amine-modified meso-macroporous resins for electrothermal adsorption. Zeolites can be efficiently regenerated by direct heating to around 430 K. One major challenge with zeolites, however, is their strong hydrophilicity, which complicates desorption. To overcome this issue, water removal is often necessary prior to direct heating via electricity.[36] In their simulation studies, Grande et al.[25] explored the capture of CO_2 using zeolite 13X-graphite composites in dry feed, and they estimated an energy consumption of 1.9 GJ/tonne of CO_2 captured with a CO_2 purity of 79.5%. They also suggested that higher CO_2 purities can be achieved by coupling the cycle with hot CO_2 as the desorbing gas via electrification. This study also suggested that, while zeolite 13X possesses high CO_2 capacity, it requires

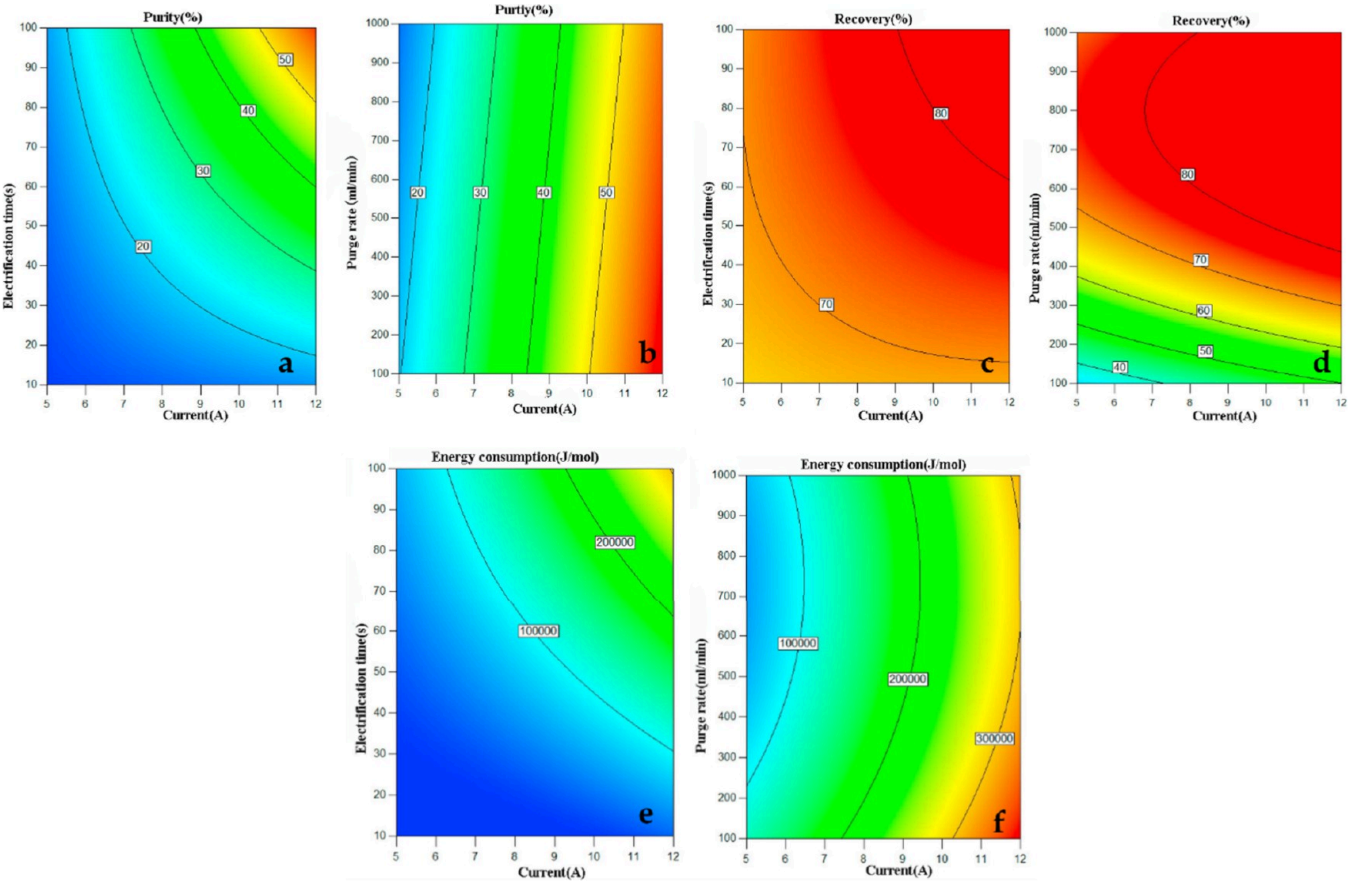

Figure 14. 2D contours of purity, recovery, and energy consumption at a fixed purge rate of 550 mL/min and fixed electrification time of 100 s for MAST carbon monolith. Image reproduced with permission from ref 60. Copyright 2017 Elsevier.

external wiring such as probes, electrodes, or other electrical components to assist in regulating and enhancing its electrical properties. Similarly, Webley's team developed a hybrid adsorbent featuring AC and zeolite NaUSY monolith, which showcased promising electrical and adsorption characteristics for ESA applications.[57]

To be suitable for use in electrically driven adsorption processes, adsorbent materials must possess high electrical conductivity, as poor and uneven conductivity can lead to unsafe hot spots.[12] Electrically conductive adsorbents can be synthesized directly or through composites of conductive/non-conductive materials. In that regard, structuring composites in the form of monoliths or hollow fibers can enable continuous electron pathways for providing uniform heating throughout the structure to avoid hot spots.[58,59] For example, Keller et al.[58] performed research on the utilization of electrically conductive CNT-zeolite and SiC-PEI hollow fibers, and reported energy requirements of 15.8 and 3.1 $J/g_{Fiber}{\cdot}K$ for these fiber bundles, respectively, when heated from ambient to 80 °C. This study highlighted the feasibility of electrical heating for CNT-zeolite and SiC-PEI fibers as a desorption strategy in a module configuration. In another study by Zhao et al.,[60] it was shown that CO_2 product purity and recovery of electro-responsive MAST carbon monolith both increase with higher electric current and electrification time, but this escalation also drives up energy consumption, as depicted in Figure 14. Specifically, it was observed that elevating the current, paired with higher N_2 flow, dilutes the CO_2 product, while lowering the N_2 flow reduces CO_2 recovery and raises the specific energy consumption. Initial trials explored the optimal performance at 12 A to achieve the desired temperature (420 K) for desorption.

In conclusion, enhancing the energy efficiency of the electrification process and optimizing electro-conductive materials' properties, striking a balance between reasonable resistance and superior adsorption, are pivotal for the commercialization of electrically driven adsorptive separation technologies. The novel adsorbent boasts significantly lower electrical resistance, potentially reducing the power consumption. However, material hybridization must consider not only the potential boost in separation efficiency but also the influence of electrical resistance on power consumption as a major challenge to address.

■ MAGNETICALLY DRIVEN ADSORPTIVE SEPARATION

In the pursuit of departure from conventional energy-intensive processes, magnetic induction swing adsorption (MISA) represents another electrification technology in the realm of adsorptive separation.[61−64] In the ESA process, the ohmic resistance within the adsorbent serves as a conduit for electricity-to-heat conversion during the desorption step. The efficient operation of the ESA hinges on the uniform distribution of electric current across the adsorbent bed, demanding a prerequisite of uniform connectivity throughout the bed. Disparities in this distribution can lead to the generation of localized hot spots, posing safety concerns and rendering the scale-up of the ESA process a formidable

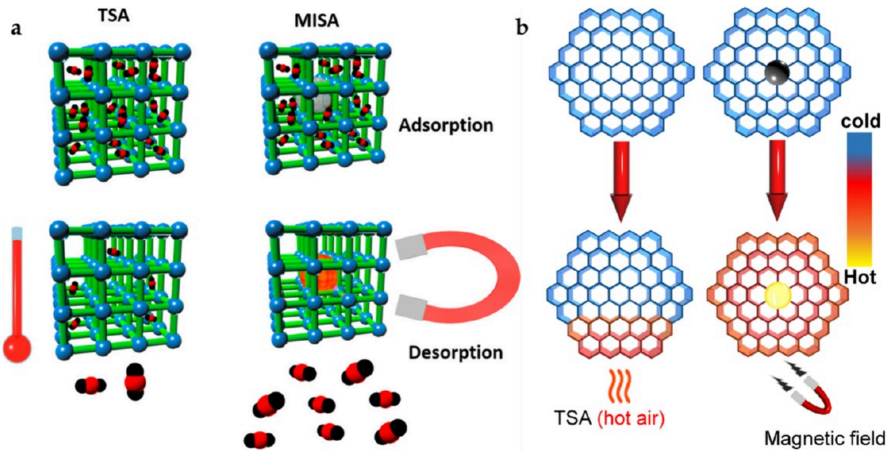

Figure 15. (a) Demonstration of the maximal regeneration capability of magnetic framework composites through MISA as compared to conventional TSA. Image reproduced with permission from ref 62. Copyright 2016 American Chemical Society. (b) Comparison of the heat-transfer mode between TSA and MISA in adsorbents. Image reproduced with permission from ref 63. Copyright 2020 Elsevier.

challenge. Unlike ESA, the adsorbent in MISA does not rely on connectivity or uniform distribution along the bed for effective heating. Instead, the induction process allows for remote heating of the adsorbent, provided the material possesses the inherent capability to be heated under magnetic induction, as demonstrated by Denayer and co-workers.[65] This unique attribute liberates the MISA process from the constraints of uniform connectivity, offering a more versatile and scalable approach. The integration of magnetic induction has emerged as a compelling approach to address the challenges associated with the regeneration and performance of adsorbent materials. By altering the magnetic field, high-frequency currents induce eddy currents, raising the bed temperature for adsorbate desorption and adsorbent regeneration.[66] As illustrated in Figure 15, MISA offers concentrated heating within the induction coil length, resulting in shorter heating times compared to conventional heating in TSA, while also resulting in full regeneration of the material.[62,63]

Magnetic field-based adsorption can be applied to various separations ranging from wastewater treatment to elimination of airborne chemicals.[67,68] Magnetic adsorbents that often contain magnetic nanoparticles (MNPs) can be employed to remove pollutants or heavy metals from contaminated water sources, in the same manner as regular adsorbents. The use of external magnetic fields induces the changes in adsorption and desorption behavior.[63]

This innovative approach aims to overcome and depart from the challenges associated with conventional thermal swing adsorption processes, such as energy consumption and temperature variations. There are advantages of this technology over conventional TSA processes:

- MISA reduces energy consumption compared to TSA, specifically to ESA, by remotely heating adsorbents through induction. Unlike ESA, it does not necessitate continuous distribution of adsorbent across the bed, allowing remote heating as long as the adsorbent exhibits susceptibility to heat under alternative magnetic induction.[62,65]
- Unlike TSA, MISA avoids issues related to temperature control and wear on adsorbents.[61]
- MISA offers fine-tuned control of magnetic field for optimal process performance, maintenance, and reversibility.

Development of MISA Processes. Development of MISA was marked by pioneering efforts that sought to harness the potential of induction heating for adsorbent regeneration. One of the early milestones in this journey was reached in 1994, when desorption of VOCs from AC through induction heating was demonstrated by Mocho et al.[68] This seminal study unveiled a critical insight: incorporating a susceptor via ligand functionalization that converts electromagnetic energy into heat, such as graphite, within the adsorbent greatly enhances the efficiency of the heating process by induction. In 2016, Hill and co-workers demonstrated a MISA process for CO_2 capture over $MgFe_2O_4$/UiO-66 composites.[62] The authors reported rapid adsorbent regeneration in just 4 min with 100% CO_2 release using induced magnetic field heating. As shown in Figure 16, these magnetic framework composites (MFCs) exhibited an instantaneous release of adsorbed CO_2 when periodically irradiated with an external magnetic field, whereas the bare UiO-66 (control) did not yield any desorption upon exposure to the magnetic field.

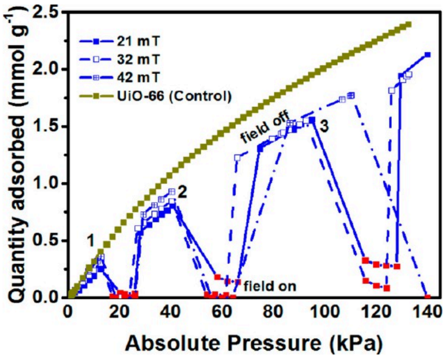

Figure 16. Dynamic uptake and release of CO_2 from the $MgFe_2O_4$/UiO-66 composites. Image reproduced with permission from ref 62. Copyright 2016 American Chemical Society.

In another MISA process over $MgFe_2O_4$-Mg-MOF-74 composite (M-74 CPT), the electromagnetic induction heating method yielded a specific regeneration energy of 1.29 MJ/kg$_{CO_2}$, signifying the remarkable energy efficiency of this process.[63] Similarly, dynamic triggered release experiments over this composite at 21 mT and 0.15 kPa conducted over 10

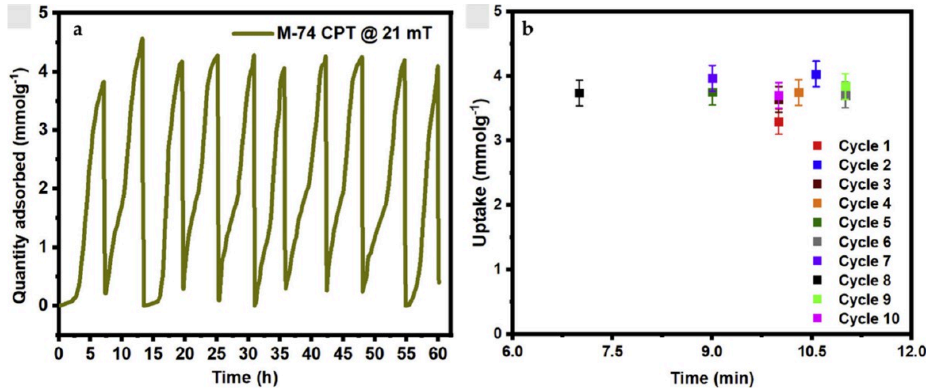

Figure 17. (a) Dynamic CO_2 uptake and release from M-74 CPT at 21 mT (145 °C) and 0.15 kPa, and (b) temporal evolution CO_2 desorption in M-74 CPT. Image reproduced with permission from ref 63. Copyright 2020 Elsevier.

cycles (Figure 17a) yielded an average CO_2 release efficiency of 85% per cycle. The induction heating's remote and localized nature, ensuring even heat distribution within the composite, resulted in a notably low regeneration time of 7−11 min across the 10 cycles (Figure 17b).[63]

In a recent study by Lin et al.,[64] the incorporation of superparamagnetic particles of Fe_3O_4 as susceptors into N-doped porous carbon was demonstrated for CO_2 capture. Their experiments revealed the potential of induction heating, achieving a remarkable energy efficiency of 79.2% with an energy consumption of 3.43 MJ/kg_{CO_2}. When the authors compared two electromagnetic induction heating modes at the regeneration temperature of 140 °C, it was found that the desorption rate was 4.02 mg/g·s in the fixed target temperature mode, significantly faster than the rate of 2.21 mg/g·s in the constant current mode at 10 A for induction heating (Figure 18).

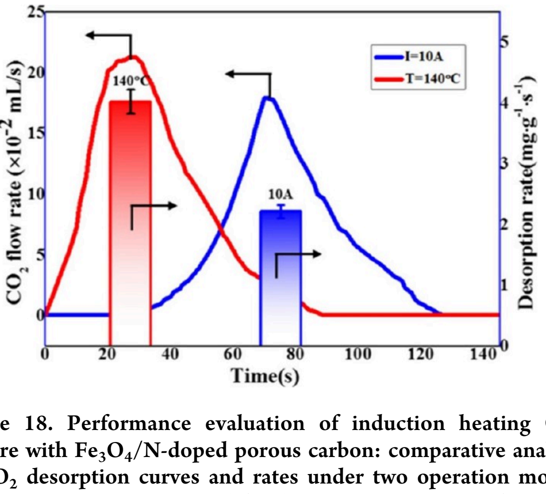

Figure 18. Performance evaluation of induction heating CO_2 capture with Fe_3O_4/N-doped porous carbon: comparative analysis of CO_2 desorption curves and rates under two operation modes, constant current at 10 A and fixed target temperature of 140 °C. Image reproduced with permission from ref 64. Copyright 2020 American Chemical Society.

Most recently, Baamran et al.[61] demonstrated the dynamic adsorption/desorption experiments utilizing the MISA principle, manifesting that magnetic field as an external stimulus can overcome the setbacks of energy utilization. This study presents the development of innovative magnetic sorbents, featuring a core−shell structure composed of MOF-74 crystals and superparamagnetic Fe_2O_3 particles, and evaluates their performance in the separation of C_2H_4/C_2H_6. By varying the loading of Fe_2O_3 (Fe_x) from 1 to 20 wt%, the electromagnetic properties of these materials were finely tuned, allowing systematic investigations of their impact on adsorption capacity, selectivity, and desorption rates which are typically governed by magnetic dissipation or ohmic resistance. As the Fe_x content increased, the surface area, microporosity, and C_2H_4/C_2H_6 selectivity decreased, while the specific heat absorption rate increased by 60−80% with varying magnetic field intensities from 12.6 to 31.4 mT. Notably, induction heating exhibited a significantly higher cooling rate of approximately 76% compared to conventional thermal heating where heat is generated by dynamic core losses and static hysteresis of ferrimagnetic particles induced by an external current magnetic field that makes the heating zone concentrated within the induction coil length, thereby making the cooling step much shorter, reducing the column size, and enhancing system throughput. This research builds upon prior studies demonstrating the potential of novel stimuli-responsive adsorbents for the next generation of olefin/paraffin separation systems.

Ensuring uniform heating throughout the bed is a crucial challenge in MISA to achieve a consistent performance. Besides, while increasing magnetic field strength can enhance desorption rates, increasing the loading at constant magnetic field strength can result in a reduced desorption rate.[61−64] Design and development of electro-magnetic field responsive adsorbent materials are necessary to critically address the challenges and trade-offs in order to maximize the process efficiency governed by such processes.

Magnetic Field-Responsive Adsorbent Materials. Magnetically responsive porous materials exhibit exceptional characteristics in adsorption processes, including magnetically induced separation and heat generation within alternating magnetic fields. These unique properties greatly facilitate recycling procedures, promote long-term operational efficiency, and enhance desorption rates, thereby significantly improving the overall effectiveness of conventional adsorption techniques. As interest in magnetic adsorbents continues to grow, substantial advancements have been achieved in the design and comprehension of magnetically responsive porous materials. These materials span a wide range of sizes, from monolithic structures to nanoscale particles, and are employed in diverse adsorption applications. MFCs represent an emerging class of materials, as magnetic field-responsive adsorbents that harness the combined attributes of magnetic

Table 1. Development and Application of MFCs in MISA Processes[a]

Metal	Magnetic Component	Ligand	Application	Refs
–	Fe_3O_4	CTF	Pollution control	69
Cu(II)	Fe_3O_4	H_3btc	Drug delivery	70
Al(III), Cu(II)	Fe_3O_4	H_3btc, H_2ndc	Catalyst separation, thermal therapy	71
Cu(II)	$Fe_3O_4@SiO_2$	H_3btc	Separation	72
Zn(II)	C-coated Co	H_2bdc, H_2Abdc	Positioning/sensing	73
Zn(II)	Fe_3O_4	Bix	Fluorescence probe	74
Zn (II), Cr(III)	$Fe_3O_4@SiO_2$, $CoFe_2O_4$, $NiFe_2O_4$	H_2bdc	PAH sequestration	75, 76
Zn(II)	Fe_3O_4	2-MI	Microreactor catalysis	77

[a] Abbreviations: CTF, covalent triazine-based framework (carbonaceous polymeric material); H_3btc, 1,3,5-benzenetricarboxylic acid; H_2ndc, 2,6-naphthalenedicarboxylic acid; H_2bdc, terephthalic acid; H_2Abdc, 2-aminoterephthalic acid; Bix, 1,4-bis(1-imidazolyl)benzene; 2-MI, 2-methylimidazole; PAH, polycyclic aromatic hydrocarbon.

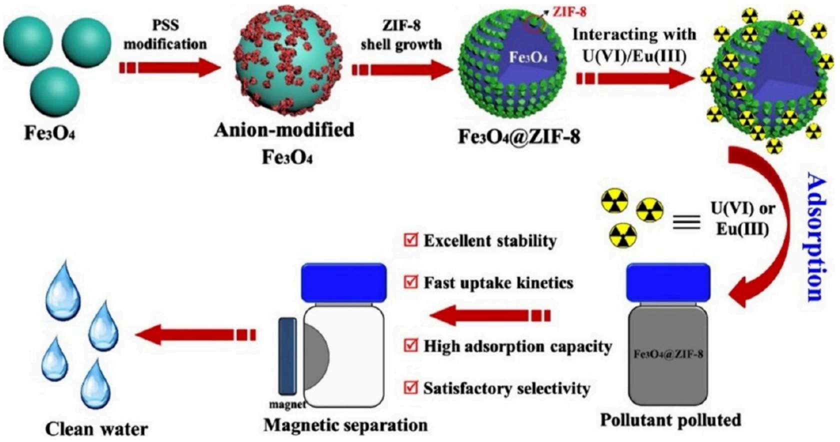

Figure 19. Fabrication of water-stable Fe_3O_4@ZIF-8 to treat radioactive wastewater via electromagnetic adsorption. Image reproduced with permission from ref 78. Copyright 2019 Elsevier.

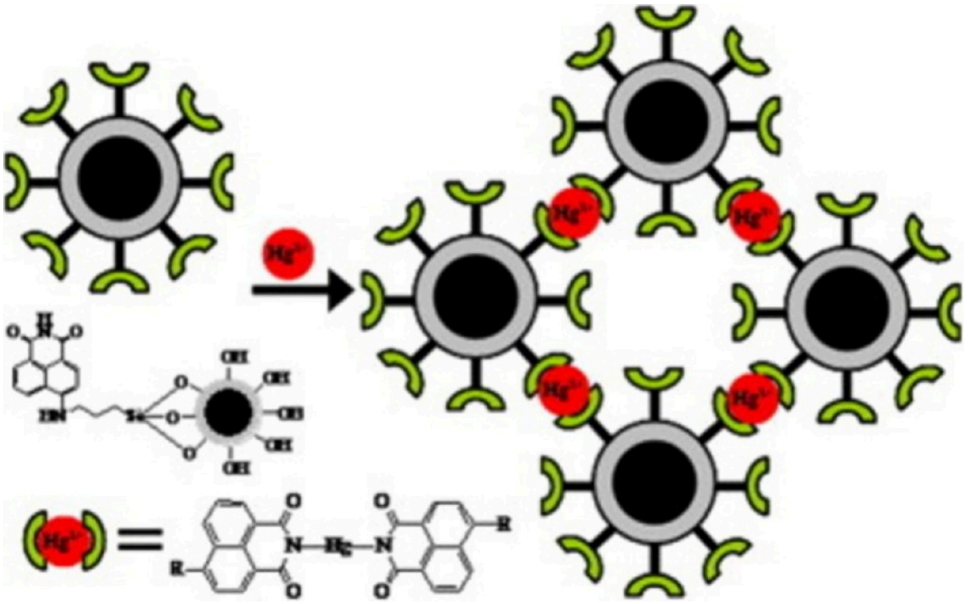

Figure 20. Rationale for extraction of Hg^{2+} over $Fe_3O_4@SiO_2$ composites. Image reproduced with permission from ref 79. Copyright 2013 Elsevier.

properties from metal or metal oxides and the functional properties of MOFs. These materials hold particular significance in applications requiring both a high surface area, a characteristic inherent to MOFs, and a facile recovery of the material, facilitated by the incorporation of magnetic particles. The versatile nature of MFCs has led to their utilization in diverse technological domains, including but not limited to drug delivery, imaging, environmental remediation, catalysis, microfluidics, sensing, and separation processes. Their multifaceted potential underscores their growing importance in a variety of fields. Table 1 shows the elements

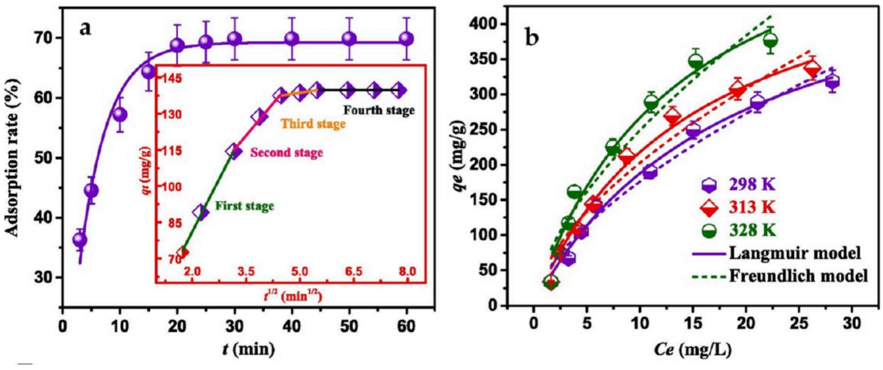

Figure 21. (a) Adsorption kinetics and inset image of intraparticle diffusion model and (b) adsorption isotherms of Fe_3O_4@ZIF-8 toward U(VI) at various temperatures. Image reproduced with permission from ref 78. Copyright 2019 Elsevier.

in development and the most significant applications of MFCs with their specific functional groups.

Iron oxides, in particular, have garnered significant attention due to their robust magnetic responsiveness and compatibility with electrification and modification within different adsorptive separation systems. Furthermore, the evolution of materials synthesis techniques has facilitated the creation of magnetic core−shell structures featuring iron oxide cores enveloped by functional outer shells made of polymers, silica, zeolites, MOFs, and carbon. For example, a water-stable core−shell microsphere (Fe_3O_4@ZIF-8) with robust magnetism was successfully synthesized (Figure 19) using a straightforward modification approach. The process involved pretreating the Fe_3O_4 core with an anionic polyelectrolyte to impart a negative charge to the Fe_3O_4 particle surface. These microspheres exhibited a large specific surface area (606.9 m^2/g) and an abundance of functional groups.[78]

In 2013, Zhu et al.[79] developed novel magnetic fluorescent nanoparticles (MFNPs) with core-shell structure composed of 1,8-naphthalimide-functionalized Fe_3O_4@SiO_2 (Figure 20). These MFNPs were designed for the simultaneous detection and adsorption of Hg^{2+}. Extensive adsorption studies were conducted, varying Hg^{2+} concentrations, temperature, time, and pH. The results indicated a high maximum adsorption capacity exceeding 30 mg/g over a wide temperature range (0, 25, 50 °C) and within the pH range of 4−10. Notably, the MFNPs demonstrated excellent reusability and displayed a high adsorption selectivity for Hg^{2+}. The propensity of MFNPs to aggregate in Hg^{2+}-containing aqueous solutions facilitated their separation, either by settling or by utilizing an external magnetic field, streamlining the removal of Hg^{2+}.

In a study by Wu et al.,[78] factors influencing the removal of radionuclides by Fe_3O_4@ZIF-8 were comprehensively investigated, such as contact time, temperature, pH, ionic strength, and co-existing ions were comprehensively investigated, and as depicted in Figure 21, the results demonstrated relatively fast adsorption kinetics (reaching equilibrium in approximately 30 min), excellent uptake capacities, remarkable selectivity, and favorable physicochemical stability. These findings indicate that Fe_3O_4@ZIF-8 can simultaneously offer stability and functionality. Moreover, its easy separation using an external magnetic field after use in the treatment of radioactive wastewater makes it highly practical for extracting radioactive waste, even in challenging conditions.

The integration of magnetic nanoparticles within MOFs has led to the development of a remarkable class of materials with a diverse applications. For example, $MnFe_2O_4$@UiO-66 aided in wastewater treatment by selectively adsorbing toxic heavy metal ions and facilitating their separation using a magnetic field.[80] Further examples include Fe_3O_4@GO in targeted drug delivery systems, which effectively released drugs at desired locations in the body with the assistance of an external magnetic field,[81] or MNPs@HKUST-1 was utilized for adsorption/degradation of a dye pollutant, Indigo Carmine, and pathogens from aqueous solutions.[82]

While these materials offer diverse opportunities for separation and purification applications, their syntheses can be complex, involving multiple steps, and they may have limitations in terms of surface area and adsorption capacity. Future developments in simplifying syntheses and improving adsorption capacities will further advance the field of electromagnetic field-responsive adsorbent materials, catering to a wider range of applications in separation and purification.[83]

CHALLENGES IN ELECTRIFICATION OF ADSORPTION PROCESSES

The electrification of adsorptive separation presents formidable challenges encompassing intricate design complexities, advancements in adsorbent materials, and techno-economic considerations concerning energy consumption and environmental pollution. In addition, the demanding research and development surrounding adsorbent material design, the impact of desorption kinetics, and the influence of crystallinity on material conductivity face significant hurdles to overcome. Challenges are further compounded by the constraints posed by electro-magnetic properties, obstacles in electrode design, and managing the heat release during the electrification process. Additionally, ensuring the compatibility and integration of electrified adsorptive separation technologies with existing infrastructure and energy systems is an alarming challenge. Therefore, the key hurdle lies in finding efficient and cost-effective methods for providing and managing the required electrical energy and ensuring that the benefits of reduced energy consumption can be realized effectively at both small and large scales. Below, we highlight some of the most daunting challenges currently facing the electrified adsorptive separation field.

Combined Energy−Environment−Economy Challenges. Electrification of adsorptive separation processes requires a significant amount of electrical energy to operate and conduct the process efficiently, which eventually increases the cost of the process and perhaps leads to environmental concerns related to increased carbon emissions if the energy is generated from non-renewable sources. Efficiently addressing

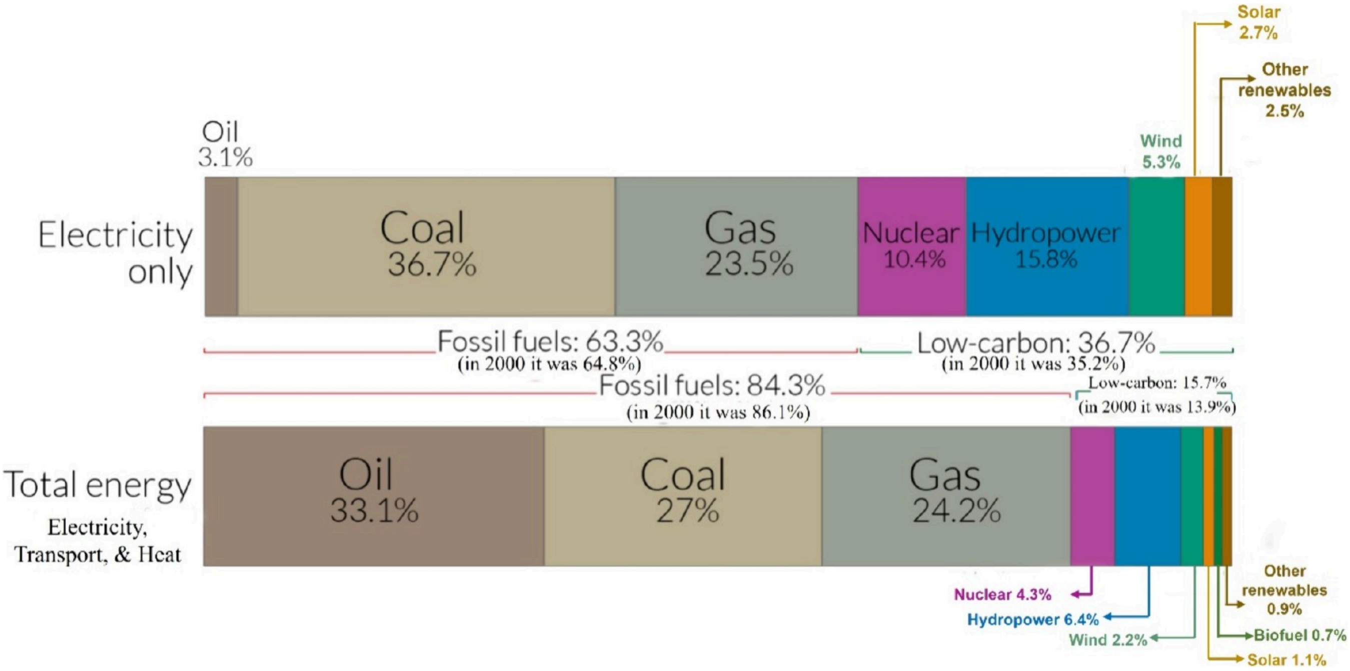

Figure 22. Sources of global electrical energy. Image reproduced with permission from ref 84. Published by Our World in Data under a Creative Commons CCBY4.0 license (http://creativecommons.org/licenses/by/4.0).

these challenges and formulating viable solutions are imperative for the effective deployment of electrified adsorptive separation processes, thereby advancing the prospects of a sustainable and decarbonized future. Researchers globally have been actively engaged in the exploration of diverse technological avenues aimed at decarbonizing national economies, where electrification of adsorptive separation techniques can play a major role. BP's Statistical Review of World Energy in 2020[84] showed that more than one-third of global electricity comes from low-carbon sources, but a much smaller fraction of total energy does, as depicted in Figure 22, which presents a combined challenge for the electrification of adsorptive separation with respect to energy requirements, environmental pollution, and economic feasibility. Various processes often require the use of electricity to power equipment such as pumps, compressors, or heaters, and thus the sustainability gets reduced conditionally during carbon emission if the electricity is generated from fossil fuels.[84] Therefore, addressing energy demands, environmental impact, and economic feasibility has become imperative to strike a balance among them concerning the current reliance on nonrenewable energy sources that underscores the dire need for a sustainable shift toward renewables.

Challenges Associated with Material Design. The effective design and implementation of adsorbents for practical applications face significant challenges within the realm of electrification in adsorption and separation processes. This complexity arises from the need to address various stimulus-responsive properties with electricity to enhance the separation efficiency. Despite the continual progress in the adsorptive separation methodologies, numerous challenges persist in the context of electrification. The selection and design of appropriate adsorbent materials in such processes hold paramount importance, presenting perhaps the most intricate challenge for design engineers. Addressing this challenge is essential to ensure the scalability and commercial viability of electrification-based adsorption and separation processes.

The development of electro-responsive adsorbents presents several challenges that researchers and engineers need to overcome. Finding materials that exhibit the desired response to electrical stimuli while maintaining adsorption capacity and selectivity, and stability can be complex. Ensuring good electrical contact and stability of the electrode materials within the adsorbent matrix can be technically demanding. Developing cost-effective and scalable synthesis methods that maintain the desired electrical properties such as conductivity or resistivity of the adsorbent is crucial for commercialization. Ensuring the long-term durability of electro-responsive adsorbents is critical for their practical application. Maintaining the electro-responsive properties over extended usage cycles, under complex operating conditions and in the presence of feed contaminants, is also a challenge that needs to be addressed. Moreover, integrating electro-responsive adsorbents into existing systems or designing new systems to leverage their capabilities can pose challenges in balancing the material inventory and capital costs, manufacturing systems, and overall performance of the processes.

Challenges Associated with Desorption Kinetics. Slow kinetics usually hamper the widespread adoption of electrified adsorptive separation processes, especially electrochemical-based systems, as diffusion of adsorbate molecules through electrolytes (typically ionic liquids) is a rate-limiting step. Moreover, the non-uniformity and uneven heating during electrification provide poor local heat dissipation and poor heat transfer to the adsorbents for the endothermic regeneration which eventually produce local "hot" or "cold" spots, preventing fast desorption kinetics.[85,86] For example, Voskian and Hatton[9] conducted an analysis of electrochemical kinetics and efficiencies, emphasizing the potential for reducing energy demands in regeneration processes during desorption through advanced designs. However, the challenges persist in slow

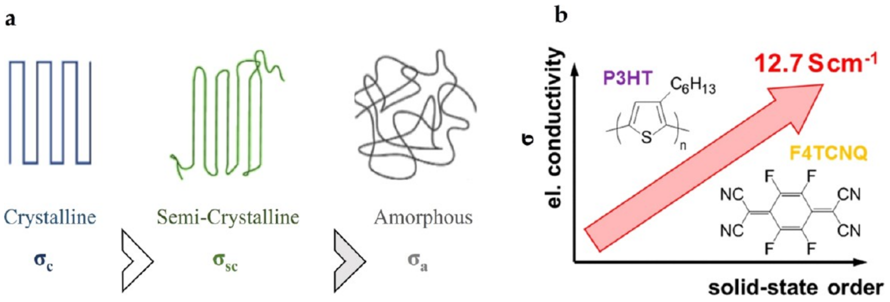

Figure 23. (a) Comparative depiction of changes in solid-state order with respect to electrical conductivity (σ), where σ_c, σ_{sc}, and σ_a denote the electrical conductivity of crystalline, semicrystalline, and amorphous materials, and (b) electrical conductivity of conjugated polymer poly(3-hexylthiophene) (P3HT) with respect to order of crystallinity as solid-state order. Image reproduced with permission from ref 89. Copyright 2017 American Chemical Society.

sorption kinetics concerning structural integrity maintenance in terms of electrification and conductivity relative to material degradation over repeated cycles.

Indeed, achieving a balance between rapid electrical response and fast desorption kinetics is necessary to attain a practical and effective separation performance. Desorption kinetics determine the speed at which molecules are released from the adsorbent material, while electro-magnetic response time refers to the time required to induce the desired electrical effect. Finding materials and operating conditions that optimize both aspects simultaneously is the main challenge in this regard. The material should possess the ability to undergo rapid electrical changes and exhibit fast sorption kinetics. In this regard, adsorbents with low electrical conductivity may impede efficient electrical response time and result in slow kinetics. Enhancing the electrical conductivity of the adsorbent material without sacrificing its sorption properties can be technically demanding. The efficiency of the charge-transfer processes between the adsorbent and the electrical source can affect both desorption kinetics and electrical response time.

Impact of Crystallinity on Electrical Conductivity. Crystalline materials, including MOFs, covalent organic frameworks (COFs), and porous organic cages (POCs), offer great opportunities to create a novel electric-responsive performance. These materials have a well-defined and regular molecular structure that facilitates energy transfer between external-stimulus signals and responsive sites, making their response faster and more efficient. Additionally, their well-defined structures enable investigation and understanding of the mechanism behind the stimuli-responsive behaviors and structure−property relationships. However, there are still challenges in developing practical applications of crystalline smart materials (CSMs) concerning the electricity responsiveness. As their crystallinity increases, their processability and mechanical properties usually decrease and vice versa, thereby hindering their practical application, which means the sustenance of mechanical properties decreases crystallinity. Additionally, CSMs mostly exist as micro-/nanosized powders, making it difficult to make them stimuli-responsive on the macroscale utilizing the efficacy of electrification in terms of conductivity.[87] The realm of CSMs, such as MOFs and COFs, offers a unique avenue for responsive performance under electrical stimuli.[87,88] Their well-defined structures facilitate swift and efficient energy transfer, aiding rapid responses to external signals. However, as their crystallinity increases, practical applications encounter hurdles, limiting their mechanical adaptability. Additionally, the micro-/nanoscale nature of CSMs poses challenges in achieving macroscale responsiveness to electrical stimuli, impacting their conductivity, as illustrated in Figure 23.

The figure underscores the pivotal role of solid-state order in enhancing conductivity, controlled by processing solvent and regioregularity choices. Nevertheless, not all stimulus-responsive adsorbents share this property, posing challenges in the application of electrical fields effectively. Therefore, optimizing properties and structure concerning electrification remains a crucial challenge in designing adsorbent materials for effective adsorptive separation processes.[89]

Constraints of Electro-magnetic Properties. Designing materials for electrified adsorptive separation encounters challenges related to poor electro-magnetic conductivity, hindering the responsiveness of sorbents to applied electric, magnetic, or electro-magnetic fields. Efforts to enhance electrothermal heating through hybridizing adsorbent crystals and magnetic particles are ongoing, targeting improved local thermal effects. Achieving magnetic susceptibility in stimulus-responsive adsorbents remains challenging due to inherent material limitations. However, the increase in temperature by direct heating during electrification adversely affects the susceptibility and magnetic moment of the adsorbents. Thus, magnetic interference complicates the use of magnetic fields in the separation process, potentially affecting the behavior of particles and the efficiency of the separation. The graph in Figure 24 illustrates the relationship between magnetic moment and susceptibility concerning temperature variations

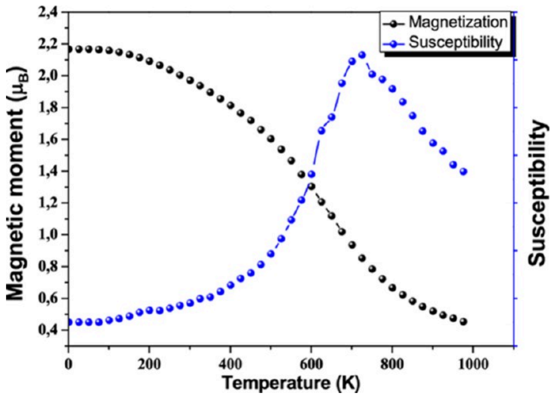

Figure 24. Magnetic moment and susceptibility as a function of temperature. Image reproduced with permission from ref 90. Copyright 2020 Springer Nature.

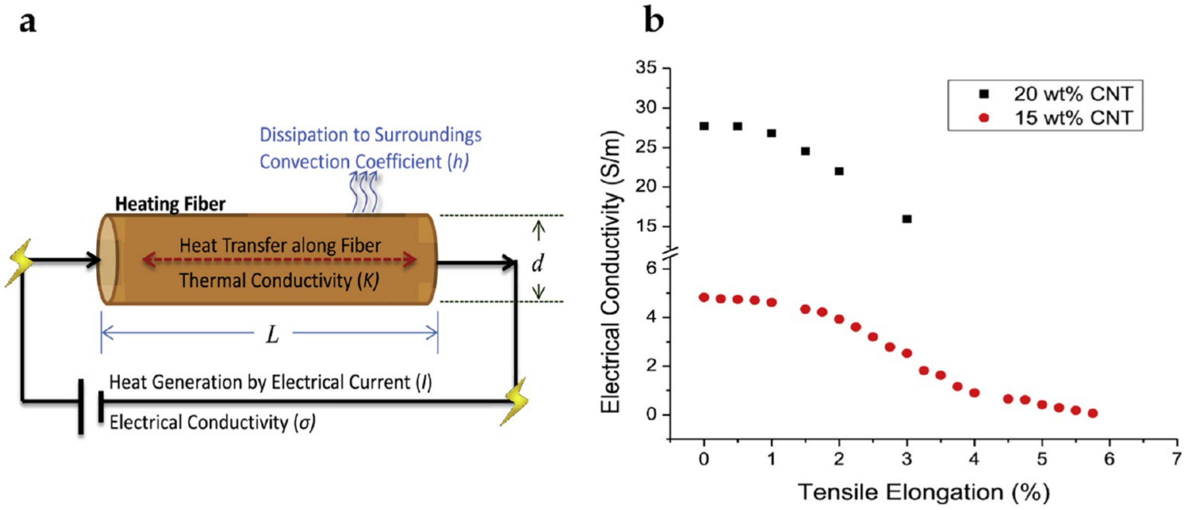

Figure 25. (a) Joule heating phenomenon and (b) the effect of direct heat stretching on electrical conductivity as a mechanical property. Image reproduced with permission from ref 91. Copyright 2014 Elsevier.

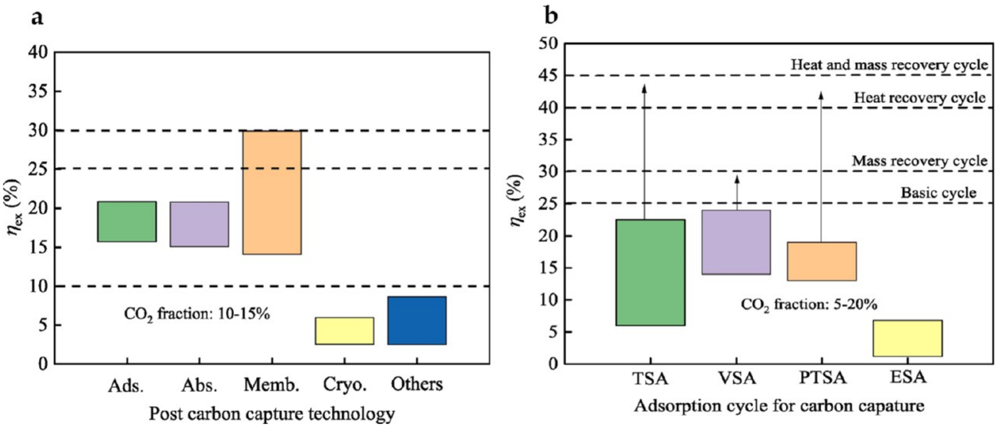

Figure 26. (a) Exergy efficiencies of various post-combustion carbon capture and storage (CCS) technologies and (b) exergy efficiencies of the selected adsorption cycles in terms of different recovery technologies. Image utilized with permission from ref 92. Published 2020 by Elsevier under a Creative Commons CCBY4.0 license (http://creativecommons.org/licenses/by/4.0).

due to direct Joule heating, highlighting the optimal susceptibility achieved amidst decreasing magnetic moment and showing that the degree of magnetization of adsorbent material gets optimized in response to the applied electromagnetic field for adsorptive separation.[90]

Hurdles of Heat Release during Electrification. Heat loss to the surroundings during electrification of adsorptive separation should be carefully managed, as it can lead to a reduction in the electrical conductivity of the elements of the system, which can in turn affect the overall efficiency of the separation process. This is mainly because the electrical conductivity of the system is directly related to the amount of electric current that can be passed through it and the amount of heat it loses. When heat loss occurs, the system may require more energy to maintain the desired level of electrical conductivity or resistivity.[91] For example, as shown in Figure 25, when composite fibers made of polymer/CNT were stretched by heat during the annealing process, a decrease in electrical conductivity was observed due to a breakdown in the CNT network. The reduction in conductivity was a result of the orientation of CNTs within the polymer matrix during the stretching process as a mechanical property. As the alignment of CNTs increased, the probability of inter-tube connection decreased, which impaired the ability of the composite to transfer electrons and thus led to lower conductivity and inefficient separation.[91]

The requirement of excess energy to maintain the desired level of conductivity during the electrification process can lead to elevated consumption of energy with higher expense for operations. When heat is generated as a byproduct, it must be removed to maintain optimal operating conditions. This requires additional energy input to cool the system, which can further increases energy consumption. Besides, heat can cause the adsorbent material to degrade and lose the degree of electrical conductivity needed for an efficient separation.

Challenges in Enhancing Exergy Efficiency. The challenges inherent in enhancing the exergy efficiency of electrified adsorptive separation processes manifest in varying ways across different technologies. For instance, while technologies such as TSA and PTSA demonstrate potential in heat recovery by leveraging internal heat recovery (IHR) mechanisms for desorption enhancement, others such as VSA predominantly rely on internal mass recovery (IMR) to bolster their performance. In contrast, electrified adsorption, for example, the ESA method, presents a unique challenge in that its primary focus on expediting desorption poses

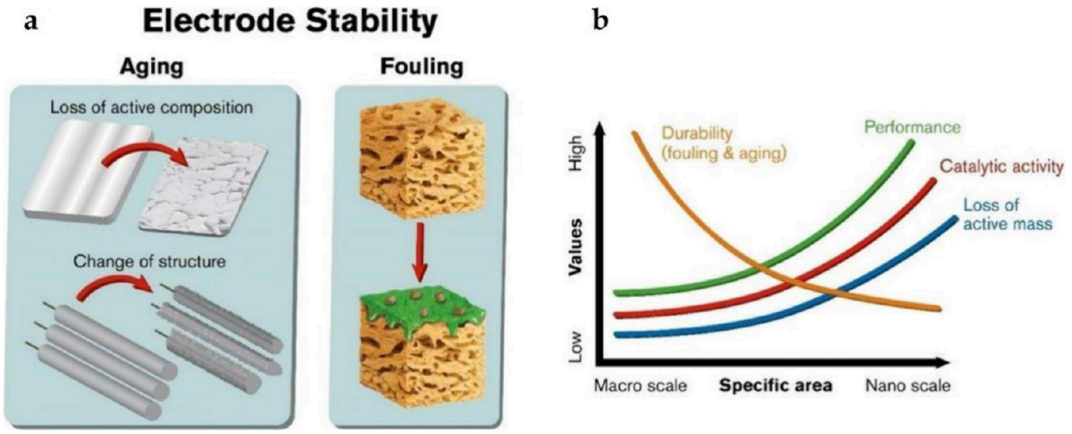

Figure 27. (a) Structural and surface impacts of aging and fouling on electrode stability and (b) impact of aging and fouling on electrode durability and long-term performance as a function of specific area transition from macroscale to nanoscale. Image reproduced with permission from ref 93. Copyright 2020 Royal Society of Chemistry.

limitations in seamlessly integrating or benefiting from specific recovery technologies tailored to optimize overall process efficiency. These intricate challenges underscore the complexity inherent in the pursuit of enhancing exergy efficiency within electrified processes, which becomes apparent in Figure 26, where the adsorption cycle for carbon capture shows that efficiency for ESA is significantly low, often below or near 5% without IHR or IMR.[92]

Deterrent of Electrode Design. Over time, the efficacy of electrochemical adsorptive systems experiences a decline, as depicted in Figure 27, primarily due to fouling and aging of the electrodes. This effect becomes more pronounced, especially concerning nano-enabled electrodes with extensive surface areas within confinement. Adsorbent's performance, evaluated based on selectivity, significantly relies on the electrode's lifespan, which dictates its overall productivity. However, aging and fouling of the electrodes diminish this lifespan by deactivating their surfaces, thereby reducing the productivity and performance of adsorbents. Fouling, encompassing the accumulation of unwanted materials like inorganic scales or magnetic particles on electrode surfaces during electromagnetization, notably hampers their electrical properties. It isolates electrodes from the solution and elevates the electrical resistance, decreasing the electrical conductivity of the system, subsequently elevating costs and compromising performance.[93]

On the other hand, aging denotes detrimental alterations in the structure and composition of electrodes surfaces during operation, further deteriorating their electrical properties. Aging results in irreversible electrode damage, necessitating electrode replacement and sacrificing structural integrity.[93] Comprehensive research aimed at understanding the mechanisms underlying these issues, coupled with the development of effective mitigation strategies, holds the key to advancing the reliability and longevity of the systems. In our shift toward a more sustainable energy landscape, traditional separation methods, which heavily rely on energy-intensive thermal changes or fossil fuel sources, encounter substantial challenges. Electrification in adsorption processes holds great potential to reduce the need for existing thermal energy, but it also brings about challenges with burgeoning opportunities. Challenges persist in the adsorption domain, from understanding factors affecting conductivity−capacity trade-offs and desorption kinetics to grasping temporal material changes across cycles and implementing effective scale-up strategies. Electrification faces complexities, from materials design to swing processes development, with techno-economic considerations for industrial process implementation. To fully embrace adsorptive electrification for separation processes, we need to continue to address the daunting challenges inherent to both adsorbents and adsorption systems. Electrification offers the potential to revolutionize adsorption processes, providing energy efficiency and sustainability, but addressing these challenges is imperative as we navigate toward a greener energy future.

> Overall, challenges persist in electrification of adsorption processes, from understanding factors affecting conductivity−capacity trade-offs and desorption kinetics to grasping temporal material changes across cycles and implementing effective scale-up strategies.

■ AUTHOR INFORMATION

Corresponding Authors

Ali A. Rownaghi − *National Energy Technology Laboratory (NETL), United States Department of Energy, Pittsburgh, Pennsylvania 15236, United States;* orcid.org/0000-0001-5228-5624; Email: ali.rownaghi@netl.doe.gov

Fateme Rezaei − *Department of Chemical, Environmental and Materials Engineering, University of Miami, Miami, Florida 33124, United States;* orcid.org/0000-0002-4214-4235; Email: rezaeif@miami.edu

Author

Abdullah Al Moinee − *Linda and Bipin Doshi Department of Chemical and Biochemical Engineering, Missouri University of Science and Technology, Rolla, Missouri 65409-1230, United States*

Complete contact information is available at:
https://pubs.acs.org/10.1021/acsenergylett.3c02340

Notes

The authors declare no competing financial interest.

Biographies

Abdullah Al Moinee earned his B.Sc. in Chemical Engineering from Bangladesh University of Engineering and Technology in 2017, followed by an M.Sc. in 2020. He is currently pursuing a Ph.D. in Chemical Engineering at Missouri S&T, where his research focuses on CO_2 capture, adsorption, electrification in separation processes, and process system engineering.

Ali A. Rownaghi, a Scientist at U.S. Department of Energy (NETL), earned his Ph.D. in Catalysis from University Putra Malaysia and worked as postdoctoral fellow at Georgia Tech. His research focuses on research involves creating, understanding, and coupling advanced materials chemistry work with process engineering for chemical, biochemical, energy, and environmental applications.

Fateme Rezaei, a Professor of Chemical Engineering at the University of Miami, earned her Ph.D. in Chemical Engineering from Monash University and Luleå University of Technology. She is an Associate Editor of *Energy & Fuels*. Her research focuses on advanced materials and processes for separation, purification, and storage applications.

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