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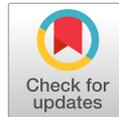
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Efficient separation of acetylene and carbon dioxide in a decorated zeolite

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Dedicated to the 100th anniversary of Chemistry at Nankai University

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Abstract: The almost identical molecular sizes and volatilities of acetylene and carbon dioxide make their separation extremely challenging in industry. Here, we report the efficient separation of acetylene and carbon dioxide ($v/v=2/1$, which is relevant to that in the industrial cracking stream) in faujasite zeolites decorated with atomically-dispersed copper(II) sites under ambient conditions. *In situ* neutron powder diffraction and inelastic neutron scattering confirm that the confined copper(II) site displays chemo-selective yet reversible binding to acetylene, whereas adsorbed carbon dioxide molecules are established by weak host-guest supramolecular interactions with the framework oxygen centres, thus resulting in the efficient separation of these two gases under flow conditions. A designed adsorption-purging-desorption system based upon Cu@FAU is established for the recovery of high purity acetylene (98–99%) from the mixture of acetylene and carbon dioxide, offering an unprecedented separation factor of 22.2 with an effective dynamic uptake of acetylene of 1.51 mmol g⁻¹ at 298 K.

Introduction

Acetylene (C₂H₂) has a wide range of industrial applications, such as metal cutting and instrumental analysis, and more importantly, it is an essential building block for the manufacture of numerous materials and fine chemicals, such as synthetic fibres, rubber, acetic acid and benzene.^[1] C₂H₂ is mainly produced by petroleum cracking and partial combustion of methane, where carbon dioxide (CO₂) as byproduct can reach a concentration up to ~50% of that of C₂H₂ in the gas stream, causing serious problems to the downstream application of C₂H₂. Therefore, the removal of CO₂ from the C₂H₂ stream is of vital importance to obtain high-purity C₂H₂ for further applications. State-of-the-art techniques to separate C₂H₂ and CO₂ are based upon extraction of C₂H₂ by organic solvents (e.g., dimethylformamide and acetone) and cryogenic distillation, which are among the most energy-intensive industrial separation processes with significant environmental impacts.^[2] The

development of energy-efficient strategies for the separation of C₂H₂/CO₂ remains an important and challenging line of research owing to their similar physical properties (Supporting Information, Table S1), for example the same molecular shape, almost identical sizes (C₂H₂, 3.3 × 3.3 × 5.7 Å³; CO₂, 3.2 × 3.3 × 5.4 Å³) and very close boiling points (C₂H₂, 189.3 K; CO₂, 194.7 K).^[3]

Adsorptive separation based upon porous sorbents affords increasing promise for gas separation at (near) ambient conditions and is attracting much interest from both academia and industry.^[4–7] Various porous materials, such as activated carbons,^[8] silica^[9] and metal-organic frameworks (MOFs),^[3,10–19] have been studied for the adsorptive separation of C₂H₂/CO₂. MOFs attracted particular attention because of their abundant structural diversity and design flexibility.^[20–24] Zeolites, as conventional sorbent materials,^[25,26] are potentially superior to MOFs for gas separations at an industrial scale owing to their high hydrothermal stability, recyclability and low production cost. However, the separation of mixtures of C₂H₂/CO₂ in zeolites has not been achieved to date because physisorption of C₂H₂ and CO₂ within pristine zeolites often exhibit similar uptakes and adsorption profiles, leading to no apparent selectivity to afford any practical separation.^[27]

We have recently developed a Ni(II)-decorated zeolite material, which shows an excellent performance in the purification of lower olefins by efficient binding and removal of alkyne impurities.^[28] The introduced Ni(II) sites have enabled chemoselective and reversible binding to alkynes through the formation of metastable [Ni(II)(alkyne)₃] complexes. This creates an exciting avenue to explore the application of decorated zeolites in challenging industrial separations. Here, we report, to the best of our knowledge, the first example of recovery of high-purity C₂H₂ via efficient separation of the mixture of C₂H₂/CO₂ in decorated zeolites. By the incorporation of coordinatively unsaturated Cu(II) sites in FAU zeolites, Cu@FAU exhibits high dynamic adsorption capacity of C₂H₂ and excellent separation of the mixture of C₂H₂/CO₂ at ambient conditions. The physisorbed CO₂ molecules on Cu@FAU can be readily removed by purging

at ambient temperature, while most bound C_2H_2 molecules are preserved. As a result, C_2H_2 with purity of 98–99% can be obtained by the regeneration of sorbents *via* conventional temperature-swing desorption. The underlying adsorption mechanisms of C_2H_2 and CO_2 in Cu@FAU have been fully elucidated by *in situ* neutron powder diffraction (NPD) and inelastic neutron scattering (INS), which revealed an unusual geometry for bound C_2H_2 molecules on the Cu(II) sites. This study represents an appealing strategy of using decorated zeolites to enable facile production of high-purity C_2H_2 from the mixture of C_2H_2/CO_2 through chemoselective adsorption of C_2H_2 .

Results and Discussion

Synthesis and characterisation. M@FAU zeolites [M=Ni(II), Cu(II) and Zn(II)] were synthesised *via* hydrothermal reactions with a bifunctional organic ligand {N-[3-(trimethoxysilyl)propyl]ethylenediamine} for the introduction of metal ions into the desired location within the zeolite matrix, which is difficult to access *via* post-synthetic ion-exchange. Coordinatively unsaturated cations at the specific positions of FAU zeolite structure can be achieved upon the removal of organic ligands by calcination.^[28] The full retention of the framework structure of FAU zeolites and location of incorporated metal ions within the M@FAU zeolites have been confirmed by Rietveld refinements of high-resolution synchrotron X-ray powder diffraction.^[28] Similar approaches based upon a (3-mercaptopropyl)trimethoxysilane ligand have been reported to deliver metal sites to specific locations in various zeolites to fabricate functional catalysts.^[29] The phase purity and porosity of desolvated M@FAU materials are confirmed by powder X-ray diffraction data and Ar adsorption isotherms, respectively (Supporting Information, Figure S1 and S2).

Analysis of gas adsorption and separation properties. Adsorption isotherms of C_2H_2 and CO_2 on M@FAU (M=Ni, Cu, Zn, Figure 1a, Supporting Information, Figure S3 and S4) and the parent Na-FAU zeolites (Figure 1b) were collected at 298 K. All samples exhibited comparable adsorption capacity for C_2H_2 and CO_2 at 1.0 bar, e.g., 3.55 and 3.88 mmol g^{-1} , respectively, for Cu@FAU. The uptakes of C_2H_2 and CO_2 of M@FAU samples are slightly lower than that of the pristine Na-FAU zeolite (4.63 and 5.32 mmol g^{-1} , respectively) due to the reduction of BET surface area on incorporation of the metal sites (Supporting Information, Figure S2 and Table S2). It is worth noting that typical contents of C_2H_2 and CO_2 in the cracking stream for the production of C_2H_2 are 7.5–8.8% and 3.2–3.5%, respectively,^[1] and importantly, these decorated zeolites show significantly higher adsorption of C_2H_2 than CO_2 under the corresponding partial pressures (e.g., 2.50 and 1.32 mmol g^{-1} , respectively, for Cu@FAU at 0.07 and 0.04 bar; Supporting Information, Figure S5 and S6), affording great promise to explore their performance in the separation of C_2H_2/CO_2 . Although ideal adsorbed solution theory (IAST) is widely employed to evaluate the adsorption selectivity for various materials,^[30] the assumption of this theory is not valid in this case due to the presence of chemoselective adsorption of one component in the competitive adsorption (see below) and we therefore determined directly the selectivity of C_2H_2/CO_2 separation by column breakthrough experiments.

Breakthrough separations of the mixture of $C_2H_2/CO_2/He$ (10/5/85; v/v/v) by M@FAU (Figure 1c, Supporting Information, Figure S7 and S8) and Na-FAU (Figure 1d) were conducted at 298 K. The dynamic adsorption capacities of C_2H_2 for Cu@FAU, Ni@FAU, Zn@FAU and Na-FAU were measured as 2.29, 2.23, 1.68 and 2.05 mmol g^{-1} , respectively, and the dynamic selectivities of C_2H_2/CO_2 are 1.71, 1.62, 1.41 and 1.63, respectively. The separation performance of Cu@FAU zeolite is comparable to that of the leading sorbent materials, such as FJU-90a,^[10] NKMOF-1-Ni,^[14] HOF-3a,^[17] JCM-1^[18] and SNNU-45,^[19] but the selectivity is lower than that of the best-behaving MOFs incorporating narrow pores or open metal sites, namely UTSA-300a,^[2] UTSA-74a,^[12] SIFSIX-3-Ni,^[15] and TIFSIX-2-Cu-*i*^[15] (Supporting Information, Table S3). It is worth noting that in a breakthrough experiment where co-adsorption occurs as observed in most reported studies, upon regeneration of the saturated sorbent, pure component gas cannot be obtained directly. For example, although Cu@FAU exhibits a decent dynamic selectivity of C_2H_2/CO_2 , the purity of C_2H_2 can only reach ~80% upon desorption of the sorbent bed, restricting its practical applications (Figure 1c).

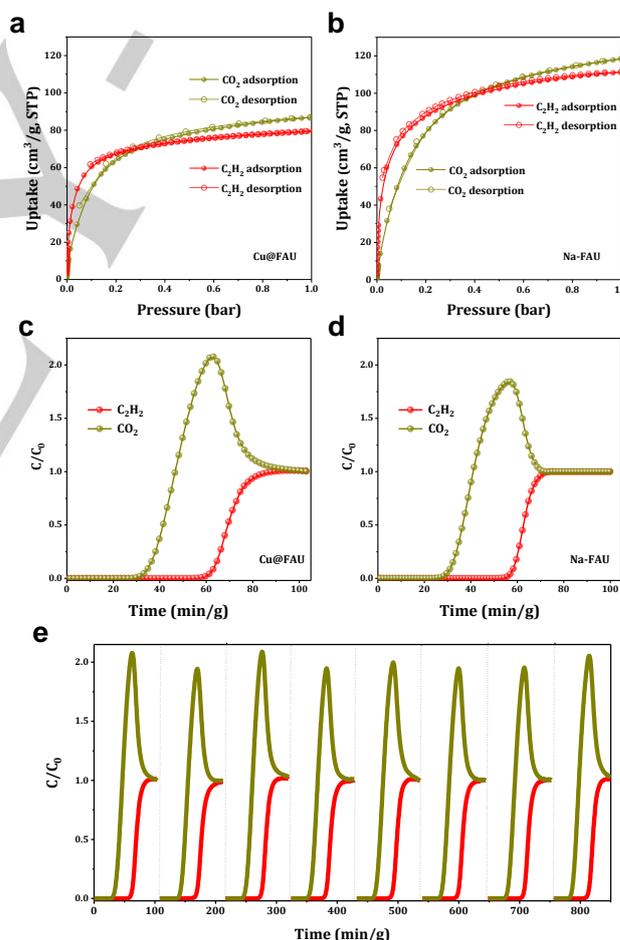


Figure 1. View of the adsorption and separation data of Cu@FAU and Na-FAU. Data for other zeolite materials are shown in the Supporting Information. Adsorption isotherms of C_2H_2 and CO_2 of Cu@FAU (a) and Na-FAU (b) at 298 K; Column breakthrough curves of the mixture of $C_2H_2/CO_2/He$ at 298 K and 1 bar (10/5/85, v/v/v; total gas flow: 8.0 mL min^{-1} , sample weight: 0.5 g) over a fixed-bed packed with Cu@FAU (c) and Na-FAU (d). (e) The recyclability of Cu@FAU for the separation of C_2H_2/CO_2 over eight cycles (C_2H_2 : red; CO_2 : dark yellow). The saturated sorbent was regenerated by heating at 473 K in a flow of Ar for 20 minutes between cycles.

Establishment of an adsorption-purging-desorption system.

To seek potential solutions, we further investigate the adsorption affinity between these two gas molecules and the sorbent. The isosteric heats of adsorption (Q_{st}) of C_2H_2 and CO_2 within these zeolites have been determined by differential scanning calorimetry (DSC) measurements (Supporting Information, Figure S9-S16).^[31,32] Na-FAU exhibits similar Q_{st} values for the uptakes of C_2H_2 and CO_2 (24.9 and 25.9 kJ mol⁻¹, respectively). In contrast, the Q_{st} of C_2H_2 adsorption in Cu@FAU is almost twice that of CO_2 (50.0 and 28.2 kJ mol⁻¹, respectively), indicating the significantly stronger binding affinity of C_2H_2 than CO_2 in Cu@FAU. Thus, desorption of CO_2 from Cu@FAU occurs under milder conditions than that would require for the desorption of C_2H_2 , affording a valid opportunity to increase the concentration of C_2H_2 within Cu@FAU by desorbing CO_2 at a mild condition, under which C_2H_2 can be largely retained in the pores.

To validate this hypothesis, temperature-programmed desorption (TPD) experiments were conducted. In a typical experiment, the zeolite sample was firstly dosed with the gas mixture of C_2H_2/CO_2 (v/v = 2/1) at 298 K and the effect of selective desorption by purging the saturated sample with a flow of Ar at 298 K was investigated. The TPD profiles of CO_2 (m/z = 44) and C_2H_2 (m/z = 26) were recorded at a heating rate of 10 K min⁻¹ from 298 to 473 K under a flow of Ar. The comparison of the TPD profiles of the purged and unpurged Cu@FAU samples (Figure 2c, d) shows that the majority (>95%) of adsorbed CO_2

molecules were desorbed upon purging, while only 34% of the adsorbed C_2H_2 molecules were desorbed at the same time (Figure 2a, b). Importantly, Cu@FAU retains the highest amount of acetylene (1.51 mmol g⁻¹) compared with Ni@FAU, Zn@FAU and Na-FAU (0.75, 0.27 and 0.55 mmol g⁻¹, respectively) after this purging process. This suggests that the proportion of C_2H_2 in the adsorbed phase within Cu@FAU is significantly increased after the selective desorption and high-purity C_2H_2 could be obtained on the complete desorption. Indeed, the concentrations of acetylene in the final desorbed gas streams are 97.8, 90.2, 95.4 and 94.4 % for Cu@FAU, Zn@FAU, Ni@FAU, and Na-FAU, respectively. Thus, the three-stage process based upon “adsorption-purging-desorption” has effectively increased the separation factor (α) of C_2H_2/CO_2 from 1.71 to 22.2 for Cu@FAU, which is ~3 times of Na-FAU (from 1.63 to 8.40; Figure 2e), demonstrating the key role of confined Cu(II) sites in enhancing the separation performance of FAU zeolites. It worth noting that the introduction of Cu(II) sites in FAU zeolites *via* post-synthesis ion-exchange (denoted as Cu-FAU) or wet impregnation (denoted as CuO/FAU) are poorly effective to promote the separation of C_2H_2/CO_2 (Supporting Information, Figure S23-S26) because the location of Cu(II) sites in Cu-FAU and CuO/FAU can undergo dynamic changes under different conditions and is often poorly accessible by the guest molecules.^[33,34] Thus, this result demonstrates the importance of location of confined Cu(II) sites introduced by the direct hydrothermal synthesis.

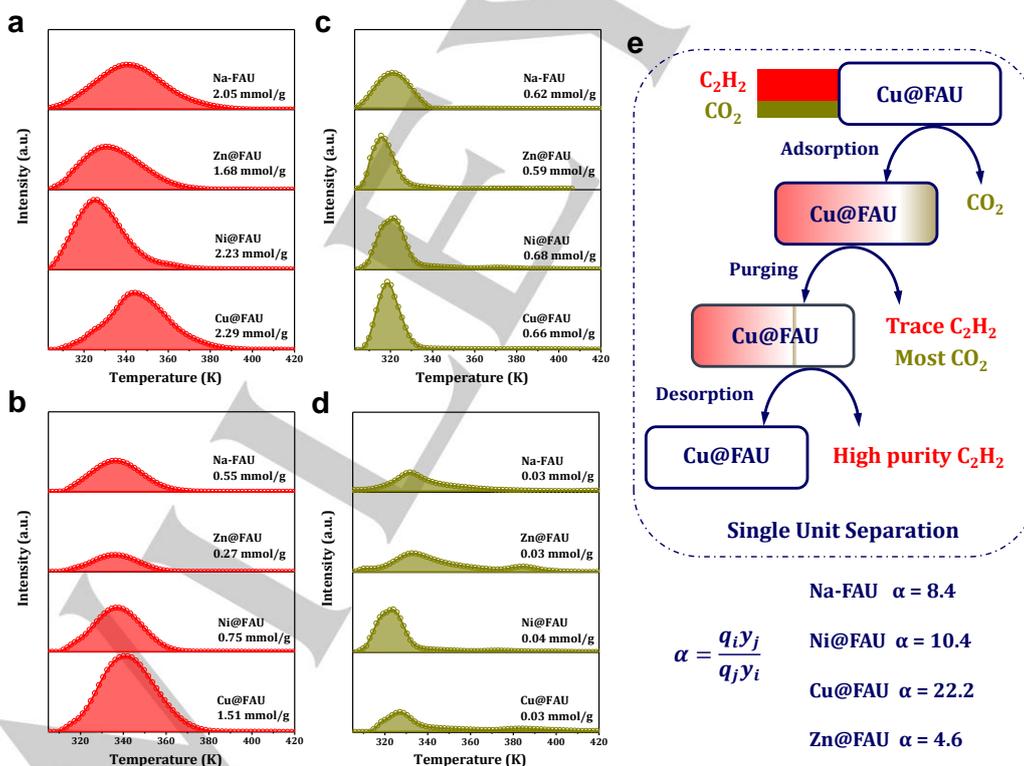


Figure 2. Views of the TPD profiles of gas-loaded M@FAU (M=Cu, Zn, Ni) and Na-FAU samples as function of temperature. TPD profiles of adsorbed C_2H_2 released from the unpurged samples (a) and from samples after purging by Ar at 298 K (b); TPD profiles of adsorbed CO_2 released from the unpurged samples (c) and from samples after purging by Ar at 298 K (d). (e) A schematic view of the designed adsorption-purging-desorption process for the recovery of high-purity C_2H_2 *via* separation of the mixture of C_2H_2/CO_2 by decorated FAU zeolites.

The effect of operating conditions on the separation of C_2H_2/CO_2 was investigated. The adsorption capacity of C_2H_2 for Cu@FAU decreases slightly with increasing flow rate of gas mixtures from 15 to 20 mL min⁻¹ or with rising temperature from

298 to 323 K (Supporting Information, Figure S27-S30). Upon dilution of C_2H_2 and CO_2 by a factor of 5 in the gas stream (*i.e.*, 10% to 2% and 5% to 1%, respectively), Na-FAU exhibited a decrease in the dynamic adsorption capacity of C_2H_2 (from 2.05

to 1.24 mmol g⁻¹) but no change in the selectivity (Supporting Information, Figure S31), whereas for Cu@FAU, although the former decreased from 2.29 to 1.68 mmol g⁻¹, the selectivity of C₂H₂ increased from 1.71 to 2.35 (Supporting Information, Figure S32). Endowed with the high structural stability and operational flexibility, Cu@FAU zeolites can function in a modular manner. For example, with a single separation unit based upon Cu@FAU, 1.51 mmol g⁻¹ C₂H₂ with the purity of 97.8 % can be obtained from an industrially relevant mixture of C₂H₂/CO₂ (v/v=2/1), and C₂H₂ can be produced with even higher purity by using multiple separation units (Figure 2e). For example, an acetylene stream with purity >99 % can be obtained with use of a second unit, demonstrating its promising potential in practical applications.

Determination of binding sites of C₂D₂ and CO₂ within Cu@FAU. The binding sites of adsorbed C₂D₂ and CO₂ molecules within the pore of Cu@FAU were determined by *in situ* NPD. Fourier difference map analysis of the desolvated Cu@FAU [Cu₁₂Na₂₀(Al₄₄Si₁₄₈O₃₈₄)] confirmed its structural integrity and the absence of residual nuclear density in the supercage. Cu(II) was found to be stabilised by framework oxygen centres of the six-membered ring of the sodalite cage. Upon gas loading, variations in Bragg peak intensities were observed, and the binding domains of gases were successfully interpreted by Fourier difference map analysis and Rietveld refinements (Figure 3). In [Cu₁₂Na₂₀(Al₄₄Si₁₄₈O₃₈₄)]·(C₂D₂)₃₀, two distinct C₂D₂ sites were identified with Site I showing strong interactions with Cu(II) centres and accounting for 63% of adsorbed C₂D₂ molecules. Unlike classic side-on interactions between transition metal and C≡C bond in cation-acetylene complexation^[35] and that observed in C₂D₂-loaded Ni@FAU^[28], C₂D₂(I) is noticeably tilted with C...Cu distances being 2.49 and 3.35 Å, and ∠C≡C...Cu of 126.0° (Figure 3a-c, Supporting Information, Figure S33). This is originated from the presence of additional strong hydrogen bonds between adsorbed C₂D₂ molecules and the oxygen centres of the framework (D_{C₂D₂}...O_{framework}=1.89 Å). *In situ* X-ray photoelectron spectroscopy confirmed the retention of the a divalent oxidation state of Cu(II) sites on acetylene binding (Supporting Information, Figure S34). C₂D₂(II) locates away from Cu(II) centres and is solely stabilised by weak host-guest hydrogen bond (D_{C₂D₂}...O_{framework}=2.64 Å). C₂D₂(II) accounts for 37% of the total adsorbed C₂D₂ molecules, in excellent agreement with the loss of C₂H₂ uptake (34%) on purging observed in the adsorption-purging process. Thus, this confirms that the combination of Cu(II) sites and strong hydrogen bonds results in the retention of C₂H₂ in Cu@FAU upon purging. Interestingly, the result is distinct to that observed C₂D₂-loaded Ni@FAU^[28], where all C₂D₂ molecules were located at a single site showing a side-on binding to the Ni(II) sites.

In comparison, only one binding site was found for adsorbed CO₂ molecules in [Cu₁₂Na₂₀(Al₄₄Si₁₄₈O₃₈₄)]·(CO₂)₁₈, which exhibits notably weaker host-guest interactions (C_{CO₂}...O_{framework}=3.08 Å; O_{CO₂}...Cu=3.18 Å; Figure 3d-f, Supporting Information, Figure S35). The above studies showed that coordinatively unsaturated Cu(II) sites confined in Cu@FAU provided unique binding sites with significantly stronger adsorption affinity to C₂H₂ than to CO₂. This leads to its superior performance in the separation of C₂H₂/CO₂ and the recovery of high-purity C₂H₂ from the designed *adsorption-purging-desorption* process.

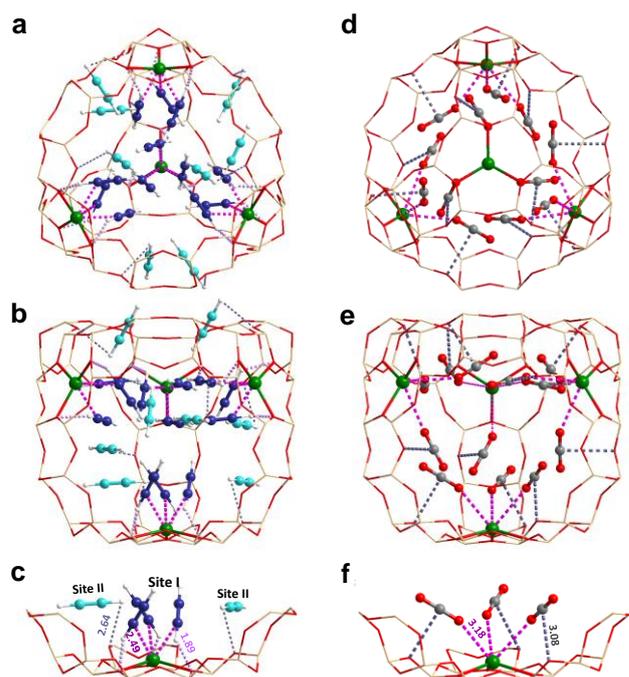


Figure 3. Views of crystal structures of the Cu@FAU zeolite as a function of gas loading. All structures were derived from Rietveld refinements of neutron powder diffraction (NPD) data at 10 K [Si/Al: yellow; O: red; Ni: green. C: grey; D: white; for clarity, C₂D₂(I) and C₂D₂(II) are highlighted in dark and light blue, respectively]. The host-guest interactions are highlighted in dash lines and the estimated standard deviation (e.s.d.) values for binding distances are typically within 0.01-0.1 Å. Views of binding sites for adsorbed gas molecules in [Cu₁₂Na₂₀(Al₄₄Si₁₄₈O₃₈₄)]·(C₂D₂)₃₀ (a) and [Cu₁₂Na₂₀(Al₄₄Si₁₄₈O₃₈₄)]·(CO₂)₁₈ (d) along the (1,1,1) axis; packing of adsorbed C₂D₂ (b) and CO₂ (e) molecules in the supercage; views of the host-guest binding interactions between C₂D₂ (c) or CO₂ (f) and the Cu(II) site and the framework oxygen centres.

Analysis of host-guest binding dynamics. The host-guest binding dynamics has been studied by *in situ* INS (Figure 4). Translational and librational modes of C₂H₂ contribute to the low energy region of the spectra (<30 meV). With both low (1.0 C₂H₂/Cu; peaks labelled as a-f) and high (3.0 C₂H₂/Cu; peaks labelled as a'-f') loading of C₂H₂ in Cu@FAU, two peaks were observed below 8 meV, which correlate with the distinct two C₂H₂ sites identified by NPD. Peak a and a' occurring at lower energy (3.5 meV) are assigned to the translational/rotational motion of C₂H₂ at Site II, which only interacts weakly with the framework oxygen centres. Peak b and b' at higher energy (5.7 and 6.1 meV, respectively) are assigned to the motion of C₂H₂ at Site I within the plane perpendicular to the Cu(II)-gas axis. The fact that the C₂H₂ molecules are more isolated and restricted in anisotropic environment upon adsorption in Cu@FAU results in such distinct INS features at low energy region, whereas coupled and dispersed modes are observed in solid state where C₂H₂ interacts with adjacent C₂H₂ molecules in all three dimensions. INS features c and c' (8-17 meV) are assigned to the rotational or twisting modes of adsorbed C₂H₂ molecules, and the broad peaks d and d' (20 meV) are due to motions of the gas molecules' moving toward or away from the binding sites on the framework.

At the high energy region, peaks e and e' are assigned to the *trans*- C-H bending mode of the adsorbed C₂H₂. Deconvolution of peak e shows two contributing peaks centred at 79.3 and 82.5 meV with a ratio of 0.22:1 (Supporting Information, Figure S36, which correspond to adsorbed C₂H₂

molecules at Site II and I, respectively. Similarly, on additional loading of C_2H_2 , deconvolution of peak e' yields two individual peaks (78.9 and 82.0 meV) and the ratio of peak areas increased to 0.29:1, in good agreement with the NPD and dynamic adsorption results. This result is in sharp comparison to that observed in C_2H_2 -loaded Ni@FAU, where a single adsorption site was observed.^[28] Peak f and f' at ~ 99 meV are assigned to the *cis*- C-H bending mode of the adsorbed C_2H_2 molecule. The apparent blue shift of these peaks comparing to that of solid C_2H_2 (97 meV) indicates that these modes are heavily hindered upon binding to the Cu(II) sites within Cu@FAU. Since both CO_2 and Cu@FAU scatter thermal neutrons weakly (compared with C_2H_2), direct observation of adsorbed CO_2 molecules with INS is challenging. Attempt has been made but no strong and convincing feature was observed in the difference spectrum, consistent with the weak Cu@FAU... CO_2 interactions. The host-guest binding mechanism of C_2H_2 and CO_2 within Cu@FAU has also been investigated by *in situ* FTIR spectroscopy, which confirmed the presence of stronger binding of C_2H_2 than CO_2 , and under dynamic and competitive adsorption conditions, adsorbed CO_2 molecules can be readily displaced by C_2H_2 molecules, consistent with the breakthrough, NPD and INS analysis (Supporting Information, Figure S37).

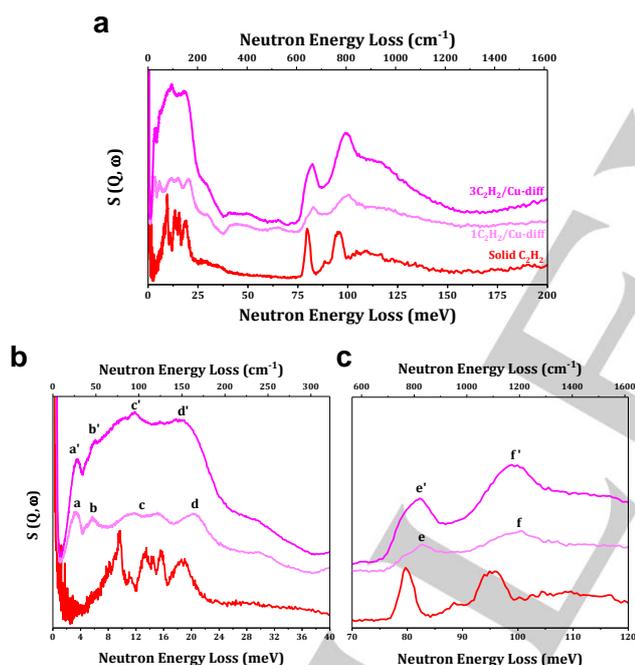


Figure 4. Inelastic neutron scattering (INS) spectra of Cu@FAU as a function of C_2H_2 loading. Diff-spectra were obtained by subtracting the spectrum of bare Cu@FAU from the spectra of C_2H_2 -loaded Cu@FAU (a). (b) and (c) are the enlarged spectrum of (a), showing detailed features at the low and high energy regions, respectively.

Conclusion

The adsorptive separation of C_2H_2/CO_2 based upon porous adsorbents represents a promising energy-efficient strategy to produce high-quality C_2H_2 for industrial applications. By confining coordinatively unsaturated Cu(II) sites in FAU zeolites, Cu@FAU has shown an excellent performance in the separation of C_2H_2/CO_2 , representing the first example of such in zeolite materials. Cu@FAU demonstrates high dynamic uptake of C_2H_2

of 2.29 mmol g^{-1} and dynamic selectivity of 1.71 under ambient conditions. The excellent stability of Cu@FAU in the processes was demonstrated by 8 cycles of binary breakthrough experiments, where Cu@FAU can be fully regenerated with complete retention of the productivity of C_2H_2 (Figure 1e and Supporting Information, Figure S38). Analyses combining *in situ* NPD, INS, and adsorption thermodynamic parameters afford full understanding on the underlying host-guest chemistry that drives the separation process. The confined Cu(II) ions play an important role in the selective binding and retention of C_2H_2 , which is further supplemented by strong host-guest hydrogen bonds, whereas CO_2 displays poor interaction to the zeolite interior and thus is readily displaced by C_2H_2 and eluted from the fixed-bed under flow conditions. By virtue of the different adsorption behaviour and mechanisms of these two molecules, a new *adsorption-purging-desorption* strategy is established for the recovery of C_2H_2 with high purity. **Figure 5** shows a direct comparison between the dynamic separation factor/selectivity of mixtures of C_2H_2/CO_2 and dynamic uptake of C_2H_2 in all leading sorbent materials. In a single separation unit, high effective dynamic uptake of C_2H_2 of 1.51 mmol g^{-1} and an unprecedented separation factor of 22.2 have been achieved on Cu@FAU, yielding the C_2H_2 stream with a high purity of 97.8%. C_2H_2 with a purity of >99% could be obtained by using a secondary separation unit in a modular manner since the large-scale synthesis of Cu@FAU can be readily achieved. This study highlights the versatility of decorated zeolite materials and their great potential in challenging gas separations in industry.

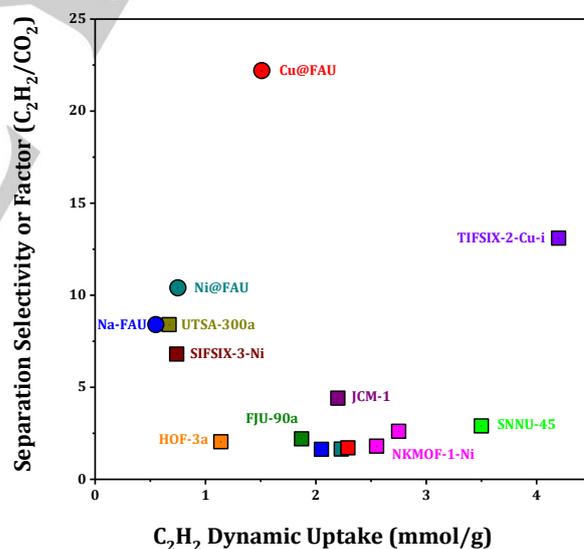


Figure 5. Comparison of the dynamic separation factor/selectivity of the mixture of C_2H_2/CO_2 against the dynamic uptake of C_2H_2 under ambient conditions for state-of-the-art sorbent materials. The three-stage process based upon *adsorption-purging-desorption* (data shown in circle) has significantly increased the effective separation factors. Table S3 (Supporting Information) lists all numerical data of this graph.

Acknowledgements

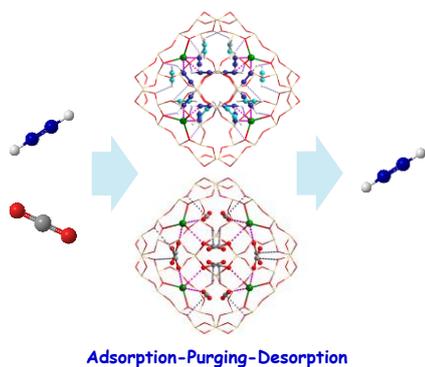
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Keywords: zeolite • gas separation • acetylene • neutron diffraction • neutron scattering

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Entry for the Table of Contents



A decorated zeolite material Cu@FAU exhibits highly selective adsorption of acetylene and thus efficient separation of acetylene and carbon dioxide, unlocking the potential of zeolite materials in challenging industrial separations.