

Effect of Support Surface Properties on CO₂ Capture from Air by Carbon-Supported Potassium Carbonate

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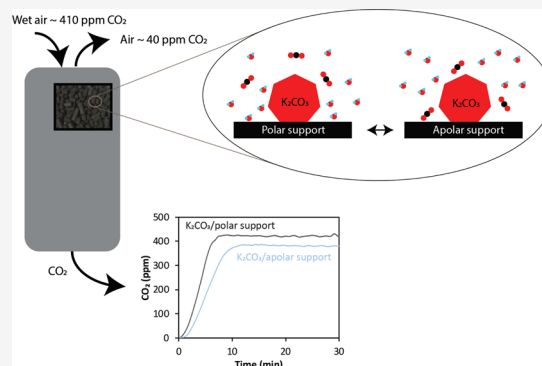


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ABSTRACT: Direct CO₂ capture from air is of prime importance to mitigate the negative effects of global warming. Potassium carbonate (K₂CO₃) is a promising sorbent for the capture of CO₂ from air. K₂CO₃ chemisorbs CO₂ from air in the presence of water (the so-called carbonation reaction), even in very low concentrations (410 ppm). To make efficient use of K₂CO₃, it is dispersed on a support. Carbon-based supports are promising because of their high hydrothermal stability. Carbons have different structural and chemical properties; however, these can be tuned during preparation. Little is known about the relation between carbon properties and their performance, after K₂CO₃ deposition. We investigated the role of support surface composition on the performance of carbon-supported K₂CO₃. By using carbons with different numbers of surface oxygen groups, that is, different polarities, we show that K₂CO₃ supported on an apolar carbon results in a higher capacity CO₂ capture. We propose that a polar support attracts polar H₂O molecules on the K₂CO₃ sorbent, resulting in a low carbonation. In contrast, an apolar support lowers the local H₂O concentration on the sorbent and favorably attracts apolar CO₂ molecules, hence promoting carbonation.



1. INTRODUCTION

The elevated level of atmospheric carbon dioxide is one of the main environmental concerns of our time.¹ We are not reducing emissions fast enough, and emissions from some sources such as non-stationary sources cannot be prevented. Hence, there is an urgent need to rapidly develop new and effective CO₂ capture technologies.² An important step in this direction is to develop sorbents that can efficiently capture CO₂ in low concentrations (410 ppm) from air, release it with little energy input, and sustain performance over a long period. A broad range of solid sorbents have been developed for CO₂ capture from air,^{3–9} such as metal–organic frameworks, immobilized amines, basic metal oxides, and alkali metal carbonates.

Alkali metal carbonates such as potassium carbonate (K₂CO₃) are promising solid sorbents. This type of sorbent can match the energy efficiency of the current commercially applied liquid-phase amines for CO₂ capture from flue gas.¹⁰ The use of solid sorbents instead of, for example, industrially applied amine solutions is beneficial for practical reasons such as minimizing pressure drop and wastewater. Notably, amines are expensive, corrosive, and have toxicity issues. Even if amines are immobilized to form amine-based solid sorbents, they are often more prone to deactivation as they decompose at high temperatures. This sensitivity compels the use of high vacuum in the desorption step or frequently reloading the expensive sorbent.¹¹ Moreover, the long-term stability of amines/imines is poorly understood.¹² As K₂CO₃ is thermally

stable, it forms a greener and more stable solid sorbent for CO₂ capture from air.

In the presence of water, K₂CO₃ reversibly reacts with CO₂ in an exothermic reaction at room temperature (RT) to produce potassium bicarbonate (KHCO₃). In a temperature swing process, full decomposition of the bicarbonate product can be achieved at temperatures below 150 °C.

For efficient use, K₂CO₃ must be dispersed on a support to prevent attrition in the reactors. Different supports have been examined for this application.¹³ Oxidic supports such as Al₂O₃ or Y₂O₃ supporting K₂CO₃ show an initial high CO₂ uptake,¹⁴ but the reaction of K₂CO₃ with these supports results in a new compound such as aluminate that shifts the desorption to a higher temperature.¹⁰ In the long term, an oxidic support dissolves K₂CO₃ in the solid state.

Investigation of other supports such as carbon shows more promise as carbon is inert and has high hydrothermal stability.¹⁰ The carbons have different structural and chemical properties such as different surface areas, porosity, and surface

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functionality; these can be tuned to support any specific application.¹⁵

We know that in heterogeneous catalysis, the support of a catalyst can affect the catalysts' performance via different mechanisms.¹⁶ The support can direct the adsorption of reactants, products, or intermediates; it can indirectly tune the structural properties of the catalyst such as dispersion of the active site over the support, or it can modify the electronic properties of the active phase. However, little is known about the influence of the support on the performance of solid sorbents, a similar category of materials, where an active sorbent material such as K_2CO_3 is also dispersed over a support. The relation between the support properties and their performance after K_2CO_3 deposition is still unknown.

We investigated this relation by comparing sorbents based on supports with a similar structure such as porosity and surface area, but with different surface properties. We chose two different commercial carbon supports, CA1 and SX ultra. These are activated carbons produced by partial burning of organic matter, where CA1 is further steam-treated. They were characterized to determine their structural and chemical properties. We studied these sorbents for the capture of CO_2 from air in a fixed-bed flow-through reactor. We observed that support surface properties affect the performance of K_2CO_3 : Apolar surfaces promote CO_2 adsorption compared to H_2O adsorption, leading to a higher capacity in CO_2 capture.

2. EXPERIMENTAL SECTION

2.1. Support and Sorbent Preparation. We purchased two pristine supports: CA1 (activated charcoal Norit, from wood, chemically activated, Sigma-Aldrich) and SX ultra (EXP 14370, Cabot). They were further treated to remove their surface functionalities. Carbon support (6–10 g) was loaded into a quartz boat and placed in a quartz tube (50 mm i.d.). The tube was placed in a tube furnace and heated under flowing N_2 (100 mL/min) up to 800 °C with step heat at 120 °C and a dwell time of 0.5 h. We named these supports CA1.reduced and SX ultra.reduced.

Sorbents were prepared by incipient wetness impregnation of K_2CO_3 (Sigma-Aldrich, 99.995% trace metals basis) on the carbon supports. In a typical preparation, the support (1 g) was dried under vacuum at 200 °C for 2 h. After cooling down to RT, it was impregnated with an aqueous K_2CO_3 solution (1.7 mL) in an appropriate concentration to prepare 10 and 25 wt. % K_2CO_3 on the carbon supports. The samples were aged at RT under vacuum for 30 min and dried in a freeze drier at –80 °C under 0.1 mbar vacuum for 18 h. The dried samples were then collected and named according to their support name and K_2CO_3 loading, for example, CA1.10 and SX10. At least two different batches of these sorbents were prepared, characterized, and tested for CO_2 capture from air.

2.2. Structural Characterization of the Pristine and Reduced Supports. Nitrogen physisorption measurements were performed at –196 °C (Micromeritics, TriStar II plus) to determine the BET surface area and porosity of the support and the sorbents.

The point of zero charge (PZC) of different supports was determined by a mass titration method.¹⁵ In summary, the carbons were suspended in 10 g of double-distilled water with six different weight loadings: 1, 2, 4, 6, 8, and 10 wt %. The pH of the solutions was measured using a VWR pH1100L meter after these solutions were stirred and de-gassed for 24 h under

N_2 in order to prevent the pH change due to CO_2 dissolution from the air.¹⁷

We characterized the type of surface functional groups on the supports using temperature-programmed desorption coupled with a mass spectrometer (TPD-MS), following evolved gases from different supports upon high-temperature treatment using an automated catalyst characterization system (Micromeritics AutoChem II 2920). Typically, 50 mg of the support was heated in a U-shaped reactor from RT to 900 °C (10 °C/min) under He flow (20 mL/min). The temperature and signal from a thermal conductivity detector (TCD) were recorded by the AutoChem analyzer. Evolved gases were further detected by an online Pfeiffer THERMO Star mass spectrometer. We followed molecules with molecular masses of 2, 4, 18, 28, 32, 40, and 44 g/mol, which correspond to H_2 , He, H_2O , CO, O_2 , Ar, and CO_2 , respectively. Then, we manually calculated the peak area and mass quantification. The functional groups on the supports were also analyzed and quantified by XPS. The analysis was performed with a PS-9200 photoelectron spectrometer from JEOL, Japan.

2.3. CO_2 Capture. CO_2 capture experiments were performed using an in-house setup. The setup configuration is schematically shown in Figure 1. It contains two mass flow

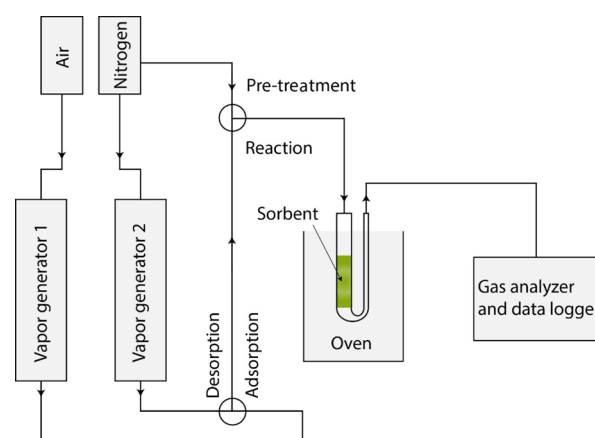


Figure 1. Schematic presentation of the in-house CO_2 capture setup.

controllers to adjust the flow of nitrogen and air. Water bubblers at adjustable temperatures were inserted in the flow to adjust flow humidity. A wet/dry flow of air/nitrogen was directed to a reactor located in a furnace and further to a gas analyzer.

In a typical experiment, the sorbent (50 mg) was placed in a U-shaped quartz plug flow reactor (internal diameter of 10 mm). The bed size was 4 mm and the gas hourly space velocity was 20,000 1/h. The sorbent was pre-treated from RT to 200 °C (ramp 10 °C/min) under flow of dry N_2 (100 mL/min) and kept at 200 °C for 30 min. Then, the sorbent was cooled to 60 °C to start the CO_2 capture experiment. The sorbent was exposed to a flow of air (100 mL/min, ~430 ppm CO_2 , water content: 3 vol %, vapor-generated, and stabilized at RT) for 1 h while it was kept at 60 °C. The outlet was analyzed every 1 s by a nondispersive infrared gas detector (LI-840A CO_2/H_2O gas analyzer, LI-COR) to record CO_2 and H_2O concentration. CO_2 breakthrough curves during CO_2 capture from air were obtained. Further, the CO_2 sorption capacity was calculated by dividing total moles of CO_2 uptake by the dry amount of sorbent applied in each test. To calculate the efficiency of CO_2

uptake, the total moles of CO₂ uptake were divided by the corresponding moles of K₂CO₃ in each test, taking the 1:1 stoichiometric mole ratio of CO₂ to K₂CO₃ into consideration. In this reaction condition, there was no pressure drop, and external and internal mass transfers were not limiting factors. Last, we performed CO₂ desorption on heat treatment (10 °C/min up to 200 °C) under a flow of dry N₂ (100 mL/min).

3. RESULTS AND DISCUSSION

3.1. Structural Characterizations of the Carbon Supports and Sorbents. Table 1 summarizes the structural

Table 1. Structural and Chemical Properties of the Selected Commercial Carbon Supports

sorbent	surface area (m ² /g)	total pore volume ^a (cm ³ /g)	micropore volume (cm ³ /g)	PZC
CA1	1000	1.0	0.27	2
SX ultra	940	0.7	0.23	7

^aMeasured at P/P_0 of 0.99.

and chemical properties of CA1 and SX ultra. These were extracted from nitrogen adsorption–desorption isotherms (Figure S.1). Both supports have a similar surface area and contain both micropores and mesopores; therefore, they have very similar structural properties.

The surface properties of both supports were evaluated by measuring their points of zero charge (PZC, Table 1) using mass titrations. The mass titrations (Figure S.2) clearly showed an acidic surface for CA1 with a PZC of 2 and a neutral surface for SX ultra with a PZC of 7. The presence of surface oxygen groups was further evaluated by TPD-MS. Figure 2 shows the

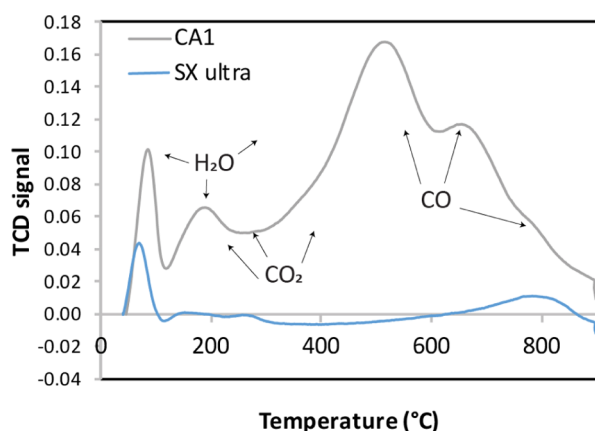


Figure 2. Surface oxygen groups evolved from CA1 and SX ultra (50 mg) on high-temperature treatment under the flow of He. Evolved gases were identified by an online mass spectrometer.

evolved gases detected by a TCD. Mass spectrometry identified desorption of H₂O, CO₂, and CO representing the decomposition of surface oxygen groups during heating to high temperature. At temperatures around 100 °C, water evolved from both supports, indicating the sample wetness. At higher temperatures, H₂O, CO₂, and CO evolved from CA1, while little CO and CO₂ were released from SX ultra. According to Li et al.,¹⁸ CO₂ evolving at high temperatures indicates different oxygen-containing functional groups such as strong acidic carboxylic groups (245 °C), weak acidic carboxylic

groups (325 °C), and carboxylic anhydrides (480 °C). Moreover, CO formation at high temperature indicates cyclic oxygen-containing functional groups such as carboxylic anhydrides (500 °C), phenols (635 °C), and quinones (760 and 878 °C).

The evolution of these groups from CA1 on heating is clearly visible in Figure 2. This shows that CA1 contains oxygen-containing functional groups on the surface, while SX ultra has little functionality compared to CA1. XPS analysis (Figure S.3) indicated that CA1 contained 90 wt % carbon and 10 wt % oxygen and no other detectable elements; this qualitatively agrees with the release of a significant amount of CO/CO₂ from that support in the TPD-MS experiments. Surprisingly for SX ultra, XPS indicated the presence of oxygen (5 wt %) while TPD-MS (Figure 2) did not show any formation of CO₂/CO during heating. Since XRD (Figure S.4) indicated the presence of SiO₂ in SX ultra, we conclude that the oxygen visible in XPS originated from SiO₂.

Since the presence of surface oxygen groups is related to polarity, the polarity of each support was evaluated by suspending the carbons in a mixture of water–ethanol (polar phase)/hexane (apolar phase) (Figure S.5). These experiments show that CA1 moves preferentially to the polar phase, while SX ultra moves preferentially to the apolar phase. This demonstrates that CA1 has a polar surface due to the presence of oxygen groups, while SX ultra lacks surface functionality, therefore has an apolar surface.

Determination of the characteristics of supported K₂CO₃, that is, average particle size, size distribution, and dispersion over the porous support, is challenging. This is mainly because common visualization techniques such as scanning electron microscopy and transmission electron microscopy do not provide enough contrast to distinguish K₂CO₃ particles from the carbon support. We extensively addressed characterization of supported K₂CO₃ particles and the influence of their size on CO₂ capture performance elsewhere.¹⁹

3.2. Performance in CO₂ Capture from Air. Potassium carbonate supported on these two supports was examined for CO₂ capture from air. Figure 3 shows the corresponding breakthrough curves after 40 min of sorption, and Table 2 summarizes the sorbent CO₂ uptake. Please note that in all breakthrough curves, the CO₂ concentration eventually reached 430 ppm (Figure S.6). K₂CO₃/SX ultra has a higher uptake compared to K₂CO₃/CA1. The differences between the samples were more pronounced when K₂CO₃ loading was lower (Figure 2b). Therefore, we show that supports have an effect on CO₂ capture by K₂CO₃.

Notably, SX ultra contains some SiO₂. As reported earlier,²⁰ K₂CO₃ on silica support has a very low capacity in CO₂ capture compared to K₂CO₃ on carbon; hence, the presence of SiO₂ impurities in SX ultra is most likely not the reason for the higher uptake of SX ultra-based sorbents. We therefore hypothesize that the lower surface polarity of SX ultra is the main cause for its superior CO₂ uptake.

To validate this hypothesis, the acidic (oxygen) groups of CA1 were removed by high-temperature treatment (up to 800 °C under N₂). Substantial weight loss (31 wt %) indicated substantial removal of the surface functions on heat treatment. Nevertheless, the support's structural properties did not change (Figure S.1). The K₂CO₃ supported on CA1 with a reduced number of acidic groups (CA1.reduced.25) showed enhanced CO₂ uptake (Figure 2c and Table 2). The capacity increased from 0.30 to 0.70 mmol CO₂/g sorbent, indicating

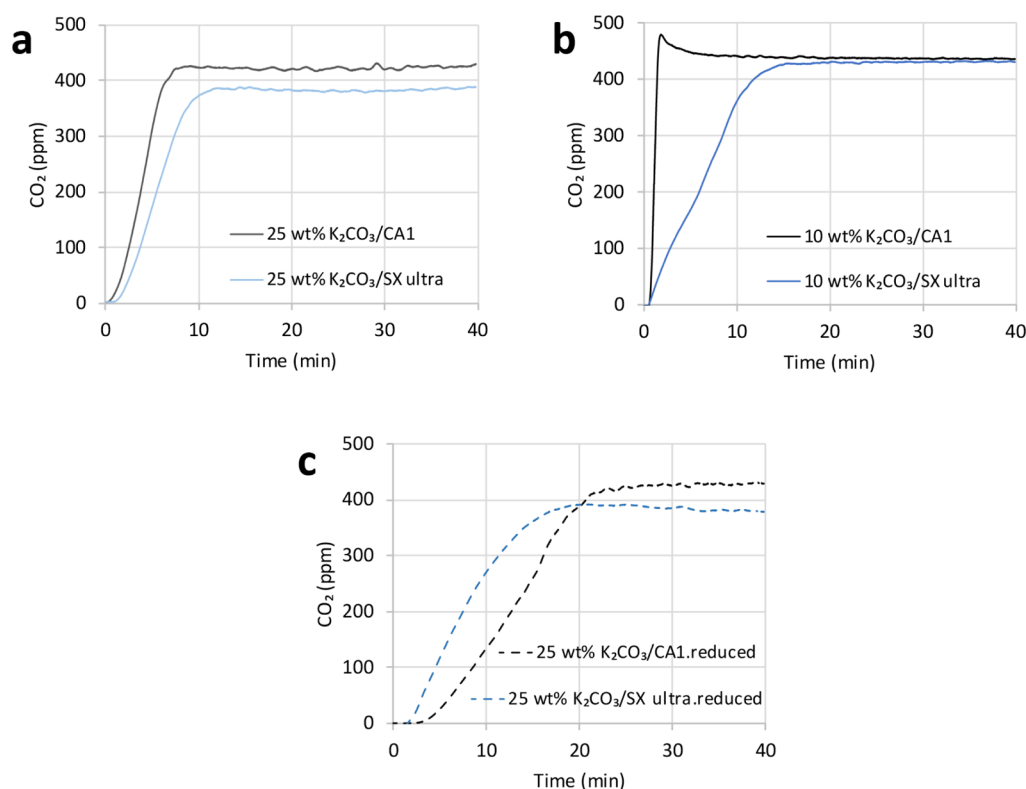


Figure 3. CO₂ breakthrough curves during CO₂ capture from air. The air passed down-flow through different sorbents: (a) 25 wt % K₂CO₃ on CA1 and SX ultra, (b) 10 wt % K₂CO₃ on CA1 and SX ultra, and (c) 25 wt % K₂CO₃ on reduced functionality CA1 and SX ultra. 50 mg, pre-treatment RT to 200 °C: 10 °C/min, dwell time: 30 min under the flow of dry N₂ (100 mL/min), sorption at 60 °C for 60 min, and flow of air (100 mL/min, 430 ppm CO₂, and 3 vol % H₂O).

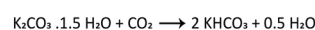
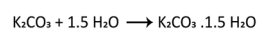
Table 2. CO₂ Sorption Capacity of the Samples

sample	K ₂ CO ₃ loading (wt %)	CO ₂ uptake (mmol CO ₂ /g sorbent)	CO ₂ uptake (mol CO ₂ /mol K ₂ CO ₃)
CA1.10	10	0.01	0.01
CA1.25	25	0.30	0.17
CA1.reduced.25	25	0.70	0.40
SX.10	10	0.29	0.40
SX.25	25	0.54	0.30
SX.reduced.25	25	0.54	0.30

that removing oxygen groups is beneficial for CO₂ uptake. The same heat treatment did not improve the SX ultra-based sorbent (Figure 2c) because SX ultra lacks surface functional groups.

Figure 4 shows a hypothesis for the effect of support surface properties on CO₂ sorption. A polar surface attracts the polar H₂O molecules on the K₂CO₃ sorbent rather than the CO₂ molecules, while an apolar surface repels H₂O molecules. Literature suggests that K₂CO₃ reacts with CO₂ and H₂O (1:1:1 stoichiometry) to form KHCO₃, the so-called carbonation reaction. This takes place either directly or via formation of intermediate hydrate, the so-called hydration reaction. In contrast to early reports, the hydration reaction is proposed to compete with the direct carbonation reaction. Therefore, hydrate formation limits the carbonation reaction rather than being a necessary intermediate.^{21–23} We propose that the support, via its polarity, affects the local concentration of H₂O and CO₂ on the K₂CO₃ sorbent. A polar support attracts polar H₂O molecules to the sorbent promoting hydration, while an apolar support repels H₂O molecules,

Intermediate hydrate formation:



Direct carbonation reaction:

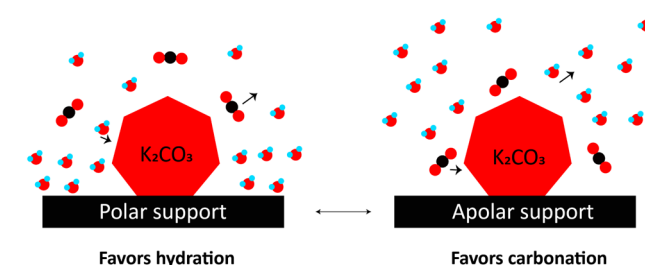
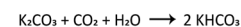


Figure 4. Schematic presentation of the effect of support surface functions on carbonation and hydration reactions.

lowers its local concentration, and attracts apolar CO₂ molecules, hence promoting carbonation reaction. In this way, the support affects the performance of K₂CO₃, and the apolar surface function promotes preferential carbonation rather than the competitive hydration reaction.

Jayakumar et al.²³ predicted that a hydrophobic support would promote carbonation above hydration because of a lower apparent H₂O concentration or a higher apparent CO₂ concentration on K₂CO₃. However, to the best of our knowledge, this has never been demonstrated experimentally.

To further investigate the water effect on CO₂ capture by K₂CO₃-based sorbents, K₂CO₃ on CA1 with reduced functionality was pre-treated with either wet or dry N₂ before a CO₂ capture experiment. In this way, the hydration reaction would be promoted (wet N₂-pretreatment) or limited (dry N₂-

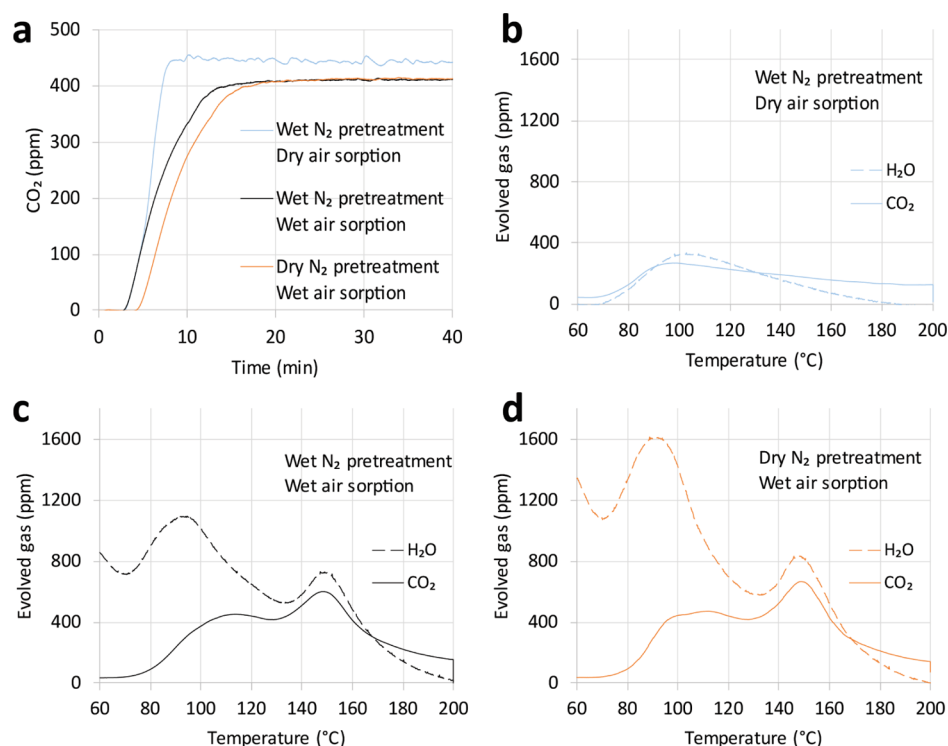


Figure 5. (a) CO_2 breakthrough curves during CO_2 capture from air passed down-flow through 25 wt % K_2CO_3 on CA1 with reduced functionality (50 mg), pre-treatment RT to 200 °C: 10 °C/min, dwell time: 30 min under the flow of dry N_2 (100 mL/min), followed by cooling and 60 min dwell time at 60 °C under the flow of dry or wet N_2 (100 mL/min, 0 or 3 vol % H_2O), sorption at 60 °C for 60 min, and flow of dry or wet air (100 mL/min, 430 ppm CO_2 , 0 or 3 vol % H_2O). (b–d) Desorption of CO_2 and H_2O after each sorption experiment upon heat treatment (10 °C/min up to 200 °C) under the flow of dry N_2 (100 mL/min).

pretreatment) compared to the carbonation reaction. Figure 5a shows the result of these experiments. After pretreatment under wet N_2 and during sorption under dry air, the breakthrough of CO_2 was the fastest (Figure 5a blue line), resulting in the lowest CO_2 uptake. In comparison, pretreatment under wet N_2 and sorption under wet air led to a higher CO_2 uptake (Figure 5a black line). This confirms that the co-presence of H_2O as one of the reactants is advantageous for the carbonation reaction. Further, pretreatment under dry N_2 and sorption under wet air led to the highest CO_2 uptake (Figure 5a, orange line). Thus, a pretreatment with water has an inhibitory effect, while the presence of water during sorption is advantageous.

Figure 5b–d shows H_2O and CO_2 evolved from sorbents during consecutive desorption steps after each sorption experiment, as described above and in Figure 5a. Figure 5b shows that after a sorption step under dry air, little CO_2 and H_2O were released at 100 °C. This confirms that pretreatment under wet nitrogen and sorption under dry air leads to low H_2O and CO_2 sorption. Moreover, no extra H_2O (compared to stoichiometric KHCO_3 decomposition) was released; thus, no extra hydrate has been formed. Therefore, one can conclude that not all K_2CO_3 , upon exposure to a wet flow, form hydrate to be consecutively transformed to bicarbonate upon exposure to CO_2 .

Figure 5c,d shows a higher release of CO_2 and H_2O , indicating that sorption under wet air leads to a higher CO_2 sorption compared to sorption under dry air. Interestingly, there are two peaks related to bicarbonate decomposition at different temperatures (110 and 150 °C). The one decomposed at 110 °C is similar to the one observed in

Figure 5b; however, the one decomposed at 150 °C is a consequence of the presence of water during sorption. Previously, we showed that diffusion of CO_2 and H_2O in and out of K_2CO_3 particles, in a cycle of sorption/desorption, follows a multilayer process.¹⁹ First, K_2CO_3 on the outer layers of particles reacts rapidly with CO_2 and H_2O and consecutively desorbs them with a lower energy input. Hence, we attribute the release of CO_2 and H_2O at 100 °C to decomposition of bicarbonate at outer layers of K_2CO_3 particles. Second, the inner layers of K_2CO_3 react with CO_2 and H_2O in a diffusion limited step. These inner layer bicarbonates decompose with a higher energy input. Hence, we attribute the release of CO_2 and H_2O at 150 °C to decomposition of bicarbonate at inner layers of particles. Since sorption under wet air leads to a higher CO_2 sorption compared to sorption under dry air and further causes appearance of bicarbonates that decompose at 150 °C, we propose that the presence of water during sorption facilitates diffusion of CO_2 and H_2O into the inner layers of bulk K_2CO_3 . Hence, the co-presence of water during sorption affects the mechanism of sorption and in this way can boost the capture.

Moreover, pretreatment under wet N_2 (Figure 5c) neither promoted CO_2 nor H_2O sorption compared to pretreatment under dry N_2 (Figure 5d).

In brief, we show a contradictory effect of water. A wet pretreatment led to a lower CO_2 uptake showing an inhibitory effect of water, even though no separate hydrate formation was detected. In contrast, sorption in the presence of water led to a higher uptake showing a boosting effect of water. We propose that the inhibitory effect of water can be diminished by applying an apolar support via lowering the local H_2O

concentration on the sorbent to an optimal level. We further propose that the co-presence of water during sorption boosts the diffusion of CO₂ and H₂O more into inner layers of K₂CO₃ particles. These outcomes may explain the contradiction reported in the literature regard the water effect on CO₂ capture by K₂CO₃-based sorbents. More investigations to reveal the role of water and optimal water concentration in the CO₂ sorption mechanism are needed.

To indicate the stability of these sorbents, CA1.reduced.25 was prepared and stored for 6 months. After that time, the material was used and the capacity was compared to that of a fresh sample (Figure S.7). After storing, the material lost 20% of its capacity, which is low but appreciable. In addition to indicate the stability of the material, the material was exposed to 10 sorption/desorption cycles in a potential industrially relevant setting for CO₂ capture from air (see the Supporting Information, Figure S.8). The sorbent showed no sign of deactivation during these 10 cycles, which makes this a promising material for application. Nevertheless, more investigation is needed to assess the overall energy consumption of the whole process. In addition, studying side reactions (such as formation of KOH) can give more insights into the potential deactivation pathways (which falls outside the scope of this paper).

4. CONCLUSIONS

We prepared potassium carbonate sorbents based on carbon supports with similar structural properties but with different surface properties. One of the supports had polar surface properties due to surface acidic functions, and the other was an apolar support with little surface functionality. The latter considerably promoted CO₂ sorption from air. Hence, we show that the support surface properties affect the performance of K₂CO₃, an important consideration for sorbent development for CO₂ capture from air.

■ ASSOCIATED CONTENT

SI Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acs.iecr.1c01229>.

Nitrogen adsorption–desorption isotherms for CA1, CA1.reduced, and SX ultra; mass titration of CA1 and SX ultra; wide XPS spectra of CA1 and SX ultra; XRD patterns of CA1 and SX ultra; pictures showing miscibility of the supports in different media; CO₂ breakthrough curves for 25 wt % on SX ultra during CO₂ capture from air for 85 min; long-term stability of CA1.reduced.25; CO₂ breakthrough curves during CO₂ capture from air on fresh and old CA1.reduced.25; and CO₂ breakthrough curves during 10 cycles of CO₂ sorption/desorption on CA1.reduced.25 (PDF)

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Notes

The authors declare no competing financial interest.

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