

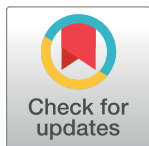
RESEARCH ARTICLE

Kinetic analysis of an anion exchange absorbent for CO₂ capture from ambient air

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Abstract

This study reports a preparation method of a new moisture swing sorbent for CO₂ capture from air. The new sorbent components include ion exchange resin (IER) and polyvinyl chloride (PVC) as a binder. The IER can absorb CO₂ when surrounding is dry and release CO₂ when surrounding is wet. The manuscript presents the studies of membrane structure, kinetic model of absorption process, performance of desorption process and the diffusivity of water molecules in the CO₂ absorbent. It has been proved that the kinetic performance of CO₂ absorption/desorption can be improved by using thin binder and hot water treatment. The fast kinetics of P-100-90C absorbent is due to the thin PVC binder, and high diffusion rate of H₂O molecules in the sample. The impressive is this new CO₂ absorbent has the fastest CO₂ absorption rate among all absorbents which have been reported by other up-to-date literatures.

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1. Introduction

As the announcement of Intergovernmental Panel on Climate Change (IPCC), the CO₂ emissions will rise between 48 and 55 Gt/yr by 2050, with the energy demands of 40% to 150% increase[1]. Atmospheric CO₂ will be ranging from 535 to 983 parts per million (ppm) by 2100, roughly double current value, 406 ppm. CO₂ concentration increment leads to a global mean temperature change from 1990 to 2100 of between 1.4 °C and 6.1 °C[2]. The significance and urgency of the development of CO₂ capture from ambient air has been discussed elsewhere[3–6].

In order to compensating for CO₂ emission to ambient air, a moisture-swing sorbent for CO₂ capture from ambient air was proposed[7], which provides a novel approach to absorb CO₂ in dilute streams. The moisture-swing CO₂ absorbent is an anion exchange resin[8–14] (IER). IER acts like a strong base, analogous to NH₄⁺, where each hydrogen has been replaced by an organic carbon chain attached to a polymer matrix. The chemical structure is shown in Fig 1. The isothermal[15, 16] and kinetic[13, 17] performance of the resin-based sorbent have been revealed systematically. The novel principle of the CO₂ absorption/desorption process over IER was well illustrated and clarified[8, 18]. The reason is that reduction of the number of water molecules presenting in the pore space promote the hydrolysis of CO₃²⁻ to HCO₃⁻ and OH⁻. This phenomenon enables a nano-structured CO₂ absorbent possible to capture CO₂

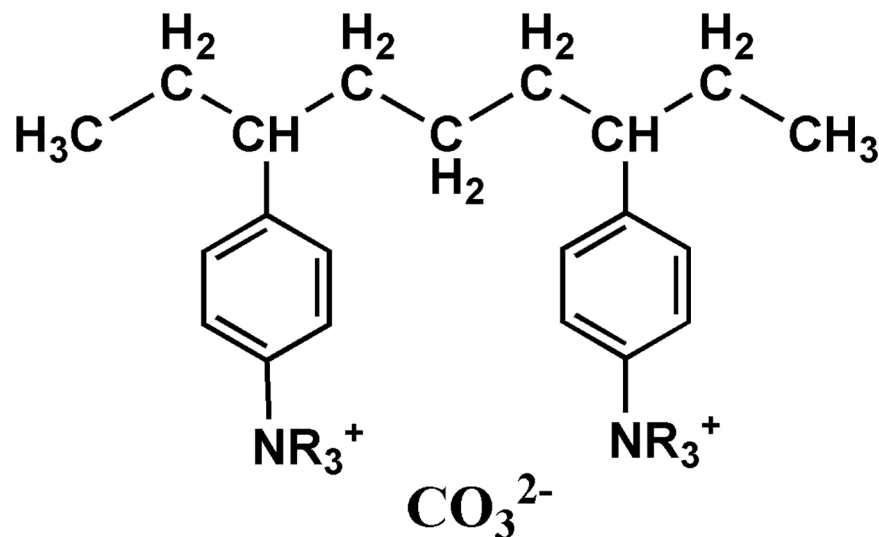


Fig 1. Chemical structure of ion exchange resin containing two side chains. The exchanged anion is CO_3^{2-} .

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spontaneously in the ambient air when the surrounding is dry, while releasing it when exposed to moisture[8, 9]. In the dry condition, a CO_3^{2-} ion splits a H_2O molecule to form a HCO_3^- and a OH^- ion which both bind tightly to their respective NR_4^+ cations. OH^- ion absorbs CO_2 even at a low partial pressure of CO_2 . This results in a CO_2 -loaded state which is entirely HCO_3^- . In the wet condition to regenerate the full-loaded absorbent, each two HCO_3^- ions react to produce a CO_3^{2-} ion, a H_2O and a CO_2 . The released CO_2 in the wet condition can be collected collectively. The underlying mechanism has been revealed by Shi[9] and the phenomenon can be applied to a series of counter-intuitive chemical reactions which is related to the hydrolysis of basic ions[10]. Meanwhile, the discovery opens a new approach for the technology of gas separation.

For these conditional CO_2 capture methods, like thermal-swing CO_2 absorbents[19], increasing absorption capacity is a significant task due to the high cost (the regeneration process consumes heat) on the absorbent regeneration. However, for the novel water-driven CO_2 absorbent, kinetics improvement is a more interesting factor due to the low cost (the regeneration process consumes water) of the regeneration part[17]. The energy consumption and cost can be reduced significantly according to improving the absorption rate of the water-driven CO_2 sorbent. The objective of this study is to propose a new moisture-swing CO_2 sorbent (P-100) by using ion exchange resin (IER) as a functional group and polyvinyl chloride (PVC) as a binder. The kinetic characterization of the new CO_2 sorbent has been enhanced significantly comparing to sorbent I-200[15], which is manufactured by Snowpure LLC, California. The preparation process of this new sorbent is introduced first, and the analysis of kinetic performance is presented next based on the studies of material structure, CO_2 absorption/desorption process and water diffusion experiments.

2. Materials and preparation process

2.1 Materials

A heterogeneous ion-exchange material in a flat sheet was prepared in this study. The material includes: 1) ion exchange resin (IER)[7]. The IER is composed of a polystyrene backbone with

attached quaternary amine ligands. 2) Polyvinyl chloride (PVC). PVC is a widely produced synthetic plastic polymer which was used as a binder. 3) Tetrahydrofuran (THF). THF is an organic compound with the formula (CH₂)₄O which was employed as a solvent to mix IER and PVC.

2.2 Preparation of anion-exchange sorbent

The heterogeneous CO₂ absorbent was prepared by using dip-coating technique[20]. First, the IER particles were grinded in a ball mill and then filtered using a mesh with 44~74 micrometer openings. Then, PVC was dispersed into THF in a glass reactor and stirred mechanically for more than 5 hours. The weight ratio of PVC to THF is 1:20. Next, powdered resin particles (44~74um) were added into the mixture of PVC and THF. The mechanical stirrer stirred vigorously at room temperature for 30 minutes to mix IER and PVC uniformly. The IER to PVC weight ratio is 1:1 and the total solid to THF ratio is 1:10 (w/v)[21]. After completed mixing, the dip coating method was conducted by using dry clean glass plate. The thickness of the produced membrane was 100 micron. The membrane was dried in the ambient air at temperature 25°C for 30 min, and then was immersed in distilled water. Last, three absorbent samples were treated by different temperatures of water for 48 hours (25°C water (P-100-25C), 50°C water (P-100-50C), 90°C water (P-100-90C)). I-200 sample was only treated by 90°C water as a reference. Three P-100 samples and an I-200 sample, containing the same load of IER, were immersed in 1.0 M sodium carbonate solution for 2–4 hours[15]. Samples were washed 4–5 times, and then washed by plenty of deionized water (DI water) to flush away the sodium carbonate solution residues on the samples. Afterwards, the samples were ready-to-use.

2.3 The absorption capacity of CO₂ sorbent

The Mohr method was used to determine the effective ion charge density ρ_c of the IER. The ρ_c is 1.58 mol/kg. CO₂ capacity Q_{est} is 17.69 L/kg, which was estimated by ρ_c at standard condition. The CO₂ capacity Q_∞ was also measured by experiments under the condition of 1000 ppm CO₂ partial pressure. The value of Q_∞ is 16.4L/kg. The effective charge density and CO₂ capacity of the absorbent can be both enhanced according to increasing the weight ratio of IER to PVC during the sample preparation process.

3 Experimental methods and models

3.1 Absorption experiment

The experimental device with humidity control was set up to measure the half-time (the time when the absorbent reaches half of its capacity) of the moisture-swing CO₂ absorbents. A layout of the device is shown in Fig 2. The CO₂ concentration change was measured by two infrared gas analyzers (IRGA). Measurements were recorded once per second. The wet and fresh samples (each sample contains 0.30g IER) were put into a sealed chamber one by one and flushed with 2L/min CO₂-free dry air. The water concentration in the air at outlet was monitored to determine whether the samples were sufficiently dried. (The water concentration in the CO₂-free dry air at inlet and water concentration in the CO₂-free dry air at outlet were same, when samples had been sufficiently dried. The water concentration was about 2% relative humidity,). The 1 L/min air containing 400 ppm CO₂, went through a dew point generator (MODEL LI-610). The air contained 30% relative humidity and flowed over all sorbent samples. Entire absorption process would last until CO₂ concentrations were same at inlet and outlet with 1% error.

3.2 Desorption experiment

The absorbed CO₂ by absorbents will be released when the absorbents are exposed to a high humidity or liquid water. Meanwhile, the absorbents continue to absorb water vapor from air when they are put into a higher humidity until the system reaches to an equilibrium state. The absorbed water molecules increase the weight of the absorbents. Here, the desorption experiment is to analyze the kinetic characteristics of the absorbents by studying the equilibrium time of the process of H₂O absorption and CO₂ desorption. The diffusion coefficients of water molecules in the CO₂ absorbents were derived by calculating the weight change of the samples. The diagram of the experimental device is shown as Fig 3

The total amount of CO₂ in the closed-loop experimental device is constant. We can track the amount of absorbed/desorbed CO₂ by measuring the CO₂ concentration in the air. The device can control the water concentration in the air by heating or cooling the condenser. The weight change of the absorbents can also be measured by weight scale in the device.

Four samples (25°C-water-treated P-100, 50°C-water-treated P-100, 90°C-water-treated P-100, and 90°C-water-treated I-200) containing the same resin load, were firstly exposed to pre-treated dry air (dew point -18°C) for two hours to be fully dried and loaded. Next, put the full-loaded samples into experimental device (as Fig 3) subsequently and increased the humidity level to 15°C. The increase of sample weight and CO₂ concentration in the experimental device were measured separately. The increase of the sample weight was mainly due to the water absorption on the samples. A humidity controller was employed to ensure a constant humidity

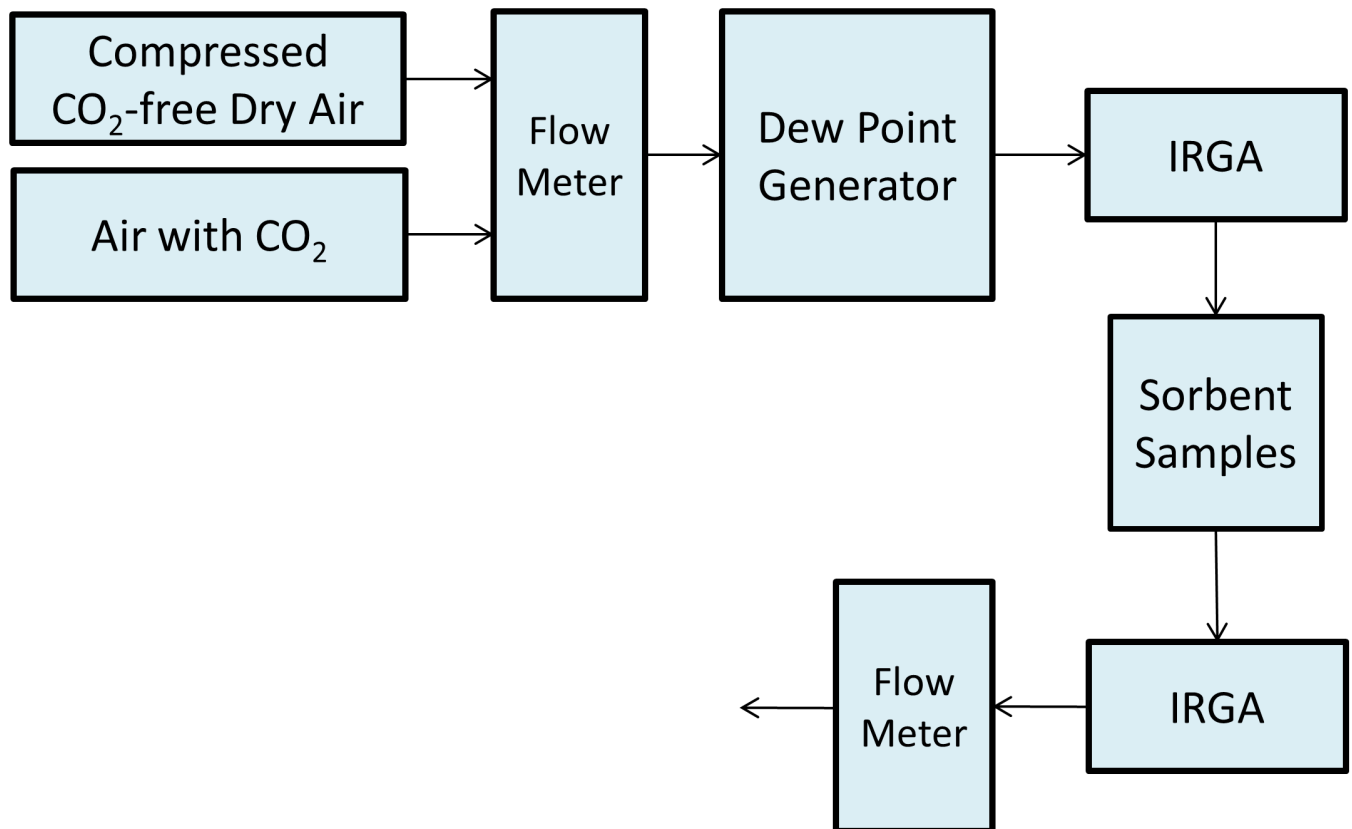


Fig 2. Schematic of experimental device. The CO₂ concentration can be tracked at inlet and outlet of the chamber of sorbent sample. The amount of absorbed CO₂ can be calculated by measuring the CO₂ concentration change with time. Dew point generator can control the water vapor concentration in the system.

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in the chamber by PID control. CO₂ concentration change was recorded every second by infrared gas analyzers (IRGA).

3.3 Absorption kinetics of sorbent

The absorption characteristics of P-100-90C sorbent was depicted by Lagergren pseudo first-order model[22], which has been most frequently employed to present absorption dynamic process under various conditions.

$$\frac{dq}{dt} = k(q_e - q) \tag{1}$$

q is absorption quantity at time t , k is constant number, q_e is equilibrium isotherm absorption capacity. Integrating Eq 1 with boundary conditions (a) $t = 0, q = 0$; (b) $t = t, q = q_e$

$$q = q_e(1 - e^{-kt}) \tag{2}$$

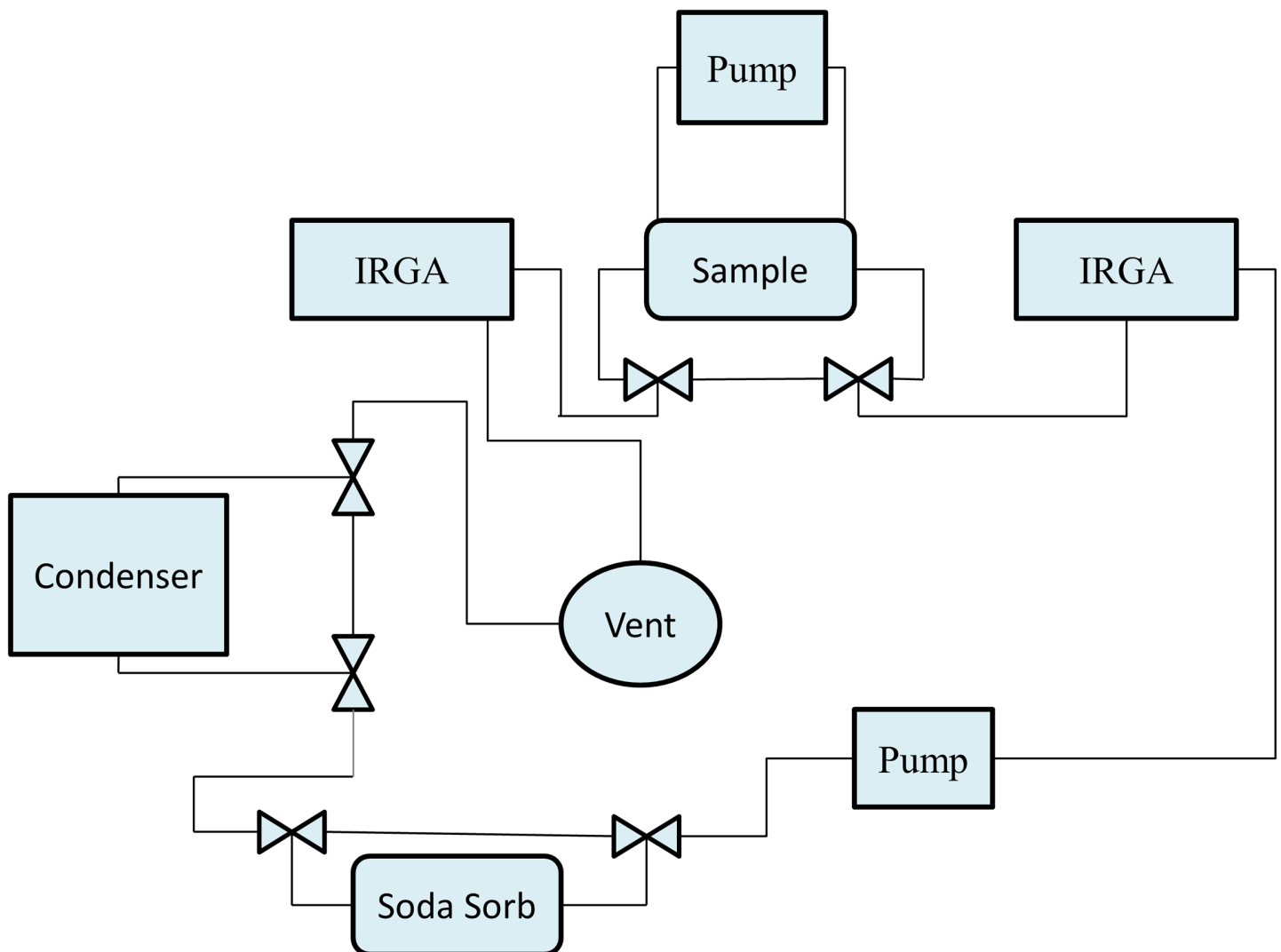


Fig 3. The schematic of the experimental device. The total amount of CO₂ in the sample and in the gas volume is constant. The process of CO₂ absorption/desorption can be identified in the experimental device.

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4 Results and discussion

4.1 Absorbent structure analysis

The structures of the P-100 sorbents, which were treated by different-temperature hot water, were studied by scanning electron microscope SEM (Agilent Technologies, SE 1000V) as shown in Fig 4. Obviously, hot water treatment results in the formation of narrow cavities between anion exchange particles and PVC matrix, as well as larger amount of pores in PVC materials.

Micro-structure schematic of P-100 sorbent can express the percolation structure of P-100 sorbent. After the treatment of 90 °C hot water, some small islands of interconnected particles appear and these connections form extended pathways. More and more IER particles are connected by channels if the connections keep growing. The chance of the appearance of percolation threshold can increase the rate and range of gas diffusion inside the sorbent. According to the observation of SEM, much more resin particles in the new sorbent P-100 are exposed into air than the ones held by I-200[15] because of the thinner thickness of membrane and the more continuous channels inside P-100. Moreover, percolation threshold may further promote the conduction level between surrounding air and resin particles. Therefore, treating P-100 by hot water may promote IER particles to be exposed to ambient air, thereby further improving the performance of the CO₂ absorbent.

4.2 Absorption half-time

The kinetic characteristics are significant factors for moisture-swing CO₂ capture sorbent. Absorption kinetics of the sorbent are determined by mass diffusivity in the materials, heat transfer into and out of the pores, and intrinsic chemical reaction rates[17, 23, 24]. As a preliminary assessment for the CO₂ absorption kinetics, absorption half time is an assessment factor²³ to evaluate the absorption rate of CO₂ absorbent. The absorption half time is expressed by Eq 3:

$$T_{half-time} = \frac{T_{CO_2}}{2} = T_{H_2O} + T_{reaction} \quad (3)$$

T_{CO_2} is the time for CO₂ absorption by sorbent from fresh-empty status to full-loaded status, T_{H_2O} is the absorption time of water on sorbent; $T_{reaction}$ is the time of intrinsic chemical

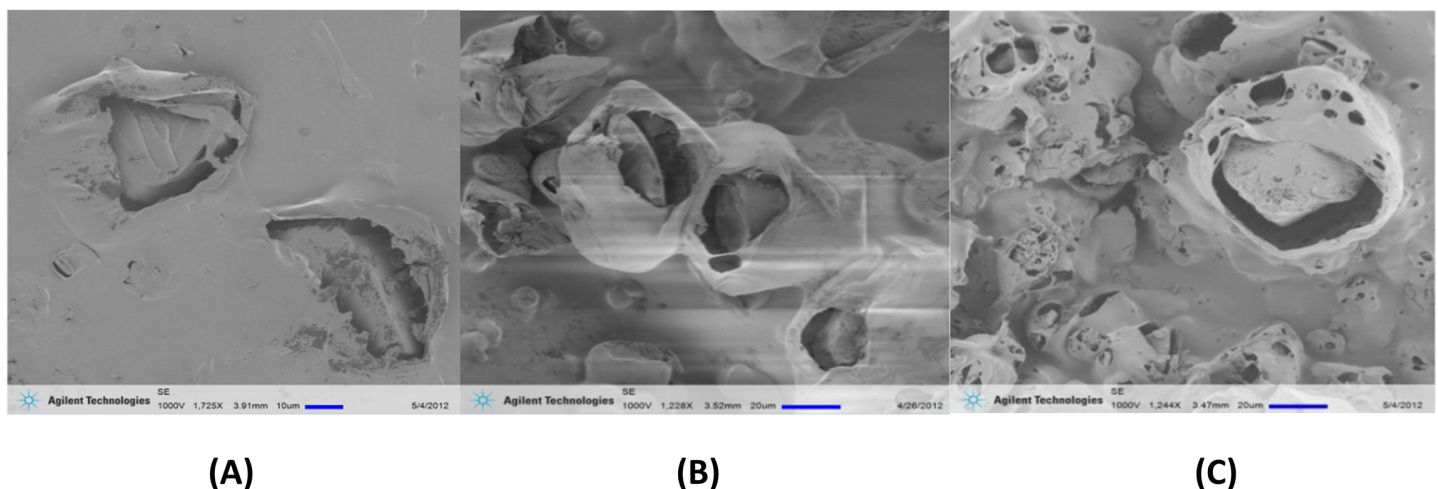


Fig 4. SEMs of P-100 sorbents treated with different water temperatures. (A) 25 °C-water-treated sample P-100-25C, (B) 50 °C-water-treated sample P-100-50C, (C) 90 °C-water-treated sample P-100-90C.

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reaction. The half time of sample P-100-25C, P-100-50C, P-100-90C, and sample I-100 was measured by experimental device, shown as Fig 2.

Fig 5 displays the half times of the four different samples under simulated air capture condition (30% relative humidity, 400ppm). Other half time of current air capture CO₂ sorbents[23, 25] in literatures have also been presented.

The sample P-100-90C owns the best kinetic characteristics comparing with the other three moisture-swing CO₂ sorbents. The 31.8 min half time is also the shortest half time in all air capture sorbents which have been reported by other up-to-date literatures. Obviously, the kinetics of P-100-90C is better than the other two P-100 sorbents, because the hot water treatment enlarges the surface area of IER to be exposed to the ambient air. This leads to a much faster absorption/desorption rate of water molecules in IER than the other two P-100 samples, meaning the smaller T_{H_2O} value. The reaction rates of moisture-swing CO₂ absorbers were estimated similar.

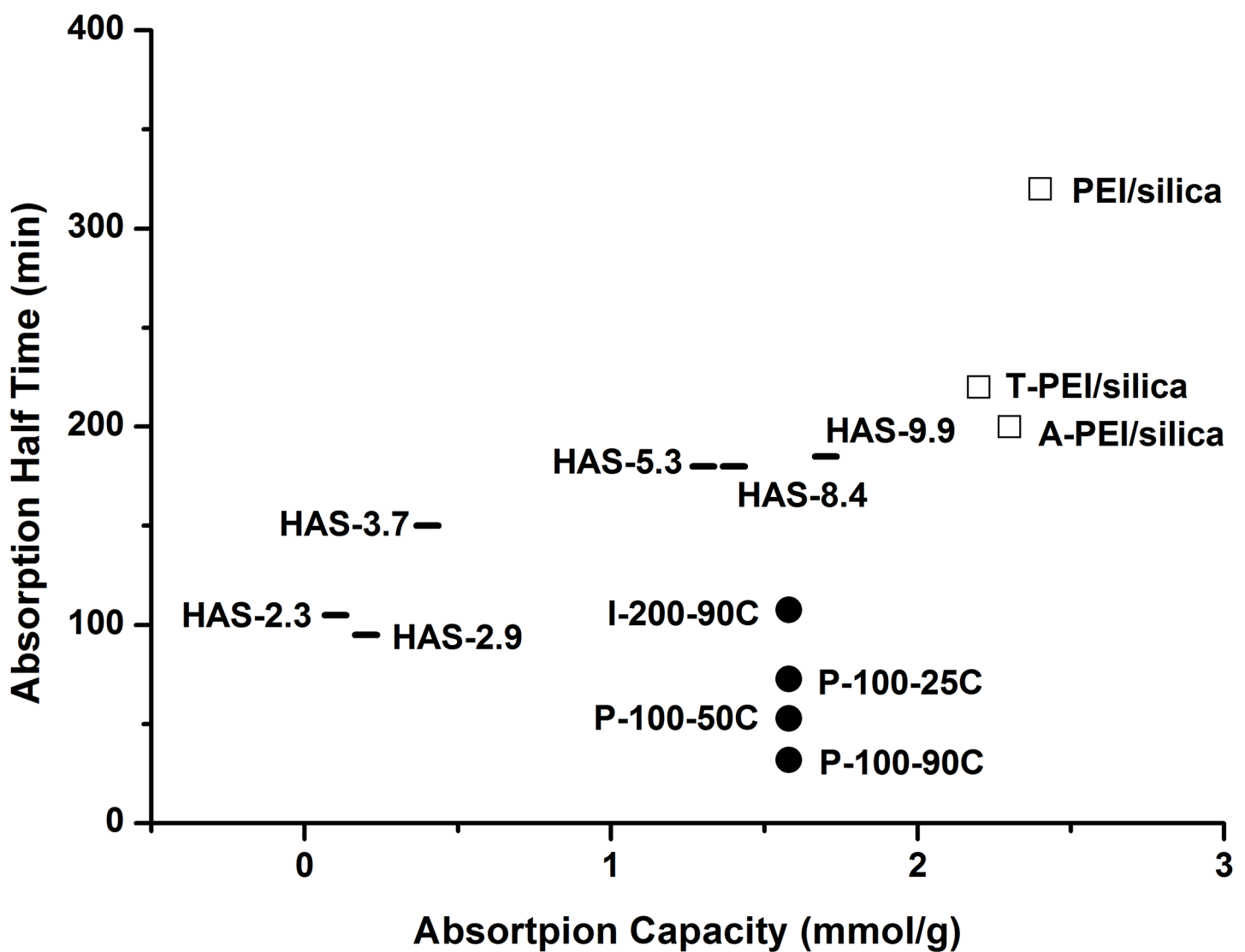


Fig 5. Comparison of CO₂ absorption half times and capacities of different sorbents. Hyperbranched aminosilica (HAS) with different amine loading (-), PEI/silica materials (□), and moisture-swing Ion Exchange Resin (IER) (●). The number of half time and absorption capacity of each adsorbent has been shown in S1 Table.

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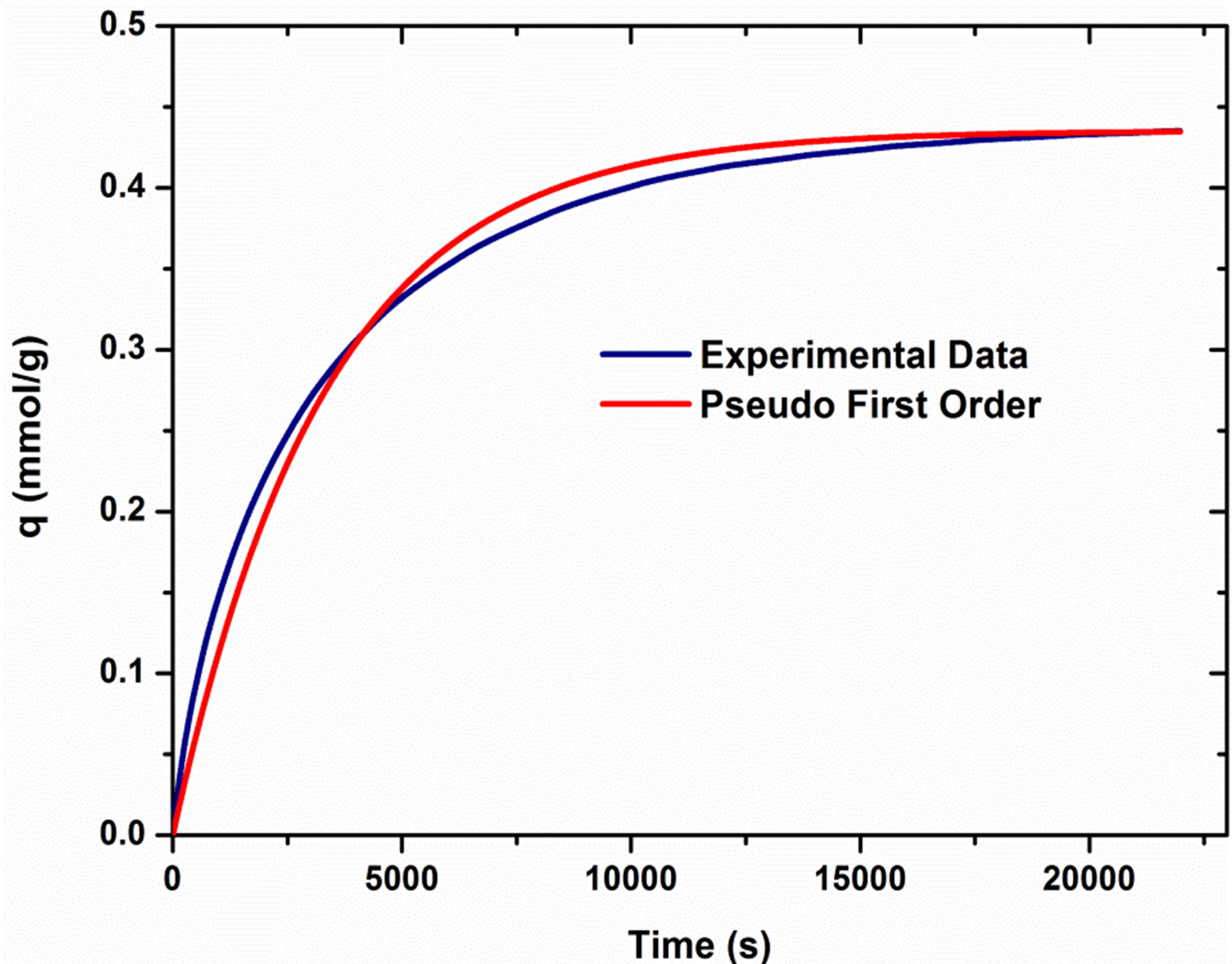


Fig 6. Comparison of kinetic model and experimental data for absorption performance of P-100-90C absorbent.

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I-200 moisture-swing CO₂ absorbent was also treated by 90°C hot water but still had a longer half time than those of three P-100 samples. The reason is the thickness of I-200 sorbent is 640 microns which is much thicker than the 100 microns thickness of P-100 sorbent. Plenty of time is consumed by water vapor to permeate into I-200 sorbent to contact the inside IER particles wrapped by polypropylene matrix binder.

Fig 6 shows that pseudo first order model as Eq 1 and 2 fits the absorption kinetic data of P-100-90C absorbent with the coefficient of determination 0.98. The amount of absorbed CO₂ by P-100-90C sorbent was recorded per second, and the k values were determined by Eq 2.

4.3 Desorption kinetic performance

Moisture-swing CO₂ absorbent can release CO₂ back to the air in a wet surrounding⁸. The desorption kinetic characteristics of the moisture-swing absorbents are mainly influenced by

the diffusion rates of H₂O and CO₂ in sorbent. This study focuses on analyzing the impact of these two factors on the desorption rate.

To determine the diffusion coefficients of water molecules in the four absorbents, the moisture uptake percentage was determined from the equation:

$$\text{Moisture Uptake}\% = \frac{M_t}{M_d} \times 100\% \quad (4)$$

Where M_t is the total amount of water content absorbed by sorbent samples at time t , M_d is the original weight of the dry samples. The diffusivity D was determined from the slope (K) of the initial linear region of the plot of the percentage moisture uptake $\frac{M_t}{M_d}$ versus \sqrt{t} curve [26, 27].

$$D = \frac{\pi}{16} \left(\frac{h}{M_\infty/M_d} \right)^2 K^2 \quad (5)$$

Where h is the thickness of the sample, t is exposure time and M_∞ is the maximum moisture gain.

Fig 7 illustrates the desorption process of the studied four absorbents. The four samples are comparable because they have the same weight of IER particles about 0.30g. The same weight of IER particles can maintain the equilibrium concentration of CO₂ is between 1900ppm and 2000ppm under the same condition of dew point 15°C in the experimental device, shown as Fig 3. The samples increasingly absorb water molecules over time, which leads to their weights increase to equilibrium values under a certain water vapor partial pressure. Absorbed water molecules are conducive to desorb CO₂ from the full-loaded absorbents. The released CO₂ from absorbent increases the CO₂ concentration in the experimental device. The coefficient of water diffusion, as well as the equilibrium time of T_w (the equilibrium time of absorbed H₂O by absorbent) and T_c (the equilibrium time of desorbed CO₂ by absorbent) of the four absorbents have been listed in Table 1.

Comparing Table 1 (A), (B) and (C), the diffusion rate of water is higher when sorbent is treated by higher temperature hot water. The higher diffusion rate is due to larger number of pores in PVC matrix, as well as larger narrow cavities between IER particles and PVC binder. These characteristics greatly promote the diffusion rates of H₂O and CO₂ in sorbents. Higher diffusion rate of water can help ions in the absorbents to move more quickly and help desorbed CO₂ to release back to air more rapidly. For the P-100-90C sorbent, the equilibrium time difference ΔT between T_w and T_c is smallest $\Delta T = 66s$. It means the desorbed CO₂ diffuses out of the sorbent costing 66s after water reaches to equilibrium in the sorbent. However, ΔT of P-100-25C is as long as 3428s. It means the released CO₂ is still trapped in sorbent for a certain amount of time.

Comparing (A) and (D), both sorbents had been treated by 90°C hot water. Though the diffusion coefficient of water in I-200 is much larger than the one of P-100-90C, the water equilibrium time of P-100-90C is much smaller than those of I-200 due to the different binder materials of two sorbents. The reason is hydrophilic polypropylene binder of I-200 may also contribute to the water diffusion than the hydrophobic PVC binder of P-100-90C. However, the polypropylene of 640 micron thickness costs more time for water diffusion than the 100 micron PVC. The polypropylene binder of I-200 (provided directly by Snowpure LLC, California) is very difficult to be thinned due to the essential characteristics of this polymer.

I-200 also needs longer time for CO₂ to release back to the ambient air. For the I-200 absorbent, the IER particles of 44~74µm diameters are inlayed in polypropylene of 640 micron thickness. The narrow, tortuous and long paths among resin particles and polypropylene are against to CO₂ diffusion. However, the same IER particles of 44~74µm diameters can almost

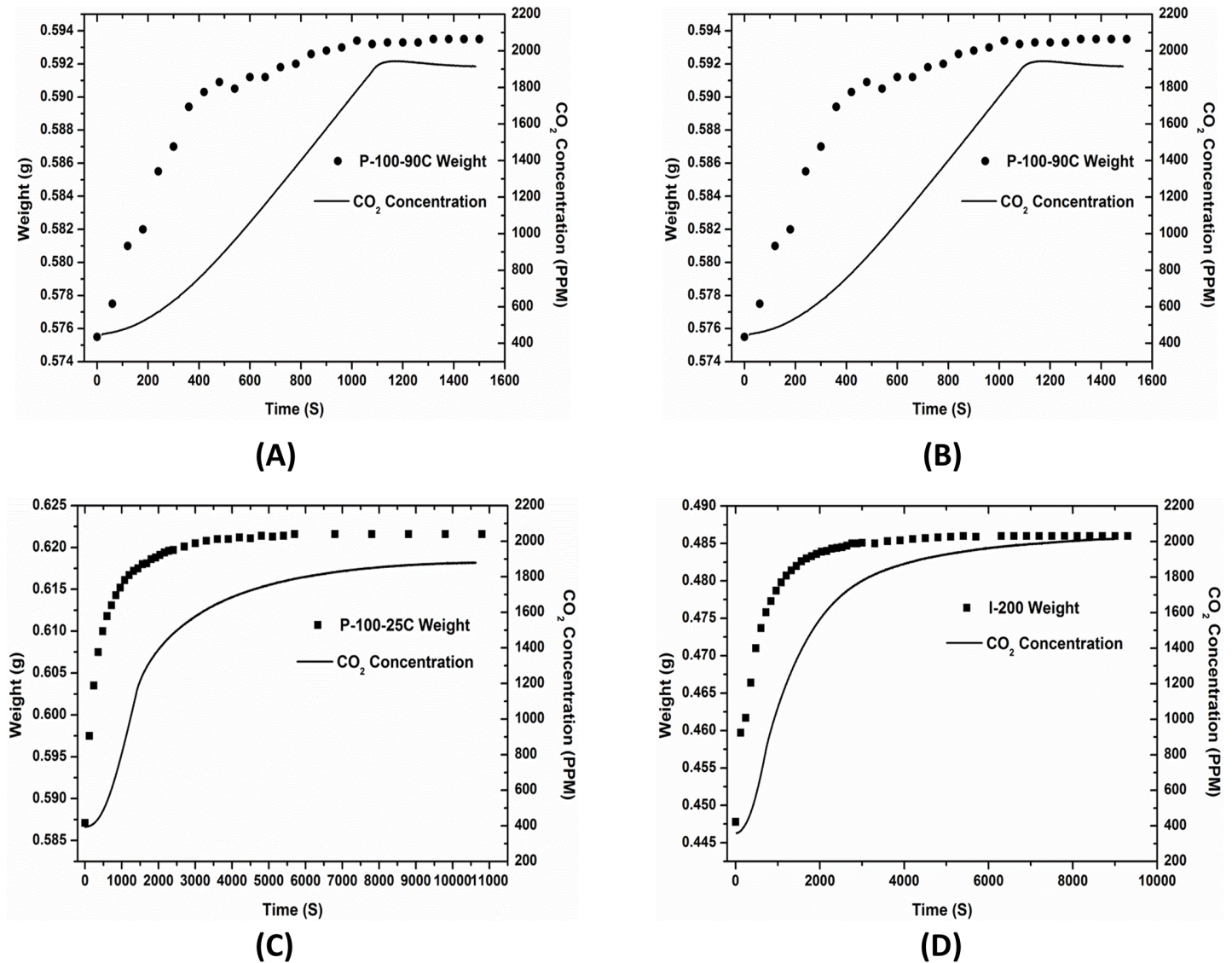


Fig 7. CO₂ desorption process of four absorbents (A) P-100-90C absorbent, (B) P-100-50C absorbent, (C) P-100-25C absorbent, (D) I-200 absorbent. Left Y-axis is absorbent weight, and right Y-axis is CO₂ concentration.

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penetrate the 100 micron PVC in P-100-90c sorbent. IER in P-100-90c sorbent can contact atmosphere directly is benefit to desorption performance. Hydrophilic polypropylene binder

Table 1. Diffusion coefficients of water, equilibrium times of water and CO₂ in four samples.

	D(m ² /s × E-12)	T _w (s)	T _c (s)	ΔT (s)
(A)P-100-90C	6.503	1020	1086	66
(B)P-100-50C	2.536	1740	2462	722
(C)P-100-25C	1.842	7200	10628	3428
(D)I-200	58.464	6300	8970	2670

D is the diffusion rate of H₂O; T_w is the equilibrium time of absorbed H₂O by absorbent; T_c the equilibrium time of desorbed CO₂ by absorbent. ΔT = T_c–T_w

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in I-200 can also attract some absorbed water molecules. This part of water molecules can not contact functional IER particles to release CO₂.

5. Conclusion

This study introduces a new moisture-swing CO₂ absorbent by employing polyvinyl chloride (PVC) as binder for ion exchange resin (IER). The manuscript analyzes the preparation process, absorbent structure, kinetic model, absorption and desorption characteristics of this CO₂ absorbent. The CO₂ absorption rate of this new produced absorbent P-100 is nearly three times as fast as the one of I-200, and also three to ten times as fast as amine-tethered solid CO₂ absorbents. This fast absorption/desorption rate is with the benefit of a thin bind holder and fast diffusion rate of H₂O. This new CO₂ absorbent provides a way of designing a moisture-swing CO₂ absorbent with a high absorption/desorption rate in the future.

Supporting information

S1 Table. Half time and absorption capacity of CO₂ absorbent.
(DOCX)

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Investigation: Xiaoyang Shi.

Methodology: Xiaoyang Shi, Tao Wang.

Project administration: Klaus S. Lackner.

Supervision: Tao Wang, Klaus S. Lackner.

Validation: Xiaoyang Shi.

Visualization: Xiaoyang Shi.

Writing – original draft: Xiaoyang Shi.

Writing – review & editing: Xiaoyang Shi, Tao Wang, Klaus S. Lackner.

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