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Research Article

Investigation of CO_2-Sorption Characteristics of Readily Available Solid Materials for Indoor Direct Air Capturing

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Direct air capturing (DAC) is an energy demanding process for CO_2-removal from air. Ongoing research focuses on the potential of indoor air as DAC-feed to profit from currently unused energetic synergies between DAC and the built environment. In this work, we investigated the performance of three different readily available, solid DAC-adsorbers under typical indoor environmental conditions of 16-25°C, 25-60% relative humidity (RH), and CO_2-concentrations of less than 800 ppm above atmospheric concentrations. The measured mass-specific CO_2-adsorption capacities of K_2CO_3-impregnated activated carbon, polyethylenimine-snow (PEI-snow), and polyethylenimine (PEI) on silica amount to $6.5 \pm 0.3\,mg\,g^{-1}$, $52.9 \pm 4.9\,mg\,g^{-1}$, and $56.9 \pm 4.2\,mg\,g^{-1}$, respectively. Among the three investigated adsorber materials, PEI on silica is the most promising candidate for DAC-applications as its synthesis is rather simple, the CO_2-desorption is feasible at moderate conditions of about 80°C at 100 mbar, and the competing co-adsorption of water does not strongly affect the CO_2-adsorption under the investigated experimental conditions.

1. Introduction

With a global demand for carbon embedded in organic chemicals and derived materials on the order of Mt yr^{-1} and a global consumption of fossil carbon on the order of Gt yr^{-1}, carbon plays a central role in the industrial value chain both for the production of bulk and fine chemicals and as energy carrier for incineration processes [1, 2]. Thus, carbon represents a cornerstone of the global economy [3]. However, because of increasing concerns related to global warming, the way of carbon utilization must radically change. Current industrial processes lead to enormous amounts of carbon being released into the atmosphere as carbon dioxide (CO_2). On a global scale, CO_2 impacts the Earth's radiative balance by absorbing radiation in the infrared region and is thereby the main course of man-made climate change. While carbon utilization allowed humanity to thrive for many centuries, it has now become one of the major environmental threats. Numerous national economies agreed on drastic measures to become climate neutral by 2050 [4]. Climate neutrality requires "net zero emissions." Net zero emissions must be achieved by an overall balance between CO_2-emissions and CO_2-sinks. Negative emissions, meaning the active removal of CO_2 from the atmosphere, are necessary and urgently needed to encounter climate change.

Negative emission technologies comprise natural and technical approaches. One natural pathway is global reforestation to enhance the CO_2-sequestration by photosynthesis [5, 6]. Another pathway resorts to CO_2-sequestration using crops or even algae and subsequent carbonization of the produced biomass [7, 8]. While undoubtedly necessary, these strategies alone are not capable of removing the vast amount of CO_2 present in the Earth's atmosphere [9]. Moreover, these approaches imply a target conflict regarding the land use for growing either food crops or fuel crops.

A further aspect to be tackled when encountering climate change is industry's immense carbon demand. While mankind might be able to become widely independent from carbon as energy carrier for stationary or mobile combustion processes under certain circumstances, the chemical

industry cannot simply abdicate carbon. Even in 2050, carbon will be required as chemical building block for the production of various chemicals, materials, and pharmaceutical products. In 2021, the production of global crude oil was 4.5 • 10^{12} kg [10]. Considering a simplified sum formula $(CH_2)_n$, this amounts to approximately 3.8 • 10^{12} kg carbon that needs to be saved or replaced in a defossilized scenario.

To date, two major technical pathways are discussed to ensure the carbon feedstock for the chemical and pharmaceutical industry. One technology is postcombustion capturing, where CO_2 is stripped from rich gas streams such as power plant exhausts or biogas. This process is relatively cost-effective due to the high CO_2-concentration in the exhaust streams (typically 5-20%) [11, 12]. However, the largest drawback is the decreasing availability of CO_2-rich exhaust streams in the future due to planned shutdowns of power plants operated with fossil fuels. Additionally, the increasing global demand for locally produced food crops as well as the necessity for land use planning to maintain biodiversity will compete with required land use for the production of fuel crops and thus with the production of biogas [13, 14]. The second technology is direct air capturing (DAC) which has been emerging in the past several years. The concept is to capture CO_2 directly from the atmosphere or air streams similar to atmospheric composition [15]. The major concern here is the high energy demand due to the fact that CO_2 is filtered at a very low concentration in the range of only 400-1000 ppm. However, several working groups were able to show that there is potential to lower the energy demand by either optimizing the capturing process itself or integrating it into a suitable environment [16–20]. To date, major limitations for using DAC on a large scale for CO_2-separation are large mass-specific energy demands of several MWh t-1/t CO_2 [21], large investment costs for CO_2-adsorber material, and the availability of suitable upstream and downstream technologies that create co-benefits by using DAC in sector-coupling applications.

In a preceding study, we investigated several potential benefits of incorporating DAC into heating, ventilation, and air conditioning systems (HVAC systems). We outlined that the building's energy demand can be significantly lowered if CO_2 is stripped from building exhaust air, followed by the partial recirculation of the CO_2-depleted and further purified air into the building. This lowers the energy demand for HVAC systems because less ambient air has to be conditioned to meet indoor requirements. We furthermore pointed out that purified recirculated air can be even of higher quality due to reduced intake of outdoor pollutants such as ozone, nitrogen oxides, volatile organic compounds, and particulate matter [18].

To take the next step towards realization of a HVAC/DAC-coupling in recirculation mode, we hereby present experimental findings on potential CO_2-adsorber materials for Indoor Direct Air Capturing (IDAC). The IDAC-technology comprises the adsorption of CO_2 in the built environment at conditions specified for the indoor environmental quality of category II laid down in the European Standard EN 16798-1 of 16-25°C, 25-60% relative humidity (RH), and CO_2-concentrations of less than 800 ppm above atmospheric concentrations [22]. Under these conditions, we measured the mass-specific adsorption capacity of selected adsorber materials. Further, we investigated the minimum desorption temperature at a predefined pressure which later is needed to estimate the mass-specific energy demand for CO_2-desorption. Complementary parameters such as co-adsorption of water, material handling, and material durability were also qualitatively assessed. Further process parameters for a reliable calculation of energy demands (adsorber heat capacity, adsorption/desorption enthalpy) were not assessed here.

2. CO_2-Adsorber Materials for IDAC

The CO_2-adsorber materials for IDAC investigated in this study were selected to enable a reversible CO_2-adsorption via a physical or chemical process. Furthermore, the adsorber materials must be readily available for the intended use. Three materials were chosen that fulfil the above-mentioned criteria: (i) K_2CO_3-impregnated activated carbon, (ii) cross-linked polyethylenimine (PEI-snow), and (iii) polyethylenimine supported on silica (PEI on silica).

2.1. K_2CO_3-Impregnated Activated Carbon. Activated carbon is a widely used material and outstanding adsorber due to its large surface area. In this study, we investigated a commercially available activated carbon impregnated with a mass fraction of 10% K_2CO_3, a basic salt that increases the ability to adsorb acidic gases such as CO_2. The activated carbon was received as a free sample from Carbotech AC GmbH. It is based on steam-activated mineral coal with a BET-surface of 1000 m^2.

2.2. Cross-Linked Polyethylenimine (PEI-Snow). Polyethylenimine (PEI) is a basic amine-based polymer, which is able to reversibly adsorb CO_2. However, pure PEI is a highly viscous liquid that needs to be further treated to function as a solid adsorber. Therefore, PEI (molecular weight = 25 kDa, branched, Merck) is cross-linked with 1,1,1-Tris-(hydroxymethyl)-propane-tris-(glycidylether) (TTE, Merck), an oligoepoxide that links the separated PEI-molecules, yielding solid PEI-snow. In this study, we synthesized PEI-snow with a mass fraction of 3 and 6% of TTE-cross-linker, respectively. The synthesis of cross-linked polyethylenimine, which appears as an aqueous gel, is described by Xu et al. [23].

2.3. Polyethylenimine Supported on Silica (PEI on Silica). An alternative to chemically cross-linking PEI is to impregnate it onto the surface of a carrier material. In this study, we used PEI (molecular weight = 25 kDa, branched, Merck) on silica with a mass fraction of 40% PEI. The synthesis of PEI on silica is described by Goeppert et al. [24].

3. Methodology

The performance of the CO_2-adsorber materials was investigated in a test chamber with a volume of (812 ± 10)L. The conditions for adsorption experiments were chosen to meet the specifications of the indoor environmental quality of category II laid down in the European Standard EN 16798-1 of

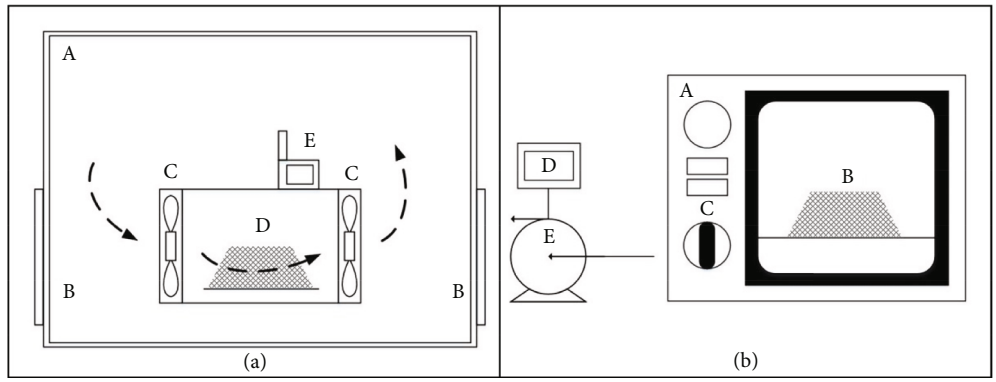

Figure 1: Adsorption setup (a) with (A) test chamber corpus, (B) sealable doors, (C) 200 mm fans, (D) adsorber material on paper tray, and (E) sensor for T, c_{CO2}, and RH. Dashed arrows indicate the circulating airflow. Desorption setup (b) with (A) vacuum drying cabinet, (B) adsorber material on steel tray, (C) vacuum valve, (D) vacuum control unit, and (E) membrane vacuum pump.

16-25°C, 25-60% RH, and CO_2-concentrations of less than 800 ppm above atmospheric concentrations [22]. Most of the experiments presented in the following were conducted under these boundary conditions with few marginal exceptions. At the beginning of each adsorption experiment, the test chamber was conditioned to a defined CO_2-level using CO_2 resulting from paraffin incineration. Subsequently, the CO_2-adsorber material was exposed to the chamber's atmosphere in a ventilation channel using two electric fans with an electrical power of 2 W and a diameter of 200 mm, each. Figure 1(a) shows the experimental setup for the adsorption experiments.

All adsorption experiments were carried out for a minimum of three hours after which all materials reached a steady state. CO_2-concentration, relative humidity, and temperature were recorded using CO_2-sensors (CL-11 NDIR, Rotronic). The CO_2-sensors have a declared accuracy of ±20 ppm and are equipped with an integrated capacitive hygrometer for the measurement of the relative humidity with a declared accuracy of <2.5% as well as a thermistor for the temperature measurement with a declared accuracy of ±0.3 K.

The CO_2-adsorption capacity was calculated exclusively based on CO_2-concentrations measured in the test chamber according to

$$\Theta_{CO_2} = \frac{(c_0 - c_1 - c_{leak}) \bullet \rho_{CO_2} \bullet V}{m_{ads}}. \quad (1)$$

Θ_{CO2} denotes the CO_2-adsorption capacity in mg g^{-1} of the respective material (mg CO_2 per g adsorber material).

c_0 is the initial CO_2-concentration in ppm when the adsorber is first exposed to the test atmosphere.

c_1 is the final CO_2-concentration in ppm after 3 hours of adsorber exposure.

c_{leak} is the CO_2-concentration loss in ppm in the test chamber caused by leakages, calculated using Equation (3).

ρ_{CO2} is the density of CO_2 under STP conditions (1.98 kg m^{-3}).

V is the test chamber's volume of 0.812 m^3 (geometrically calculated, ±0.01 m^3).

m_{ads} is the mass of adsorber used for the experiment.

The test chamber showed an unavoidable leakage, which was considered in the data evaluation. Therefore, some experiments were prolonged overnight. The steady decline of the recorded CO_2-concentration after achieving adsorber saturation was used to calculate the leakage for the experiment using

$$k_{leak} = \frac{\ln (c_3 - c_{out}/c_2 - c_{out})}{t_{end} - t_{start}}. \quad (2)$$

k_{leak} is the leakage rate of the test chamber in h^{-1}.

c_2 is the CO_2-concentration for the leakage measurement in ppm, taken 4 hours after initial adsorber exposition and therefore at least one hour after adsorber saturation is achieved.

c_3 is the final CO_2-concentration in ppm after the leakage experiment's duration of 6 hours.

c_{out} is the CO_2-concentration outside the test chamber in ppm. This value is recorded at the beginning of each experiment and assumed to be constant.

t_{start} is the starting time for determination of the leakage rate, recorded 4 hours after initial adsorber exposition.

t_{end} is the end time for determination of the leakage rate, recorded 10 hours after initial adsorber exposition.

Solving Equation (2) for c_2 and subtracting c_3 yield Equation (3) where c_{leak} denotes the CO_2-concentration loss in ppm in the test chamber caused by leakages. c_{leak} is needed for leakage correction of Equation (1).

$$c_{leak} = c_2 - c_3 = (c_3 - c_{out}) \bullet e^{[-k_{leak} \bullet (t_{end} - t_{start})]} + c_{out} - c_3. \quad (3)$$

During the adsorption experiments, the leakage rate k_{leak} was individually calculated eight times. The mean value was found to be $k_{leak} = (0.015 \pm 0.01)\ h^{-1}$. This value was used as a constant leakage factor for all adsorber performance corrections in this study. The mean value of $k_{leak} = 0.015\ h^{-1}$ was used for the determination of c_{leak} according to Equation (3). We found that the absolute decrease in CO_2-concentration due to passive leakage was 24.1 ± 1.1 ppm, which is on the order of the declared sensor accuracy of ±20 ppm.

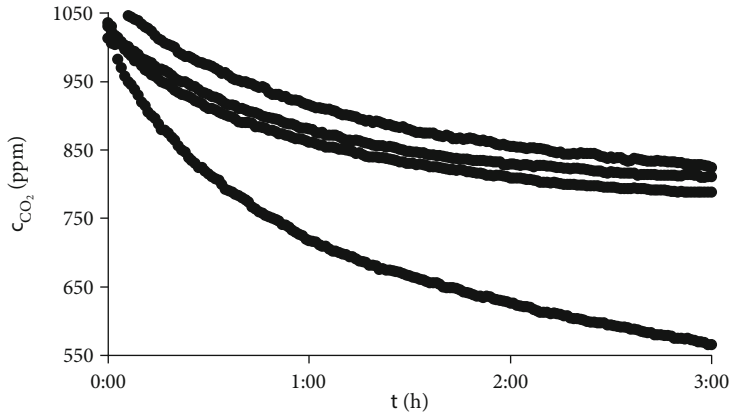

FIGURE 2: Exemplary CO_2-adsorption curves for each investigated material. From top to bottom: PEI-snow with a mass fraction of 3% TTE, PEI-snow with a mass fraction of 6% TTE, PEI on silica, and K_2CO_3-impregnated activated carbon. The absolute adsorber performance is no indicator for adsorption capacity as different adsorber masses were used for activated carbon and PEI-based materials, respectively.

The minimum regeneration temperature for the materials was investigated in desorption experiments to achieve complete regeneration when held at this temperature for one hour at 100 mbar. The parameter set consisting of pressure, temperature, and time for regeneration can be further optimized. This issue has not been assessed systematically in this study. Figure 1(b) shows the experimental setup for the desorption experiments. Pressure and regeneration time were kept constant. Pressures below 100 mbar greatly increase the vacuum pump's energy demand and require more effort in future machine design to achieve the necessary tightness. A longer regeneration time increases the thermal stress on the adsorber material, which might affect the durability.

The three materials were compared and evaluated towards their potential for IDAC-applications. Furthermore, potential degradation of the materials due to the thermal stress was qualitatively assessed. Therefore, the CO_2-adsorption capacity was compared over all regeneration cycles in which the same batch of the material was used. The mass of the adsorber used for the experiments was selected in a way that the expected CO_2-adsorption remains at around 40% (K_2CO_3-impregnated activated carbon) or 20% (PEI-snow and PEI on silica) of the initial CO_2-mass in the chamber.

4. Results and Discussion

Figure 2 shows exemplary CO_2-adsorption curves for each investigated material. Batches of 6 g material were used of PEI-snow and PEI on silica, respectively. K_2CO_3-impregnated activated carbon was used in a 100 g batch.

All materials show a comparable adsorption behavior. When exposed to a CO_2-enriched atmosphere, the material adsorbs CO_2, resulting in a rapid decline of the CO_2-concentration. In the course of the adsorption experiment, the material becomes saturated with CO_2 while the gas-phase concentration of CO_2 is reduced. Both effects result in reduced CO_2-uptake over time until a steady state is reached. At this point, the measurements start to show a constant decline solely caused by the test chamber's leakage. Figure 3 shows a slow and steady decline of the CO_2-concentration in the test chamber exclusively caused by leakage.

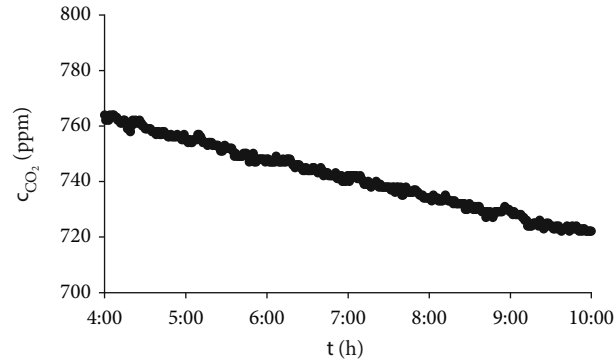

FIGURE 3: Exemplary decay of the CO_2-concentration exclusively caused by passive leakage observed 4 to 10 hours after initial adsorber exposition. The regression analysis of the experimental data using Equation (2) yields a leakage rate of $k_{leak} = 0.015\,h^{-1}$ with a determination coefficient of $R^2 = 0.993$. The data set shown here was recorded after a regular adsorption experiment with 6 g PEI on silica.

In the following, we present and discuss the experimental results for each investigated CO_2-adsorber material for IDAC. The dependence of the CO_2-adsorption capacity on the initial CO_2-concentration was not systematically investigated in this study.

4.1. K_2CO_3-Impregnated Activated Carbon. In general, activated carbon is a robust material. The CO_2-adsorption-performance was found to be consistent over the range of experiments. The necessary regeneration temperature at 100 mbar over one hour was found to be 120°C. Adsorber regeneration at 100°C leads to a steady decline in CO_2-adsorption capacity over three cycles. In contrast, regeneration at 120°C restored the material's initial CO_2-adsorption capacity (cycle 5 in Table 1). In addition to CO_2, activated carbon co-adsorbs water as seen in decreasing relative humidity during the experiments. However, no correlation between air humidity and CO_2-adsorption capacity was found under the prevailing experimental conditions ranging from 42 to 66% relative humidity over a set of 10 adsorption/

Table 1: Experimental results for CO_2-adsorption on K_2CO_3-impregnated activated carbon. Experiments shown here comprise desorption at 120°C, with the exception of cycle 1, which was initially treated at 100°C before first adsorption.

Starting conditions			End conditions			Capacity[1]	Cycle[2,3]	Capacity corrected[1,4]	Standard dev.[1]
CO_2 (ppm)	RH (%)	T (°C)	CO_2 (ppm)	RH (%)	T (°C)	(mg g^{-1})	#	(mg g^{-1})	(mg g^{-1})
1018	58.7	24.5	552	54.5	24.2	7.5	1	7.0	
1069	56.9	25.2	660	46.9	25.4	6.6	5	6.1	0.3
1069	66.0	26.2	651	51.7	25.9	6.7	6	6.2	
1044	51.6	23.8	596	44.6	24.3	7.2	7	6.7	
1055	54.6	23.3	594	44.7	24.4	7.4	8	6.9	
1076	47.7	24.3	625	43.5	24.7	7.3	9	6.8	Arithm. mean[1]
1021	42.0	25.2	612	40.1	25.9	6.6	10	6.1	(mg g^{-1})
1070	43.8	23.3	655	38.5	23.9	6.7	11	6.2	
1013	60.7	23.1	565	44.9	23.8	7.2	12	6.8	6.5
1050	52.2	24.1	615	42.7	24.7	7.0	13	6.5	

[1]CO_2-adsorption capacity given in milligram CO_2 per gram adsorber material. [2]Number of adsorption/desorption cycles for this material batch. [3]Missing cycle numbers are not listed here because the corresponding experiments were conducted with different CO_2-concentrations and/or humidity. [4]CO_2-adsorption capacity corrected for leakage according to Equation (1).

Table 2: Experimental results for CO_2-adsorption on PEI-snow with a mass fraction of 3% and 6% TTE, respectively. Experiments shown here comprise desorption at three different temperatures.

	Starting conditions			End conditions			Reg. temp.	Capacity[1]	Cycle[2,3]	Cap. corr.[1,4]	Standard dev.[1,6]
	CO_2 (ppm)	RH (%)	T (°C)	CO_2 (ppm)	RH (%)	T (°C)	(°C)[5]	(mg g^{-1})	#	(mg g^{-1})	(mg g^{-1})
	1066	50.0	22.3	816	70.0	22.1	60	67.1	1	58.9	
	1090	44.3	22.1	872	66.5	23.3	60	52.9	4	44.3	
3% TTE	1073	51.2	22.5	883	69.6	22.9	80	43.5	5	35.2	4.9
	1076	66.8	23.7	845	75.7	23.3	100	53.7	6	45.3	
	1081	63.0	23.5	824	76.9	23.1	100	58.8	8	50.3	
	1079	57.4	25.2	718	61.1	24.5	100	66.5	2	58.1	Arithm. mean[1,6]
6% TTE	1035	68.3	24.3	807	74.2	25.0	100	60.1	3	52.3	(mg g^{-1})
	1063	59.0	24.0	815	70.0	24.3	100	66.6	4	58.4	52.9

[1]CO_2-adsorption capacity given in milligram CO_2 per gram adsorber material. [2]Number of adsorption/desorption cycles for this material batch. [3]Missing cycle numbers are not listed here because the corresponding experiments were conducted with different CO_2-concentrations and/or humidity. [4]CO_2-adsorption capacity corrected for leakage according to Equation (1). [5]Temperature used to regenerate material at 100 mbar for 1 hour prior to adsorption experiment. [6]Standard deviation and arithmetic mean calculated only for experiments with adsorber fully regenerated at 100°C.

desorption cycles. The acquired data is summarized in Table 1.

K_2CO_3-impregnated activated carbon shows a comparably (see Table 2 and Table 3) low adsorption capacity for CO_2 of 6.5 ± 0.3 mg g^{-1}. However, it is a commercially available product. An increased mass fraction of K_2CO_3 might even lead to an increased CO_2-adsorption capacity. Additionally, activated carbon can be yielded from renewable resources such as coconut shells [25, 26]. Furthermore, activated carbon is capable of co-adsorbing water. Obviously, this will increase the energy demand for regeneration since water has to be co-evaporated during the regeneration process. However, co-adsorption of water might be beneficial to lower the additional effort for air dehumidification without additional installations in HVAC systems. Moreover, activated carbon might yield additional co-benefits as it is expected to remove further airborne trace constituents such as particulate matter, volatile organic compounds, and odorous substances. Both air dehumidification and further air purification are process steps in indoor air treatment that often become necessary in partially recirculating HVAC systems. Therefore, impregnated activated carbon can be regarded as a promising material for IDAC.

4.2. PEI-Snow. PEI-snow adsorbed on average 52.9 mg CO_2 per g adsorber with a standard deviation of 4.9 mg g^{-1}. A correlation between adsorption capacity and TTE-concentration could not be found.

PEI-snow cross-linked with a mass fraction of 3% TTE was tested over several cycles. It was found that the necessary regeneration temperature was 100°C. If regenerated with lower temperature, the CO_2-adsorption capacity steadily decreases as shown in Table 2 (cycles 1-5). However, the regeneration at 100°C at 100 mbar for 10 minutes resulted in a significant color change of the material from white to yellow. Further regeneration cycles at 100°C

Table 3: Experimental results for CO_2-adsorption on PEI on silica. Experiments shown here comprise desorption at 80°C.

Starting conditions			End conditions			Capacity[1]	Cycle[2,3]	Capacity corrected[1,4]	Standard dev.[1]
CO_2 (ppm)	RH (%)	T (°C)	CO_2 (ppm)	RH (%)	T (°C)	(mg g^{-1})	#	(mg g^{-1})	(mg g^{-1})
1010	55.2	24.1	763	55.8	24.6	66.3	1	58.9	
1010	48.4	23.1	750	50.5	24.0	69.8	3	62.4	4.2
1031	49.2	25.2	788	48.3	25.7	65.2	6	57.5	
1050	49.4	25.6	821	47.2	26.9	61.5	7	53.5	
1019	57.5	23.9	763	56.4	24.8	68.7	8	61.2	
1002	54.4	25.3	788	54.5	26.1	57.4	9	50.2	Arithm. mean[1]
1067	56.7	25.6	837	54.4	26.9	61.7	10	53.5	(mg g^{-1})
1031	57.1	25.7	775	55.4	25.8	68.7	11	61.0	
1019	64.1	24.9	802	58.3	25.3	58.2	12	50.7	
1061	41.2	25.8	821	43.9	26.2	64.4	17	56.3	56.9
1018	45.0	24.6	763	51.7	25.1	68.4	22	60.9	

[1]CO_2-adsorption capacity given in milligram CO_2 per gram adsorber material. [2]Number of adsorption/desorption cycles for this material batch. [3]Missing cycle numbers are not listed here because the corresponding experiments were conducted with different CO_2-concentrations and/or humidity. [4]CO_2-adsorption capacity corrected for leakage according to Equation (1).

resulted in an intensive orange color. Oxidative decomposition even under reduced pressure of 100 mbar is observed. Orange color might be caused by the formation of nitrogen oxides, potentially through oxidation of amine groups. The loss of the amine groups is expected to lead to the deterioration of CO_2-adsorption capacity.

The same effect was observed when regenerating PEI-snow with a mass fraction of 6% TTE at 100°C. The material was furthermore found to release a significant amount of water due to evaporation during the adsorption experiments as well as during the regeneration at 100°C. It was not effective to use regenerated and therefore drier material since it showed a significantly reduced CO_2-adsorption capacity and was not able to resorb water from the test chamber's atmosphere to rehydrate itself. To use the material effectively, it was first regenerated, and distilled water was then added to rehydrate the polymer prior to use for adsorption. The water lost during adsorption results in a significantly increased air humidity within the test chamber as depicted in Table 2.

The necessary regeneration temperature of 100°C and thus the induced thermal stress and inevitable degeneration of the adsorber material hamper a potential usage of PEI-snow for IDAC. To be able to use this material, it might be viable to use hot steam instead of dry air and reduced pressure for regeneration as suggested by Xu et al. [23]. If such regeneration is feasible under certain conditions, PEI-snow might be a promising material that is easily synthesized without waste products.

4.3. PEI on Silica. PEI on silica showed comparable CO_2-adsorption capacities as PEI-snow. The material adsorbed on average 56.9 mg CO_2 per g adsorber with a standard deviation of 4.2 mg g^{-1}. However, the material had a smaller influence on air humidity than the other investigated materials.

PEI on silica regenerated at 80°C at 100 mbar for one hour. Under these conditions, no visible degradation occurred during the first ten cycles. Upon further experimenting, a pale yellow color was observed. This might indicate a slow formation of oxidized PEI on the material's surface. However, the material's performance did not significantly change over 22 cycles.

In the range of relative humidity between 40 and 60%, no significant correlation between CO_2-adsorption capacity and relative humidity was found. However, increasing humidity exceeding 75% led to reduced CO_2-adsorption capacities in both investigated scenarios with 1000 and 2000 ppm initial CO_2-load, respectively.

4.4. Comparison of Experimental Results to CO_2-Adsorption Capacities Reported in Literature. CO_2-adsorbers are often grouped for either DAC-application in atmospheric air (about 400 ppm CO_2) or in a simulated flue gas (5-15% CO_2). In this study, we report CO_2-adsorption capacities determined at initial CO_2-concentrations of about 1100 ppm. Zhang et al. and Leonzio et al. [27, 28] published a list of CO_2-adsorption capacities for their own and further materials based on PEI or metal organic frameworks (MOFs). The reported mass-specific CO_2-adsorption capacities range between 3 and 7% for atmospheric air and 6-11% for simulated flue gas. The adsorption capacities found in this work for PEI-based materials fit to the data for atmospheric air capturing. Activated carbon is regularly investigated to capture CO_2 in flue gases or even pure CO_2 atmospheres [29–31]. Mass-specific CO_2-adsorption capacities of 5-10% in pure CO_2 are documented. However, considering the CO_2-concentration difference of at least two orders of magnitude as well as the co-adsorption of other species (like, e.g., N_2), this data is not transferable to a DAC-case as investigated here where we documented a mass-specific CO_2-adsorption capacity of around 0.7%. Not addressed in this paper, but promising anyway is the material class of MOFs, which have been intensively investigated in the last decade. The flexibility in chemical composition and pore properties renders MOFs to be promising candidates for future DAC applications. However, some basic issues are yet to be overcome such as costs, durability, potential health issues, and tolerance towards humidity [32–34].

4.5. General Findings. According to the conducted experiments, we consider PEI on silica to be the most suitable candidate for DAC in the built environment. Looking at necessary chemicals and industrial scale production, PEI is expected to be more expensive than activated carbon in the near future. However, it provides the best combination of durability and CO_2-capacity under the experimental conditions of this study. The synthesis is rather simple and does not require any complex technical equipment. The necessary regeneration temperature of 80°C is also comparably low and thereby offers potential to reduce the overall energy demand of the IDAC-process. In our setup, we regenerated the loaded adsorber in a vacuum drying cabinet where the adsorber is initially exposed to increased oxidative stress until the reduced desorption pressure of 100 mbar is reached. If the reduced pressure was applied prior to heating or the remaining oxygen was additionally purged out of the vacuum drying cabinet, this oxidative stress might be suppressed in future applications. Alternatively, regeneration time or pressure might be altered to achieve a more sensitive regeneration and thereby increasing the material's durability further. K_2CO_3-impregnated activated carbon might play a valuable role in future IDAC-applications as additional adsorber or even additive to use its air-cleaning and moisture-capturing behavior.

5. Outlook

There are several additional parameters, which have to be evaluated to assess a material's potential for IDAC. Not only affordability and durability have to be suitable for an endured application. The material's emission behavior has to be known if air is to be recirculated back into a building. This includes the emission of volatile organic compounds, particles, or odors from the material itself or secondary pollutants that might originate from chemical reactions on the material's surface. Additionally, microbial infestation must be prevented, especially when hygroscopic materials are used. Another important issue is the regulatory aspect of realizing IDAC-installations. Indoor air quality is generally evaluated based on its CO_2-content. IDAC, however, actively reduces the CO_2-concentration. This might lead to the false assumption of high indoor air quality, although it is in fact loaded with secondary pollutants such as particles, microbial constituents, or volatile organic compounds. To ensure a hygienically harmless indoor air quality, alternative parameters for assessing indoor air quality have to be implemented. Most likely, further air purification technologies have to be used in addition to IDAC to ensure high indoor air quality in a partially recirculated system and to avoid accumulation of pollutants. The evaluation of physical properties and emissions will be subject of further investigations.

Furthermore, IDAC is an energy demanding process. While a certain amount of energy can be saved by recirculating already properly conditioned air back into a building and CO_2 is yielded as valuable product, both factors are currently unlikely to overcome the needed amount of energy for IDAC [18]. It is therefore important to reduce the processes' energy demand by using efficient materials and reactor setups. This includes compact packaging and realizing low pressure drops for ventilated air as well as an efficient heating of the material itself. Furthermore, a material with low heat capacity would be favorable to reduce the amount of energy needed to heat it up to its regeneration temperature. Beside the process itself, the main energy input has to be optimized. While electricity from the public grid is expensive, on-site generated electricity from solar power or nearby wind turbines might be significantly cheaper [35]. Additionally, waste heat from other processes could be used to preheat the adsorber material for regeneration.

We come to the conclusion that there is indeed potential usage of already available and comparably affordable CO_2-adsorber materials for IDAC. However, any IDAC-process needs to be optimized towards the active adsorber material as well as to the application, especially when an implementation in the built environment is planned.

Data Availability

Data is available on request.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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