

NOVEL K₂CO₃-BASED COMPOSITE SORBENT FOR CO₂ CAPTURE FROM AMBIENT AIR

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E-mail: jvv@catalysis.ruНОВЫЙ КОМПОЗИТНЫЙ СОРБЕНТ НА ОСНОВЕ K₂CO₃ ДЛЯ ПОГЛОЩЕНИЯ
УГЛЕКИСЛОГО ГАЗА ИЗ ВОЗДУХА

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Аннотация. Сорбционные свойства композитного сорбента K₂CO₃/активированный уголь были изучены в циклическом процессе при чередовании стадий сорбции CO₂ из воздуха и термической регенерации сорбента. Установлено, что значения абсорбционной емкости по диоксиду углерода существенно зависят от относительной влажности воздуха. Показано, что композитный сорбент может быть эффективно регенирован при нагреве до 150 °С, а при повышении температуры регенерации до 200 °С начинается процесс окисления углеродного материала кислородом воздуха.

Introduction. It is well known that CO₂ is the major anthropogenic greenhouse gas, which contributes to global climate change. Potassium carbonate is a solid inorganic chemisorbent, which reacts with atmospheric CO₂ in the presence of water vapor forming potassium bicarbonate: $K_2CO_3 + H_2O + CO_2 \rightarrow 2KHCO_3$. However, bulk potassium carbonate is not widely used as a material for CO₂ capture due to low reaction rate and insufficient mechanical strength. These problems can be solved by dispersing K₂CO₃ in pores of a support material. Recently, it was shown that K₂CO₃/γ-Al₂O₃ composite is a promising material for absorbing CO₂ directly from ambient air [1]. However, this material needs to be heated up to 300 °C in order to be fully regenerated, unlike bulk KHCO₃, which decomposes around 130 °C releasing CO₂ and H₂O. K₂CO₃/γ-Al₂O₃ needs higher regeneration temperature due to interaction between the active component and the porous support, which results in formation of potassium dawsonite KAlCO₃(OH)₂ [1, 2]. Increase in the regeneration temperature raises energy costs and narrows the range of heat sources that can be used. Therefore, there is a need for developing new sorbents for CO₂ absorption from ambient air, which can be effectively regenerated at T=150-200 °C. To solve this problem, it is necessary to select a porous support that does not interact with potassium carbonate. Other widely used porous oxides, such as SiO₂, MgO, TiO₂, do not fit, because they also react with K₂CO₃, forming either mixed oxides or mixed carbonates [2]. Activated carbons (AC), on the other hand, are considered to be perspective porous supports for K₂CO₃, but the major concern is a possibility of carbon material oxidation/destruction upon heating in air. Thus, this work is focused on performance of K₂CO₃/AC material in temperature-swing absorption cycles with regeneration temperatures of 150 and 200 °C. In order to separate KHCO₃ decomposition and carbon oxidation processes, both of which result in CO₂ release upon heating, the composite sorbent regeneration was carried out in both oxidative atmosphere (air) and inert atmosphere (argon).

Experimental. Composite sorbent K_2CO_3/AC was prepared by dry impregnation method, described in detail in [1]. Granules of a mesoporous active carbon AG-3 ($S_{BET} = 860 \text{ m}^2/\text{g}$, $V_{\text{pore}} = 0.5 \text{ cm}^3/\text{g}$) were filled with 40 wt. % aqueous solution of K_2CO_3 , followed by drying at 100°C for 12 h and then at 200°C for 1 h. Estimated K_2CO_3 content in the resulted composite material is 14 wt. %.

Processes of carbon dioxide absorption from ambient air and consecutive thermal desorption was studied in a temperature-swing adsorption cycles using an experimental set-up, schematically presented in Fig. 1. The composite sorbent was placed into a cylindrical fixed bed adsorber with the inner diameter of 19 mm, which was located inside an electrical heater. The mass of the composite sorbent inside the adsorber was 2 g. A single TSA cycle comprised 3 steps: 1) CO_2 absorption from ambient air for 2 h; 2) thermal desorption of CO_2 for 2 h; 3) the adsorber cooling for 2 h. The experimental conditions are summarized in Table 1. During the first step of each TSA cycle the composite sorbent was saturated with CO_2 as indoor air with was pumped through the adsorber using a gas pump. Relative humidity of the inlet air was 7 - 25 %. During the regeneration step the adsorber was rapidly heated up to the preset temperature (150 or 200°C), while the inlet flow rate of air or argon was maintained at $50 \text{ mL}/\text{min}$. Outlet concentrations of CO_2 were measured using a NDIR CO_2 sensor.

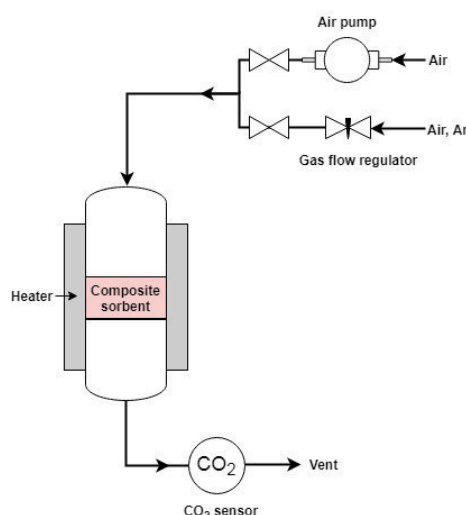


Fig. 1. The experimental set-up for performing TSA tests

Table 1

Parameters of TSA cycles

№	Description of a step	Parameter	Value
1	CO_2 absorption from air	Duration	2 h
		Gas flow	Air, $1300 \text{ mL}/\text{min}$
		Adsorber temperature	30°C
2	CO_2 desorption	Duration	2 h
		Gas flow	Air or Ar, $50 \text{ mL}/\text{min}$
		Adsorber temperature	$30^\circ\text{C} \rightarrow 150; 200^\circ\text{C}$
3	Cooling	Duration	2 h
		Gas flow	No flow
		Adsorber temperature	$150; 200^\circ\text{C} \rightarrow 30^\circ\text{C}$

Results and Discussion. It was shown that amount of CO₂ desorbed at 150°C in either argon or air flow strongly depended on relative humidity (RH) of indoor air during the previous CO₂ absorption step (Fig. 2). The maximal CO₂ uptake was obtained when RH was 12-14 %. Increase of regeneration temperature up to 200°C in argon atmosphere does not lead to any additional desorption of CO₂. However, when the material was heated up to 200°C in air flow, the amount of released CO₂ was significantly higher than the previous results, which is likely to be a result of carbon support oxidation by oxygen containing in air.

It should be noted that K₂CO₃ utilization extent in the TSA cycles is < 30%. XRD analysis showed that the composite sorbent K₂CO₃/AC after 12 h of CO₂ absorption from ambient air contains crystalline phases of KHCO₃ and K₂H₄(CO₃)₃·1.5H₂O, which means that even after much longer CO₂ absorption step this material absorbs less than 1 mol of CO₂ per 1 mol of K₂CO₃.

Conclusions. The obtained results show that the composite material can be effectively regenerated at 150°C in air flow, but the major drawback is that K₂CO₃ does not fully convert to KHCO₃ during the CO₂ absorption step. Further research is needed to improve performance of K₂CO₃/AC materials in the process of CO₂ absorption from ambient air.

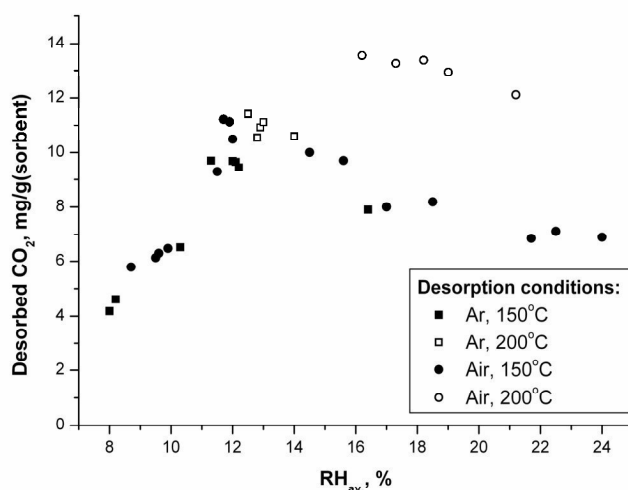


Fig. 2. Amount of desorbed CO₂ (in mg per 1 g of the composite sorbent) depending on average relative humidity throughout the CO₂ absorption step and conditions of the CO₂ desorption step (gas-carrier: argon or air; temperature: 150 °C or 200 °C)

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