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

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## НОВЫЙ КОМПОЗИТНЫЙ СОРБЕНТ НА ОСНОВЕ К₂СО₃ ДЛЯ ПОГЛОЩЕНИЯ УГЛЕКИСЛОГО ГАЗА ИЗ ВОЗДУХА

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**Аннотация**. Сорбционные свойства композитного сорбента  $K_2CO_3$ /активированный уголь были изучены в циклическом процессе при чередовании стадий сорбции  $CO_2$  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  $150\,$ °C, а при повышении температуры регенерации до  $200\,$ °C начинается процесс окисления углеродного материала кислородом воздуха.

**Introduction.** It is well known that CO<sub>2</sub> is the major anthropogenic greenhouse gas, which contributes to global climate change. Potassium carbonate is a solid inorganic chemisorbent, which reacts with atmospheric  $CO_2$  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 CO<sub>2</sub> capture due to low reaction rate and insufficient mechanical strength. These problems can be solved by dispersing K<sub>2</sub>CO<sub>3</sub> in pores of a support material. Recently, it was shown that K<sub>2</sub>CO<sub>3</sub>/γ-Al<sub>2</sub>O<sub>3</sub> composite is a promising material for absorbing CO<sub>2</sub> directly from ambient air [1]. However, this material needs to be heated up to 300°C in order to be fully regenerated, unlike bulk KHCO<sub>3</sub>, which decomposes around 130°C releasing CO<sub>2</sub> and H<sub>2</sub>O. K<sub>2</sub>CO<sub>3</sub>/γ-Al<sub>2</sub>O<sub>3</sub> needs higher regeneration temperature due to interaction between the active component and the porous support, which results in formation of potassium dawsonite KAlCO<sub>3</sub>(OH)<sub>2</sub> [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 SiO<sub>2</sub>, MgO, TiO<sub>2</sub>, do not fit, because they also react with K<sub>2</sub>CO<sub>3</sub>, forming either mixed oxides or mixed carbonates [2]. Activated carbons (AC), on the other hand, are considered to be perspective porous supports for K<sub>2</sub>CO<sub>3</sub>, but the major concern is a possibility of carbon material oxidation/destruction upon heating in air. Thus, this work is focused on performance of K<sub>2</sub>CO<sub>3</sub>/AC material in temperature-swing absorption cycles with regeneration temperatures of 150 and 200°C. In order to separate KHCO<sub>3</sub> decomposition and carbon oxidation processes, both of which result in CO<sub>2</sub> 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 m<sup>2</sup>/g,  $V_{pore}$ = 0.5 cm<sup>3</sup>/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<sub>2</sub> absorption from ambient air for 2 h; 2) thermal desorption of CO<sub>2</sub> 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<sub>2</sub> 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 he inlet flow rate of air or argon was maintained at 50 mL/min. Outlet concentrations of CO<sub>2</sub> were measured using a NDIR CO<sub>2</sub> sensor.

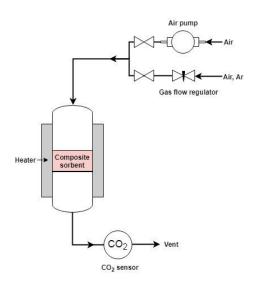


Fig. 1. The experimental set-up for performing TSA tests

Table 1

## Parameters of TSA cycles

No	Description of a step	Parameter	Value
1	CO <sub>2</sub> absorption from air	Duration	2 h
		Gas flow	Air, 1300 mL/min
		Adsorber temperature	30 °C
2	CO <sub>2</sub> desorption	Duration	2 h
		Gas flow	Air or Ar, 50 mL/min
		Adsorber temperature	30 °C → 150; 200 °C
3	Cooling	Duration	2 h
		Gas flow	No flow
		Adsorber temperature	150; 200 °C → 30 °C

**Results and Discussion.** It was shown that amount of CO<sub>2</sub> desorbed at 150°C in either argon or air flow strongly depended on relative humidity (RH) of indoor air during the previous CO<sub>2</sub> absorption step (Fig. 2). The maximal CO<sub>2</sub> 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 CO<sub>2</sub>. However, when the material was heated up to 200°C in air flow, the amount of released CO<sub>2</sub> 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  $K_2CO_3$  utilization extent in the TSA cycles is < 30%. XRD analysis showed that the composite sorbent  $K_2CO_3/AC$  after 12 h of  $CO_2$  absorption from ambient air contains crystalline phases of KHCO<sub>3</sub> and  $K_2H_4(CO_3)_3\cdot 1.5H_2O$ , which means that even after much longer  $CO_2$  absorption step this material absorbs less than 1 mol of  $CO_2$  per 1 mol of  $K_2CO_3$ .

Conclusions. The obtained results show that the composite material can be effectively regenerated at  $150^{\circ}$ C in air flow, but the major drawback is that  $K_2CO_3$  does not fully convert to KHCO<sub>3</sub> during the  $CO_2$  absorption step. Further research is needed to improve performance of  $K_2CO_3/AC$  materials in the process of  $CO_2$  absorption from ambient air.

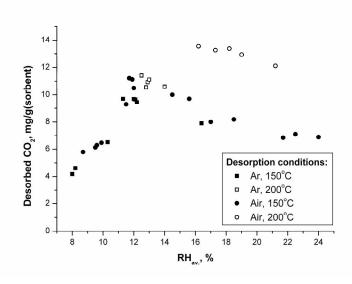


Fig. 2. Amount of desorbed  $CO_2$  (in mg per 1 g of the composite sorbent) depending on average relative humidity throughout the  $CO_2$  absorption step and conditions of the  $CO_2$  desorption step (gas-carrier: argon or air; temperature:  $150 \, \text{C}$  or  $200 \, \text{C}$ 

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