Experimental investigation of CO₂ adsorption onto a carbon based consolidated composite adsorbent for adsorption cooling application

Animesh Pal^{1,2,5}, Ibrahim I. El-Sharkawy^{2,3}, Bidyut Baran Saha^{1,2,4,*},
Skander Jribi^{2,5}, Takahiko Miyazaki^{2,5}, Shigeru Koyama^{2,5}

¹*Kyushu University Program for Leading Graduate School, Green Asia Education Center, Interdisciplinary Graduate School of Engineering Sciences, Kyushu University, Kasuga-koen 6-1, Kasuga-shi, Fukuoka 816-8580, Japan*

²*International Institute for Carbon-Neutral Energy Research (WPI-I2CNER), Kyushu University, 744 Motoooka, Nishi-ku, Fukuoka 819-0395, Japan*

³*Mechanical Power Engineering Department, Faculty of Engineering, Mansoura University, El-Mansoura 35516, Egypt*

⁴*Mechanical Engineering Department, Kyushu University, 744 Motoooka, Nishi-ku, Fukuoka 819-0395, Japan*

⁵*Interdisciplinary Graduate School of Engineering Sciences, Kyushu University, Kasuga-koen 6-1, Kasuga-shi, Fukuoka 816-8580, Japan*

*Author to whom correspondence should be addressed, E-mail:

saha.baran.bidyut.213@m.kyushu-u.ac.jp

Abstract

Adsorption of carbon dioxide onto highly porous activated carbon based consolidated composite adsorbent has been experimentally investigated. Experiments have been conducted at temperatures of 30, 50, 70 °C and pressures up to 7 MPa using magnetic suspension adsorption measurement unit. The innovative adsorption isotherms data have been correlated using three isotherm models namely, Langmuir, Tóth, and modified Dubinin-Astakhov (D-A). The studied models successfully fitted with the experimental

data and Tóth isotherm model shows a better fitting. Results showed that the volumetric adsorption capacity of CO₂ onto the studied consolidated composite is higher than that of CO₂ onto parent activated carbon powder (Maxsorb III). The isosteric heat of adsorption of the studied pairs has been calculated from isotherm data. The performance of ideal adsorption cooling cycle, employing consolidated composite adsorbent/CO₂ pair, has also been simulated at three different evaporator temperatures, namely 5, 10 and 15 °C along with a coolant temperature of 25 °C and heat source temperatures ranging from 45 to 90 °C. The estimated thermodynamic parameters and isotherm data are important for further development of CO₂ based adsorption cooling systems.

Keywords: adsorption cooling; adsorption isotherm; carbon dioxide; composite adsorbent

Nomenclature

A	adsorption potential [kJ mol ⁻¹]
b _o	equilibrium constant [-]
C	adsorption uptake [kg kg ⁻¹]
C _o	saturated amount adsorbed [kg kg ⁻¹]
c _p	specific heat capacity [kJ kg ⁻¹ K ⁻¹]
CE _v	cooling effect per unit volume [MJ m ⁻³]
COP	coefficient of performance [-]
E	adsorption characteristic energy [kJ mole ⁻¹]
k	fitting parameter of modified D-A equation [-]
n	exponential parameter of D-A equation [-]
P	equilibrium pressure [MPa]
P _s	saturated pressure [MPa]
P _c	pressure at critical point [MPa]
q	heat input [kJ kg ⁻¹]
Q	isosteric heat of adsorption [kJ mol ⁻¹]
R	universal gas constant [J mol ⁻¹ K ⁻¹]

SCE	specific cooling effect [kJ kg^{-1}]
T	temperature [K]
T _C	temperature at critical point [K]
t	heterogeneity factor [-]
V _m	molar volume [$\text{cm}^3 \text{ g}^{-1}$]
V _t	specific volume of liquid carbon dioxide [$\text{cm}^3 \text{ g}^{-1}$]
W	volumetric adsorption uptake [$\text{cm}^3 \text{ g}^{-1}$]
W _o	maximum volumetric adsorption capacity [$\text{cm}^3 \text{ g}^{-1}$]
ΔH	vaporization enthalpy [kJ mol^{-1}]

Subscripts

ad	Adsorbent
cond	Condenser
evap	Evaporator
max	Maximum
min	Minimum
ref	Refrigerant
reg	Regeneration
s	Saturation
sens	Sensible

Greek symbols

α	thermal expansion of the adsorbed gas [K^{-1}]
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1. Introduction

Utilization of HCFCs and HFCs in heat pump systems leads to an increase in environmental temperature as these have high global warming potential (GWP). According to the Paris agreement in December 2015, the goal is to hold the increase in the global average temperature to well below 2°C [1]. Accordingly, research and development on the utilization of natural and/or alternative refrigerants in heat pump systems has been intensified. Studies showed that adsorption heat pump system can utilize natural or alternative refrigerants and it can be driven by waste heat or solar heat of temperature below 100°C [2-15]. Among all natural refrigerants, carbon dioxide (CO₂) offers a number of advantages including excellent thermo-physical properties, high volumetric capacity. Moreover, it is a non-toxic, nonflammable refrigerant having zero ODP and GWP as low as 1. Therefore, it is suitable for adsorption cooling applications where the flammability and toxicity of ammonia and hydrocarbons refrigerants for example, may be a problem. Besides, adsorption cooling systems using CO₂ as refrigerant are not limited only to cooling and air-conditioning applications above 0 °C like those using water, but can be applied for freezing and ice-making applications.

Adsorption isotherm and isosteric heat of adsorption are important in order to investigate the performance of CO₂ based adsorption cooling systems. Saha et al. [10] studied adsorption isotherms of CO₂ onto highly porous activated carbon (AC) fiber of type A-20 and AC powder of type Maxsorb III. The authors found that the Tóth and modified Dubinin-Astakhov (D-A) isotherms well matched with the experimental data within $\pm 5\%$ root-mean-square deviation. Guo et al. [16] investigated adsorption of CO₂ on raw AC and three modified AC samples at temperatures ranging from 303 to 333 K. They found that Dubinin-Radushkevich (D-R) and Freundlich adsorption models were the best fitted models with the adsorption data on carbon samples. Singh et al. [17] measured adsorption isotherms in the pressure range of 0–45 bar at various temperatures using Sievert's type experimental setup. Experimental data of CO₂ adsorption isotherms were modelled using Langmuir and D–A isotherm models and found that D–A model was well matched with the experimental data. Himeno et al. [18] experimentally conducted adsorption of CO₂ onto five microporous ACs at different temperatures and pressure ranges using a static volumetric apparatus. The experimental data were

associated using the Tóth model and the D-A model. Shen et al. [19] measured CO₂ adsorption gravimetrically at different temperatures and pressures ranges on pitch-based AC beads. They showed that experimental data were best fitted with the Virial isotherm model at low and high pressures, whereas the multisite Langmuir model only fits well with at low pressures. However, activated carbon of type powder and fiber suffers are prone to low thermal conductivity.

Recently, several researchers have reported that consolidated composites adsorbents can enhance the heat transfer inside the adsorber unit [20-26]. From the above perspective, the present authors have been experimentally investigated the adsorption of carbon dioxide onto highly porous activated carbon based consolidated composite adsorbent. Notable contributions presented in this article are:

- (1) Consolidated composite has been synthesized by the authors.
- (2) Carbon dioxide (CO₂) is used as a refrigerant.
- (3) Adsorption isotherm data of CO₂ onto the developed composite adsorbent are original.
- (4) Isothermic heat of adsorption data are calculated from the isotherm data.
- (5) Ideal cooling cycle performance and effect of packing density of composite adsorbent on volumetric cooling capacity have been discussed.

2. Experiments

2.1. Materials

The material used in the present study is consolidated composite adsorbent. It has been prepared by mixing a highly porous activated carbon powder (ACP) namely Maxsorb III, binder, water, and expanded graphite (EG) with certain ratios. The binder used in this study is Polyvinyl alcohol (PVA) supplied by Kanto Chemical Co., Inc., Japan. Maxsorb III is supplied by Kansai Coke & Chemicals Co. Ltd., Japan whilst expanded graphite (EG) type EC500 is provided by Ito Graphite Industries Co., Ltd., Japan. [Synthetization](#) of consolidated composite adsorbents can be elucidated as follows; Maxsorb III and EC500 are dried in the oven at a temperature of 120°C for 6 hours to remove the moisture content. Secondly, PVA has been mixed with water to make it solvent. Maxsorb III and EC500 powder mixture are then added to the water-binder

solvent. After that the mixer of Maxsorb III, binder, water and EC500 is compressed using a pressing machine at pressure of 34 MPa. Lastly, the consolidated sample is dried in the oven at the temperature of 120 °C about 10 hours to remove water. Adsorbent used in this study is comprised of 70% Maxsorb III, 20% EC500, and 10% binder. The dry sample mass is 208 mg, height is 1.9 mm, and 17 mm diameter with 480 kg m^{-3} packing density. The photograph of prepared consolidated composite is shown in Fig. 1. The total surface area and micropore volume of a sample with similar composition (70% Maxsorb III, 20% EC500, and 10% binder) are $2000 \text{ m}^2 \text{ g}^{-1}$ and $1.094 \text{ cm}^3 \text{ g}^{-1}$, respectively [20].

2.2. Instrumentation

Adsorption isotherms of consolidated composite adsorbent/ CO_2 pair has been measured gravimetrically using magnetic suspension adsorption measurement unit (MSB-GS-100-10M) supplied by BEL Japan. Figure 2 shows the schematic diagram of the adsorption isotherms measuring apparatus. It consists of magnetic suspension balance; an evaporator; isothermal circulation oil bath to control the adsorption/evaporation temperatures; isothermal air bath to avoid condensation within the system; a series of vacuum pumps including rotary, diaphragm and turbo-molecular pump; and pressure gauges and thermocouples. The magnetic suspension unit contains of a permanent magnet, sensor code, and electronic control unit. Adsorption amount is measured as weight the gravity created between the electromagnet mounted on the balance and the load mechanism including a sample hanging from a permanent magnet. To make a measurement under a high temperature and a high pressure without the chance of the balance becoming eroded, the balance section (electromagnet) and the measurement section (load mechanism) are separated from each other by a wall and are not exposed to a measurement atmosphere. It is worth mentioning that buoyancy effect of adsorption quantity measurement is automatically considered by the software. The resolution of magnetic suspension balance is $\pm 10 \text{ }\mu\text{g}$ to record the sample mass. The weight measurement repeatability of the magnetic balance is $\pm 30 \text{ }\mu\text{g}$ with relative error $\pm 0.002\%$ of the reading; a 10000 kPa absolute pressure gauge of type Keller PAA-35X is used to measure the pressure with an uncertainty of ± 0.1 of full scale [27].

2.3. Procedure

The magnetic suspension balance unit has been used to measure the adsorption isotherms of studied adsorbent/CO₂. Experimental procedure can be described as follows; Firstly, consolidated composite adsorbent sample is put into the sample basket and connected to the measuring unit of the system. To remove any adsorbed gas, sample is then regenerated at 130 °C for several hours under vacuum condition for each isotherm. After that temperature is stabilized for measurement condition. The following technique has been used for the measurements of adsorption isotherms.

- a) To measure the adsorption uptake at low to medium pressure ranges, CO₂ gas is charged into the tube from CO₂ cylinder and it is then directed to magnetic suspension balance unit. The pressure in the connecting tube is controlled by control valve C1, as shown in Fig. 2. The pressure could not go above the saturated vapor pressure of carbon dioxide cylinder at ambient temperature (about 5.7 MPa at 20 °C) in this method.
- b) As shown in Fig. 2, liquid reservoir is submerged into a controlled temperature oil bath. In this method, CO₂ is charged into the liquid reservoir and it is directed to the balance unit. Therefore, adsorption pressure is the saturated carbon dioxide pressure in the vaporizer at a set temperature and this method allows to measure pressure up to 7 MPa.

3. Adsorption isotherms

Adsorption uptake of CO₂ onto composite adsorbent has been conducted at temperatures of 30, 50, 70 °C and pressure ranging from 0 to 7 MPa. The Langmuir, Tóth, and modified D-A isotherm models have used in this study to fit the experimental data. The Langmuir and Tóth isotherm models can be expressed by equations 1 and 2, respectively [10, 28].

$$\frac{C}{C_0} = \frac{b_o e^{\frac{Q}{RT}} P}{1 + b_o e^{\frac{Q}{RT}} P} \quad (1)$$

$$\frac{C}{C_o} = \frac{b_o e^{\frac{Q}{RT}} P}{\left(1 + \left(b_o e^{\frac{Q}{RT}} P\right)^t\right)^{1/t}} \quad (2)$$

The Dubinin-Astakhov (D-A) model is defined by the following equation [10, 28].

$$W = W_o \exp \left[- \left(\frac{RT \ln \left(\frac{P_s}{P} \right)}{E} \right)^n \right] \quad (3)$$

$$\text{and } W = CV_m \quad (4(a))$$

The molar volume V_m of adsorbed phase can be calculated using Eq. 4 (b).

$$V_m = V_t \exp (\alpha (T - T_t)) \quad (4(b))$$

At triple point, value of parameter V_t of liquid carbon dioxide is $0.84858 \text{ cm}^3 \text{ g}^{-1}$. The thermal expansion α of the adsorbed gas is 0.0025 K^{-1} or $1/T$ as suggested by Saha et al. [10], and Himeno et al [18]. Amankwah et al. [29] proposed a modified D-A model for estimating pseudo-vapor pressures that is given by Eq. 4 (c). This methodology introduces a parameter, k , which accounts for interactions in the adsorbate-adsorbent systems. It is a fitting parameter and can be obtained from experimental adsorption isotherm data.

$$P_s = \left(\frac{T}{T_c} \right)^k P_c \quad (4(c))$$

Figures 3 to 5 represent a good agreement between the experimental and predicted adsorption uptake using Langmuir, modified D-A equation, and Tóth, respectively. The marker points in the figures show the measured uptakes whereas solid lines indicate the fitted data with studied adsorption models. It can be seen from Figs. 3 to 5 that the

adsorption uptake increases with increasing in equilibrium pressure. It is also observed from the experimental data that the maximum adsorption uptake of CO₂ onto studied composite adsorbent is about 1 kg of CO₂ per kg of adsorbent. Isotherm parameters of the studied adsorption isotherm models have been optimized to fit the measured data for temperatures 30, 50, and 70 °C. Table 1 shows regression coefficient and average relative error for the fitting isotherm models. Numerical value of isotherm parameters has been furnished in Table 2. It can be mentioned that among the three isotherm models, Tóth isotherm model fits well with the experimental isotherms. The effect of k parameter on adsorbed volume W versus adsorption potential A is shown in Figure 6. For k is 4.886, a single characteristic curve is obtained for adsorption temperatures 30, 50, and 70 °C, which indicates that characteristic energy is independent of temperature. A similar observation was also reported by Saha et al. [10] and Himeno et al. [18]. A comparison between present study, Saha et al. [10], and Himeno et al. [18] of adsorption uptake of CO₂ onto Maxsorb III in kilogram per cubic meter have been depicted in Fig. 7. In this comparison packing density of prepared composite adsorbent is found to be 480 kg m⁻³, whilst Maxsorb III powder is found to be 290 kg m⁻³ [20]. It is noticed that present study shows higher adsorption capacity of CO₂ per cubic meter of adsorbent. In addition, El-Sharkawy et al. [20] reported that consolidated composite adsorbent with similar composition (70% Maxsorb III, 20% EC500, and 10% binder) possess higher thermal conductivity compared to Maxsorb III powder.

4. Isosteric heat of adsorption

Isosteric heat of adsorption has been estimated from isotherms data using well-known Clausius-Clapeyron equation that is derived from modified D-A equation and can be written as [10, 28].

$$Q_{CC,D-A} = RT^2 \frac{\partial \ln P_s}{\partial T} + A + \frac{\alpha}{n} TE^n A^{1-n} \quad (5)$$

It is also calculated from heat of adsorption equation proposed by Chakraborty et al. [30] and can be given as [10];

$$Q_{Chak,D-A} = kRT + A + \frac{\alpha}{n} TE^n A^{1-n} + Tv_g \left(\frac{\partial P}{\partial T} \right) \quad (6)$$

The plots isosteric heat of adsorption versus the adsorption uptake of CO₂ has been presented in Fig. 8. It is derived from experimental data using equations 5 and 6. Estimated isosteric heat of adsorption using Tóth model is also depicted on the same figure. It is clearly observed that isosteric heat of adsorption calculated from Tóth model is constant. However, it decreases with increasing fractional adsorption uptake in case of derived from modified D-A and proposed equation by Chakraborty et al. [30]. As can be seen from Fig. 8, heat of adsorption estimated using the equation proposed by Chakraborty et al. [30] shows higher value compare with other models.

5. Thermodynamic analysis of adsorption cooling cycle

Figure 9 shows the schematic diagram of adsorption cooling system which composes an evaporator, two adsorber/desorber heat exchangers that are act as a thermal compressor and a condenser. The adsorber and evaporator connecting through a throttling valve to keep the pressure difference. The prepared consolidated composite sample and CO₂ are used as the adsorbent and refrigerant pair. The thermodynamic relation among pressure, adsorption temperature, and amount of refrigerant adsorbed per unit mass of adsorbent at equilibrium state can be expressed using P-T-W (pressure-temperature-concentration) diagram. The Tóth equation (Eq. 2) is used to draw the P-T-W diagram of composite adsorbent/CO₂ pair, which is shown in Fig. 10. This diagram provides a simple prediction of the performances of an ideal adsorption cooling cycle. The adsorption/desorption cycle of consolidated adsorbent/CO₂ pair has also been depicted on P-T-W diagram as can be seen in Fig. 10. The ideal cycle can be described as follows; in adsorption process (1 → 2, see Fig. 10), the pressure is kept constant at P_{evap}. The refrigerant vapor evaporates in the evaporator and is adsorbed by the adsorbent.

The refrigerant concentration in the adsorber increases from $W_{\min}$ to $W_{\max}$. In pre-heating ($2 \rightarrow 3$) process, the adsorbent is isolated and heated continuously with high temperature heat source and hence the pressure increases from P_{evap} to P_{cond} . In desorption process ($3 \rightarrow 4$), refrigerant vapor is liquefied inside condenser and is then returned to the evaporator through the expansion valve. In this time, refrigerant concentration into adsorbent decreases from $W_{\max}$ to $W_{\min}$. During the pre-cooling process ($4 \rightarrow 1$), the adsorbent is cooled by cooling load and hence pressure decreases from P_{cond} to P_{evap} . The basic adsorption cooling is assumed to be an ideal, time independent and as a result the pressure drops as well as heat loss have been neglected.

The specific cooling effect (SCE) can be described by the equation 7.

$$SCE = (C_{\max} - C_{\min}) \left[\Delta H_{\text{evap}} - \int_{T_{\text{evap}}}^{T_{\text{cond}}} c_{p,\text{ref}} dT \right] \quad (7)$$

The coefficient of performance (COP) can be defined as:

$$COP = \frac{SCE}{q} \quad (8)$$

q can be expressed as:

$$q = q_{\text{sens}} + q_{\text{latent}} \quad (9)$$

where,

$$q_{\text{sens}} = \int_{T_2}^{T_4} c_{p,\text{ad}} dT + C_{\max} \int_{T_2}^{T_3} c_{p,\text{ref}} dT + \int_{T_3}^{T_4} C c_{p,\text{ref}} dT \quad (10)$$

and

$$q_{\text{latent}} = Q \int_{C_{\max}}^{C_{\min}} dC \quad (11)$$

In Eq. 10, the first term defines the sensible heat added to the adsorbent consequently its temperature increases from T_2 to T_4 whilst the sensible heat added to the refrigerant during pre-heating and the desorption process are represented by second and third terms, respectively. The numerical value of all parameters used to calculate SCE and

COP have been furnished in Table 3. The change of specific cooling effect (SCE) with regeneration temperature, T_4 , at three different evaporator temperatures, namely 15, 10 and 5 °C is shown in Fig. 11. The value of SCE increases linearly with the increasing of regeneration temperature owing to the increases of uptake difference with the increasing of regeneration temperature at fixed evaporator and adsorption temperature. Figure 12 shows the comparison of cooling effect per unit volume (CE_v) of present study with different packing densities of composite adsorbent and Maxsorb III/ CO_2 pair [31]. The cooling effect per unit volume can be estimated by multiplying the specific cooling effect and the adsorbent packing density. For knowing the density effect on CE_v , different packing densities adsorbent with same composition have been considered. The packing densities of adsorbent are varied from 480 kg m⁻³ to 750 kg m⁻³ whilst Maxsorb III is 290 kg m⁻³ [20]. It is observed that, the consolidated composite adsorbent with packing density 480 kg m⁻³ shows cooling effect per unit volume as high as 29 MJ m⁻³, which is 32% higher than that of Maxsorb III/ CO_2 (22 MJ m⁻³). It is noteworthy to mention that increasing the packing density of composite increases volumetric cooling effect. According to Zhao et al., 2015 (32), and Choudhury et al 2013 (33) consolidated composite adsorbents based adsorber/desorber heat exchanger offered higher overall heat transfer coefficients even though adsorbent's permeability was decreased. However, very high packing density may reduce the vapor diffusion into the adsorbent material. Fig. 13 illustrates the effect of regeneration temperature on coefficient of performance (COP). It is seen that the COP increases sharply up to a regeneration temperature of 70 °C, after that; there is no significant change in COP although SCE increases. This occurs as a result of the requirement of heat input becomes considerably large once the temperature difference between driving heat source and heat sink is relatively high.

6. Conclusions

Adsorption of carbon dioxide onto a consolidated composite adsorbent has been experimentally investigated at three different adsorption temperatures namely, 30, 50, and 70 °C and pressures ranging from 0 to 7 MPa. The measured adsorption isotherms data have been successfully fitted with the three isotherm models namely, Langmuir,

Tóth, and modified Dubinin-Astakhov (D-A) equation. The volumetric adsorption capacity of CO₂ onto the consolidated composite is compared with that of Maxsorb III/CO₂ pair. The isosteric heat of adsorption of studied adsorbent/refrigerant pair has been predicted from the isotherm data and average value of Q is found to be 22.5 kJ mole⁻¹. The performance of adsorption cooling cycle employing the studied pair in terms of coefficient of performance (COP) and specific cooling effect (SCE) has also presented. It was found that the consolidated composite with packing density 480 kg m⁻³ can achieve cooling effect per unit volume as high as 29 MJ m⁻³, which is 32% higher than that of the Maxsorb III/CO₂ pair at evaporator temperature of 15 °C along with driving heat source and coolant temperatures of 90 °C and 25 °C, respectively. With the advent of the technological improvement for heat exchangers, composite adsorbent materials can be packed inside the adsorber/desorber heat exchanger which will lead to notable performance improvement in adsorption cooling and heat pump systems.

Acknowledgements

This work was financially supported by Japan Science and Technology Agency (JST), Core Research Evolution Science and Technology (CREST).

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Table 1

Regression coefficient and average relative error for the fitting isotherm models.

Table 2

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Table 1. Regression coefficient and average relative error for the fitting isotherm models.

Temperature [°C]	Fitting models	Regression coefficient	Average relative error (ARE)* [%]
			$* ARE = \frac{100}{N} \sum_{i=1}^N \left(\frac{ C_{cal.} - C_{exp.} }{C_{exp.}} \right)$
30	Langmuir	0.9965	2.12
50		0.9996	2.37
70		0.9947	7.04
30	Tóth	0.993	2.53
50		0.9995	1.41
70		0.9957	2.75
30	Modified D-A	0.9948	2.45
50		0.999	0.94
70		0.9923	4.49

Table 2. Fitting parameters of the isotherm models for CO₂ adsorption onto composite adsorbent.

Isotherm Parameters	Langmuir	Tóth	Modified D-A
C_o [kg kg ⁻¹]	1.4	2.05	-
b_o [MPa ⁻¹]	$7.90 \cdot 10^{-5}$	$6.67 \cdot 10^{-5}$	-
Q [kJ mole ⁻¹]	21.8	22.5	-
t [-]	-	0.6	-
k [-]	-	-	4.844
W_o [cm ³ g ⁻¹]	-	-	1.109
E [kJ mole ⁻¹]	-	-	5.42
n [-]	-	-	1.29

Table 3. Numerical value of parameters used for calculation of SCE and COP.

Parameters	Values
Q [kJ mole ⁻¹]	22.5
C_{max} [kg kg ⁻¹]	1.02
C_{min} [kg kg ⁻¹]	0.55
T_{cond} [°C]	25
T_{evap} [°C]	5, 10, and 15
T_{reg} [°C]	45 to 90
$c_{p,ref}$ [kJ kg ⁻¹ K ⁻¹]	2.73
$c_{p,ad}$ [kJ kg ⁻¹ K ⁻¹]	0.806
ΔH [kJ kg ⁻¹]	214.98, 197.15, and 176.65 at $T_{evap} = 5$, 10, and 15 °C, respectively.

Fig. 1. Photograph of synthesized consolidated composite adsorbent; (a) diameter of adsorbent (b) thickness of adsorbent.

1. Magnetic suspension balance; 2. Sample cell; 3. Circulation oil jacket; 4. Isothermal circulation oil bath; 5. Sheathed heater; 6. Refrigerant container; 7. Helium cylinder; 8. Carbon dioxide cylinder; TMP: Turbo-molecular pump; R.P.: Rotary pump; D.P.: Diaphragm pump; T: Thermocouple; P1-P4: Pressure gauges; C1-C2: Controlled valves; V1-V12: Valves.

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Fig. 12. Comparison of cooling effect per unit volume of present study with different packing densities of studied adsorbent and Maxsorb III/CO₂ pair with function of regeneration temperature.

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