

Experimental Investigation of Granular Activated Carbon/R-134a Pair for Adsorption Cooling System Applications

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ABSTRACT

The objective of this study is to investigate the adsorption characteristics of granular activated carbon GAC/R-134a pair in the temperature range of 20°C - 60°C and pressure up to 10 bars. The Dubinin-Astakhov adsorption equilibrium model is fit to experimental data with acceptable error limit. The pressure-temperature-concentration (P-T-X) diagram of the pair is also presented. The isosteric heat of adsorption of R-134a on activated carbon has been calculated using the Clausius-Clapeyron equation as a function of adsorption capacity. The maximum adsorption capacity was found to be 1.92 kgR134a/kg carbon at 20°C after 1200 s. The experimental results also show that the increase of heat transfer area improves the adsorption capacity per kg of adsorber, which leads to the design of a finned tubes heat exchanger adsorption unit.

KEYWORDS

Adsorption; Activated Carbon; R-134a; Isosteric Heat

1. Introduction

Over the past two decades, adsorption refrigeration system has gained considerable interest due to its ability to combat ozone depletion problem that was caused by the utilization of CFCs and HCFCs in cooling system [1,2]. The thermo-physical properties of adsorbent/refrigeration pairs as well as the operating conditions have significant effects on the system performance [3,4]. The adsorption cooling and refrigeration systems have many advantages of being compact, free moving parts, and free of toxic and environmentally harmful materials and need low temperature to work, and don't require synthetic lubrication [5,6]. It is a matter of regret that adsorption data are unavailable from the manufactures of adsorbents [7,8].

Just the data of surface area and pores volume of the adsorbent are available which are insufficient to design a thermally driven adsorption system [9,10]. The design adsorption based cooling cycle is inevitable to evaluate

adsorption isotherms of the assorted adsorbent/adsorbate pairs as well as the isosteric heat of adsorption.

The selection of any pair of adsorbent/adsorbate for refrigeration applications depends on certain desirable characteristics ranging from their thermodynamic and chemical properties to their physical properties and even to their costs or availability [11]. The debate is on whether to continue with hydro fluorocarbon (HFC) refrigerants or if a time has come to switch fully to natural refrigerants such as ammonia and carbon dioxide [12]. In addition, the even flammable hydro fluorocarbon is gaining prominence in the wake of global warming issue that is rocking the world. Hydro fluorocarbon (HFC-134a) is considered friendly refrigerant on ozone layer. Therefore, all countries will continue developing it at least for the next two decades [12].

The following are some representative examples of thermally driven adsorption systems. Akkimaradi *et al.* [13] measured adsorption isotherms of R-134a on three types of activated carbons namely Chemviron, Fluka and

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Maxsorb II. Askalany *et al.* [11] estimated adsorption characteristics of granular activated carbon/R-134a pairs. Pons and Guillemot [14] designed and tested an activated carbon-methanol icemaker driven by solar heat. It was observed that the coefficient of performance (COP) of icemaker is 0.15. Wang *et al.* [8] studied the adsorption characteristics of ACF/methanol and activated carbon (AC)/methanol pairs. They showed that the coefficient of performance of ACF/methanol pair is about 10 - 20 percent higher than that of AC/methanol pair. Vasiliev *et al.* [15] and Vasiliev [16] developed the adsorption heat pump with activated carbon fiber as an adsorbent and ammonia as the working fluid. They explained the effect of adsorbent material, which combined activated carbon fiber and some salts on adsorption capacity. The full adsorption capacity of the activated carbon fiber methanol system reaches up to 0.55 kg of methanol/kg of ACF. Banker *et al.* [17] introduced a mathematical model for activated carbon R-134a pair based two-stage thermal compression adsorption refrigeration using low-grade thermal energy sources. It derived a COP of 0.16 for the system. In addition, they showed that the system achieved low COP of less than 0.04 at 0.93°C driving source temperatures in another study [18].

Enhancement of heat and mass transfer in the adsorber/desorber heat exchanger is the most important factor to improve the system COP [9]. There are two major points for the achievement of heat and mass transfer enhancement. First is to improve heat and mass transfer mechanically between adsorbent bed and the surface of the heat transfer material, such as extended heat transfer. The second point is to develop new adsorbent material having highly uptake and adsorption rate. Chang *et al.* [19] used flat-tube type heat exchangers for adsorption beds to increase the heat transfer area and improve the heat transfer ability between the adsorbent and heat exchanger fins. The study was conducted under the standard test conditions of 80°C hot water, 30°C cooling water, and 14°C chilled water inlet temperatures. It has provided a specific cooling power (SCP) of about 176 W/(kg adsorbent). Watanabe *et al.* [20] investigated the possibility of producing cold energy by a super active carbon SAC/ethanol adsorption system with a packed bed type adsorber. The SAC/ethanol adsorber system enables to increase the heat transfer fluid operating temperature range, while the amount of cooling energy production is the same as the conventional silica gel-water based adsorption system [9,21]. Ito *et al.* [22] made an adsorber heat exchanger composed of a consolidated bed of silica gel particles to enhance heat transfer. They reported the feasibility of adsorption refrigeration operating using the consolidated bed heat exchanger.

From the above perspective, this study investigated the adsorption characteristics of GAC/R-134a Pairs for cool-

ing system applications. A circular tube heat exchanger with fins was adopted as an adsorber bed. Adsorption isotherms of R-134a on granular activated carbon were measured within the temperature from 20°C to 75°C. The instantaneous capacity of R-134a has been recorded at each 60 s under constant volume, constant temperature and variable pressure. In addition, the Dubinin-Astakhov (D-A) equation was used to model equilibrium data of the adsorption and desorption experiments. The present data are compared with those of Saha *et al.* [5] and Askalany *et al.* [11].

2. Experimental Section

2.1. Materials

Adsorption technology can be used for not only air conditioning and refrigeration but also for upgrading heat with thermal transformers, and the type of refrigerant should be selected according to the application [4]. The requirements for a suitable refrigerant are generally as follows: 1) high latent heat of vaporization, 2) thermal stability, 3) environmental harmless, 4) inflammability, 5) innocuousness and 6) saturation pressure between 1 and 5 atm in working temperature [4,23]. Unfortunately, there are no refrigerants that have all the characteristics shown above. The refrigerant R134a of 99.9% purity has been used in the present study and supplied by Foshan Lianyou Environmental Protection Chemicals Co., Ltd China. All properties of R134a used in this paper were evaluated from REFPROP7 software, which is based on the generalized equation of state proposed [5,24].

The activated carbon is made of materials such as wood, peat, coal fossil oil, chark, bon coconut shell and nut stone [4,23]. The microcrystal for the activated carbon product from bon is a six-element carboatomic ring, [23]. While the adsorption performance is influenced by the functional group that increases adsorption, as sulfonic group decreases it [25,26]. Acidic functional group increases adsorption selectivity. The functional groups on the surface of activated carbon are different if the original carbonaceous material and activation method are different [4]. The specific area of activated carbon is between 500 and 1500 m²/kg [4]. **Table 1** shows the thermo-physical properties of granule activated carbon [11,12].

2.2. System Description

The schematic diagram of the experimental setup of adsorption test unit is illustrated in **Figures 1(a)** and **(b)**. The experimental setup includes two systems. One is the heating system; the other is the cooling system. The heating system includes an electric heater capacity range from 1.75 kW to 4 kW is used to drive the system by heating intermediate water. Circulating the intermediate

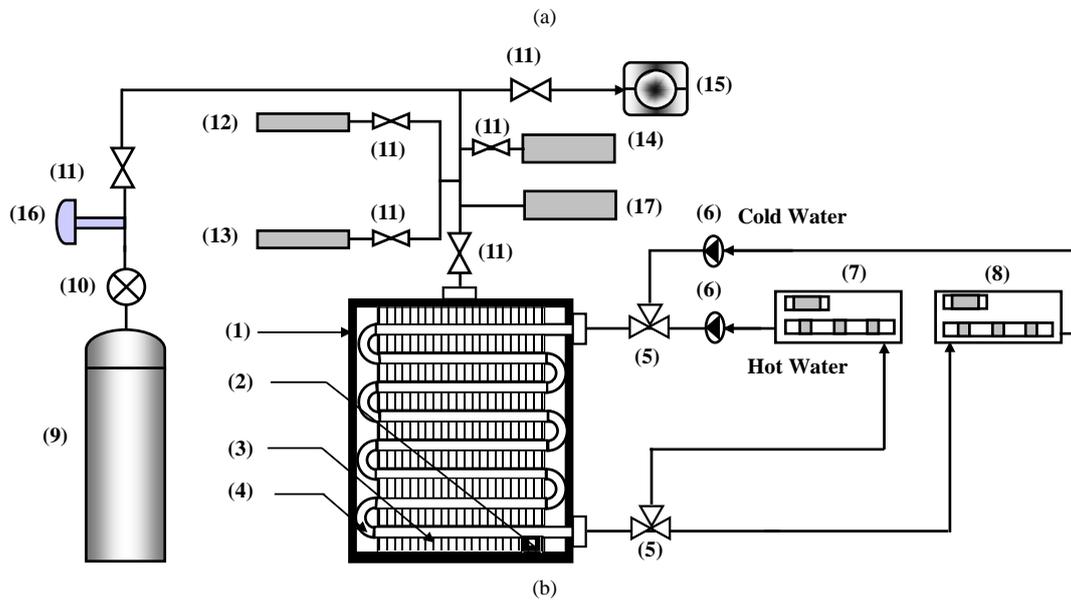
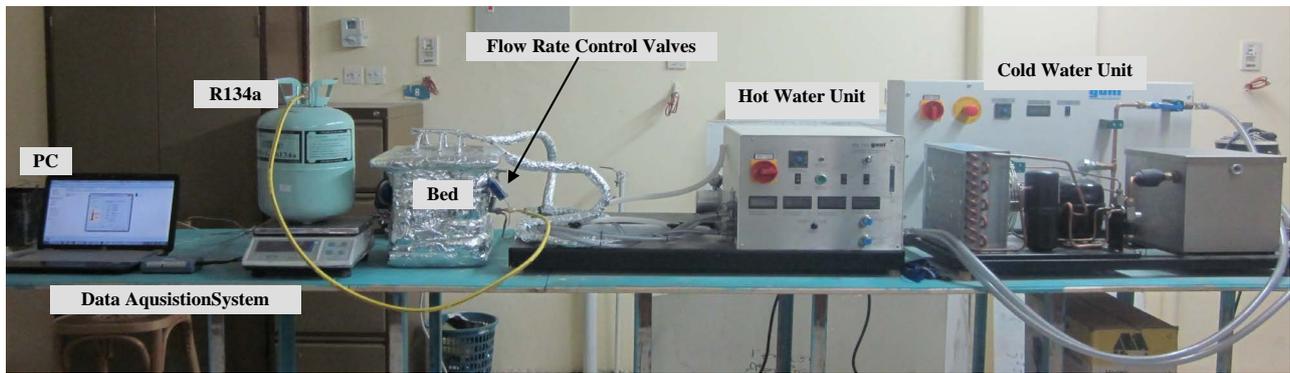


Figure 1. 1. Vacuum Chamber; 2. Adsorbent; 3. Flat Fins; 4. Tube; 5. Three Way Valve; 6. Water Pump; 7. Hot Water Unit; 8. Cold Water Unit; 9. Refrigerate Cylinder; 10. Pressure Regulator; 11. Valve; 12. Pressure Transducer; 13. Vacuum Gauge; 14. Mass Flow Control; 15. Vacuum Pump; 16. RTD Temperature Measure; 17. Temperature Measuring Unit.

Table 1. Physical properties of commercial granule activated carbon.

Properties	Value
Bulk density (g/mol)	0.57
Apparent density (g/mol)	0.85
Density (g/mol)	2.0
Void fraction	0.38
Pore volume (mol/g)	0.78
Specific surface area (m ² /g)	950
Average pore diameter (mm)	3.0
Iodine value (mg/g)	950

water inside the system is done by using a 0.4 kW pump. The cooling system includes a thermostatic water tank while an electronic controller keeps the water temperature constant.

The adsorption bed is a finned tubes heat exchanger

with dimensions of 280 mm × 260 mm × 190 mm. It consists of 54 tubes and 85 fins where the tubes are made of copper and the fins are made of aluminium. Every tube has an inner diameter of 8 mm and outer diameter of 10 mm and every fin has a thickness of 0.35 mm. They are wrapped with a hollow sheet to prevent the granular activated carbon (GAC) from falling and filled with 2 kg of the GAC. They are put in insulated stainless steel casing with dimensions 295 mm × 280 mm × 215 mm provided with pressure gauges, thermocouples and inlet and outlet tubes for water and refrigerant. The characteristics of the finned tubes heat exchanger used are listed in **Table 2**.

2.3. Instrumentation

The experimental facility was more instrumentation devices such as pressure transducer, thermocouple and flow rate. The bed pressure was monitored by Wika A-10 pressure transducer with the accuracy of ±0.5 percentage

Table 2. Characteristics of finned tubes heat exchanger.

Item	Value
Heat exchanger dimension (mm)	280 × 260 × 190
Number of tubes	54
Number of fins	85
Outer tube diameter (mm)	10
Inner tube diameter (mm)	8
Fins thickness (mm)	0.35
Casing dimension (mm)	285 × 280 × 215

full scale (0 - 15 bar). The inlet and outlet temperatures and temperatures in the bed were measured with M100-RTD with the accuracy of ±0.06%. The mass flow rates of hot and cold water measured with TUR FLO flow meters with the accuracy of ±0.7%. A data acquisition system consists of a DAQ-TC connected to a computer and is used to measure and record all thermocouple and pressure gage measurements. The software TRACER DQA is used to program the DAQ-TC and download and visualize the data. The digital balance was used to measure the amount of refrigerant with maximum reading 50 kg and accuracy about ±0.001 g.

2.4. Procedures

The goal of the experimental procedure is to obtain adsorption characteristics relating to the equilibrium adsorption capacity of the granule activated carbon/R-134a pair. These characteristics are important as they define the thermodynamic operation of an ideal cycle and therefore set limits with respect to performance and system size [27]. Prior to adsorption process, the GAC is placed in an oven for 12 h. The oven temperature is kept constant at 145°C. Before starting adsorption test the bed was evacuated using a vacuum pump to 0.05 bar. The bed was heated by using hot water circulation at 96°C during 6 h, while the vacuum process is still running. The vacuum process has been occurred to take out any gases and moisture from the bed.

The bed charging (adsorption process) is starting by opening the valve between the refrigerant cylinder and the bed as shown in **Figure 1(b)**. The pressure of the bed increased gradually with the refrigerant entering the bed. The pressure does not allow more than the saturation pressure at such a temperature 20°C to avoid condensation [11,12]. The valve between the refrigerant cylinder and bed was closed every 60 s or reached to equilibrium state. The mass of refrigerant was estimated from the difference of the test section mass at the beginning and end of each test. After the bed reaches to the equilibrium state, the bed is then heated gradually up to 100°C again. This process was repeated five times at different temper-

atures 20°C, 27°C, 34°C, 41°C, 48°C and 60°C. The equilibrium adsorption is the mass of adsorbed refrigerant divided by the mass of dry adsorbent in the adsorber.

After charging the bed by the R-134a, the system started to run by heating the bed. The heating process (desorption process) started by opening the valve between the bed and the refrigerant cylinder. The desorption process was done at different temperatures 50°C, 60°C, 75°C, 85°C, 95°C and 105°C. The experimental uncertainty of mass ±0.03 g, and the measured equilibrium adsorption has the uncertainty between 3.6% and 5.7%.

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3. Results and discussions

3.1. Adsorption Equilibrium

The Dubinin-Astakhov D-A [28,29] “Equation (1)”, was chosen to model the experimental adsorption data since it is used extensively to estimate the equilibrium uptake of GAC/R134a pair.

$$X = X_o \exp \left\{ - \left[\frac{RT}{E} \ln \left(\frac{P_s}{P} \right) \right]^n \right\} \quad (1)$$

where X is the instantaneous uptake, X_o is the maximum uptake capacity which was estimated experimentally to be 1.92 kg/kg, and R denotes the universal gas constant. The parameter n is an exponential constant which estimated experimental isotherms. In the present study, the values of E and n were determined to be 9.574 kJ/mol and 1.92 respectively. By knowing X_o , n and E “Equation (1)”, could be written in form of “Equation (2)”.

$$X = X_o \exp \left\{ - \left[0.0008686 \times T \ln \left(\frac{P_s}{P} \right) \right]^{1.92} \right\} \quad (2)$$

It is well known that the adsorption cooling potential of adsorbent is determined by both maximum adsorption capacity and the rate of adsorption and desorption, among other parameters. These parameters are weight of the bed, temperature and pressure which measured at every 90 s. **Figure 2** shows experimental results for the adsorption capacity of GAC/R-134a pair against relative pressure at six different temperatures. As can be seen from **Figure 2**, the equilibrium adsorption capacity of temperature 20°C is considerably larger than those of other temperatures. Also the maximum adsorption capac-

ity that could be reached is 1.92 kg/kg at 20°C adsorption temperature with an interval time of 1200 s. The figure shows the adsorption capacity rises rapidly in the first 110 s and then has a slow progress and the adsorption capacity approaches a limiting value as the relative pressure approaches unity. This order is corresponding to that of the lower vacuum pressure in the bed at starting process.

Figure 3 shows the isobaric adsorption GAC/R-134a pair at four different pressures of 1, 2, 4, 6 and 8 bar corresponding to the saturated temperature of R134a. It is observed that the isobaric adsorption is the maximum value at low temperature and then decreases with the increase of temperature.

The relation between desorption capacity during desorption process and relative pressure is shown in Figure 4. The desorption process was measured at six different temperatures, 50°C, 60°C, 75°C, 85°C, 95°C and 105°C. It can be noticed that the instantaneous desorption capacity decreases rapidly at the start of the desorption process. This is due to that the relative pressure is high of the bed at the start of the desorption process. After that, the desorption capacity decreases slowly with decreases in the relative pressure.

The isotherms obtained from the present study are compared with the results obtained by Saha *et al.* [5] and Askalany *et al.* [11] and the results of the comparison are shown in Figure 5. It is observed that the maximum uptake that Saha *et al.* [5] and Askalany *et al.* showed [11] is lower than that of the present study. This is due to increasing of the heat transfer area achieved by using a finned tubes heat exchanger.

Figure 6 shows the adsorption isotherms for GAC/R-134a pair over the temperature range of 10°C - 95°C. The isotherms are expected by using D-A “Equation (2)”

where the equilibrium pressure for different values of equilibrium adsorption capacity, X at constant temperature [11,12]. The relation between pressure and temperature at different adsorption capacities could be predicted by using “Equation (2)”, as shown in Figure 7. This figure is known as P-T-X diagram [5]. It is observed that, at certain equilibrium pressure, the equilibrium adsorption capacity decreases with increasing adsorbent temperature. This is due to that the heat of adsorption is generated and hence the adsorbent temperature increases during the adsorption process. The generated heat is usually removed by using water as a cooling fluid. Therefore, it is important to enhance the heat transfer characteristics of the adsorber so that it can achieve by using a finned tubes heat exchanger, where the heat transfer area increases.

3.2. Heat of Adsorption

The isosteric heat is defined as the heat of desorption under constant adsorption capacity [5]. The isosteric heat of adsorption for GAC/R-134a pair can be expressed by using the following Clausius-Clapeyron equation [5,11, 12] is shown.

$$R \frac{\partial \ln p}{\partial (1/T)} = -Q_{st} \tag{3}$$

where Q is the isosteric heat adsorption (kJ/kg_w),

$$h_{fg} + E \left\{ \left[\ln \left(\frac{X_o}{X} \right) \right]^{-1/n} + a \left(\frac{T}{T_c} \right)^b \right\} = Q_{st} \tag{4}$$

where E is the characteristic energy which is equal to 98.848 kJ/kg and T_c is the critical temperature for R-134a it is equal to 100.9°C.

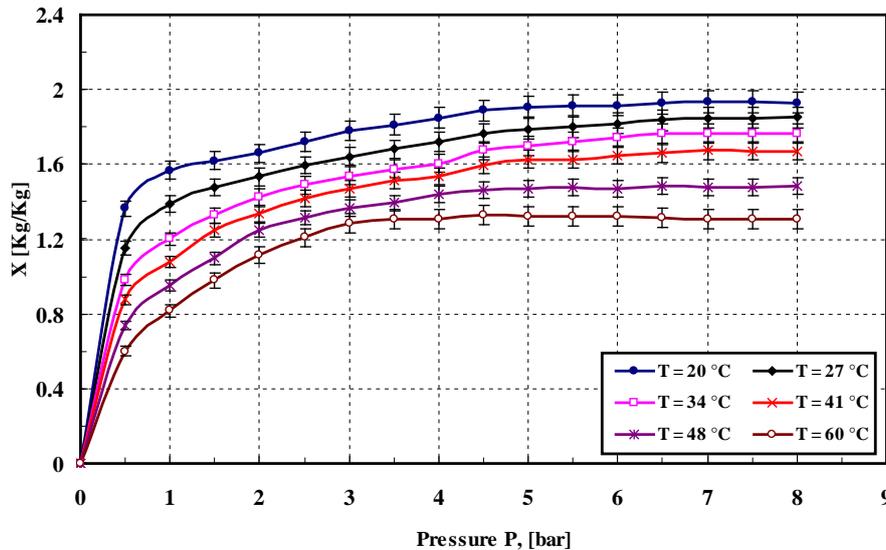


Figure 2. Adsorption isotherm of R-134a on granular activated carbon.

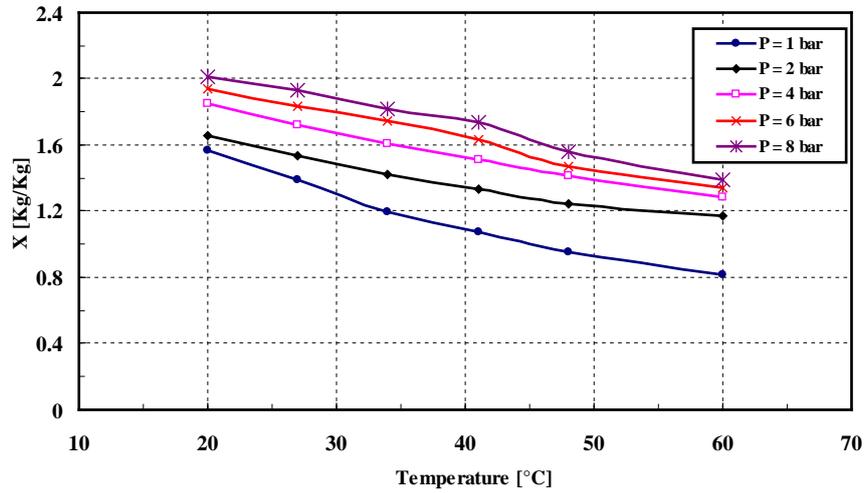


Figure 3. Effect of pressure on adsorption isotherm of R-134a on granular activated carbon.

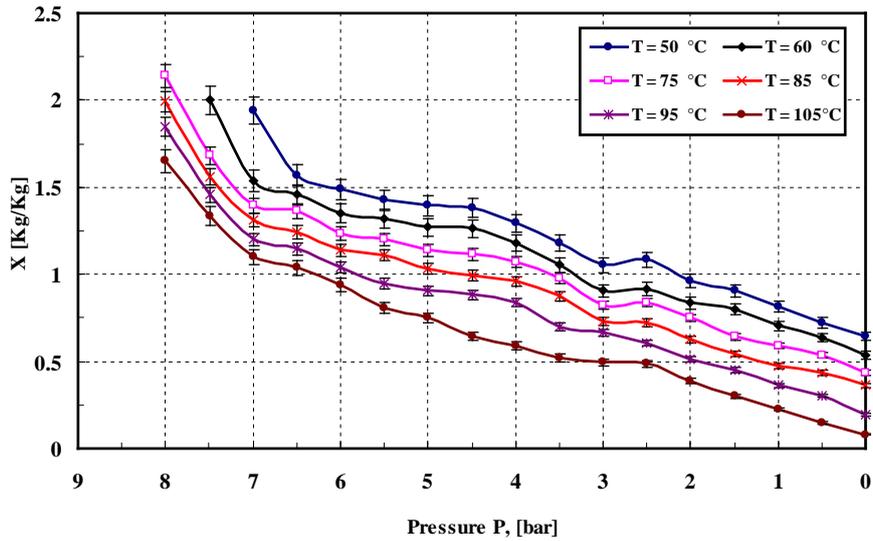


Figure 4. Desorption isotherm of R-134a on granular activated carbon.

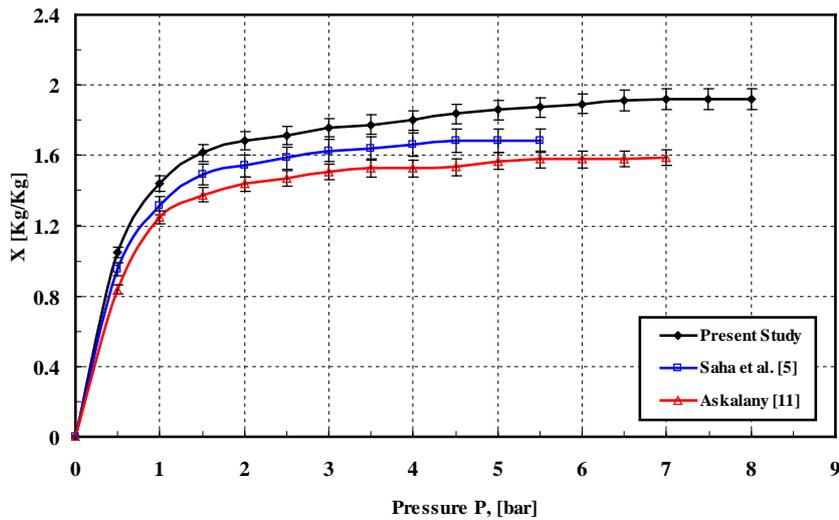


Figure 5. Adsorption isotherm of R-134a on granular activated carbon at 25°C.

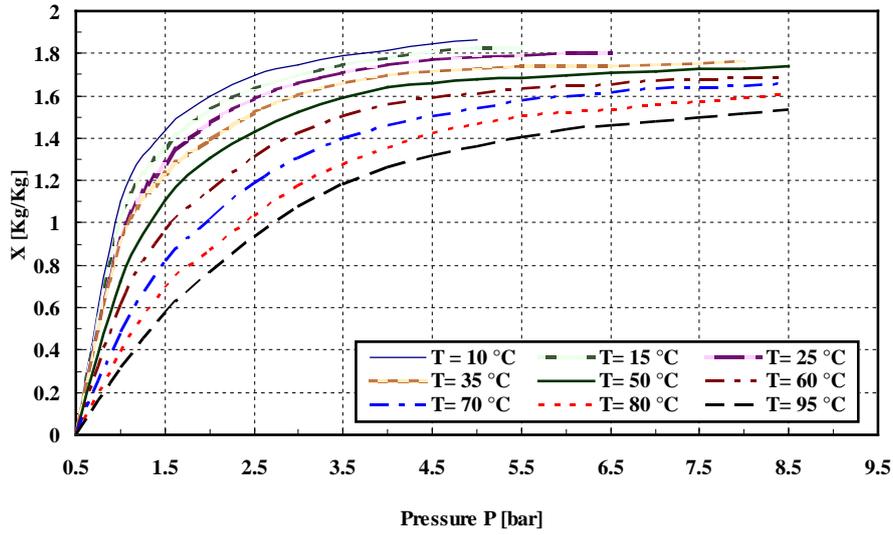


Figure 6. Adsorption equilibrium of GAC/R-134a pair.

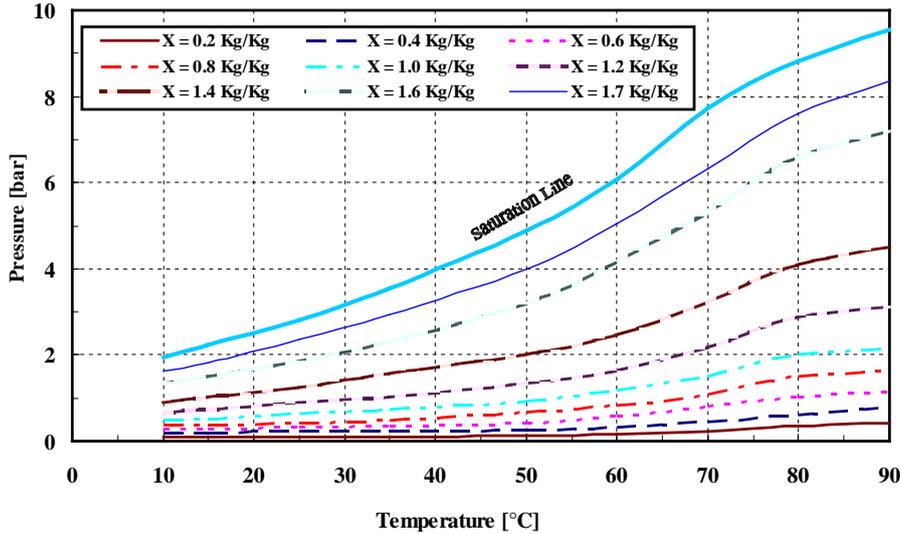


Figure 7. P-T-X diagrams of the GAC/R-134a pair.

El-Sharkawy *et al.* [5] and Habib *et al.* [10] approximated the values of the constants *a* and *b* for R-134a of equal 1.81 and 6.25 respectively. Using the above values, the “Equation (4)”, could be written in the following form;

$$Q_{st} = h_{fg} + 9.574 \left\{ \left[\ln \left(\frac{1.92}{X} \right) \right]^{0.55} + 1.81 \left(\frac{T}{374.05} \right)^{6.25} \right\} \tag{5}$$

From “Equation (5)”, the variation of isosteric heat of adsorption with adsorption capacity at different temperatures is shown in **Figure 8**. One observes from this figure that the isosteric of adsorption decreases with increasing adsorbate uptake of R-134a. In addition the temperatures of bed have more effect on isosteric heat, where the ma-

ximum value of isosteric heat obtained with low temperature at 20°C.

Similar results were recommended by Saha *et al.* [5] where the adsorption characteristics and heat of adsorption of R134a were measure on activated carbon. Saha *et al.* [5] illustrated that the Maxsorb III consists mainly of micropores with different width adsorbs rapidly onto sites and R-134a of high energy, and as adsorption progresses, molecules adsorb onto sites of decreasing energy.

The R-134a molecules first penetrate into narrow pores of Maxsorb III, resulting in a stronger interaction between adsorbate and Maxsorb III. This implies a higher value of isosteric heat of adsorption at lower loading. After completely filling the smaller pores, R-134a molecules are gradually accommodated in larger pores, in

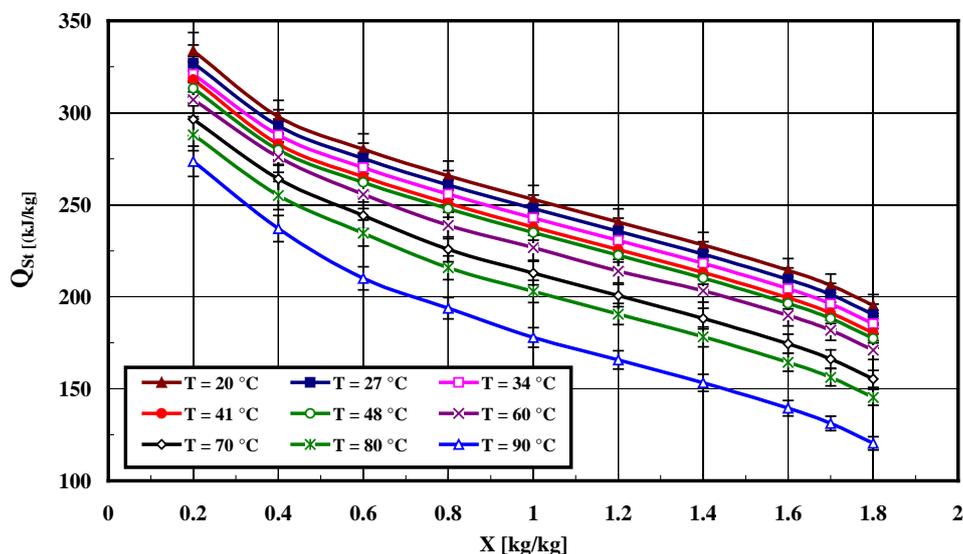


Figure 8. Isosteric heat of adsorption at different temperature.

which the adsorption affinity becomes weaker [5,12]. Therefore a monotonic decrease in isosteric heat of adsorption as a function of adsorbate R-134a uptake.

4. Conclusions

The adsorption capacity (X) of R-134a on a granular activated carbon for adsorption/desorption process has been experimentally studied. The bed was designed and built with finned tubes heat exchanger to increase the heat transfer area. The experiments were conducted over a temperature range from 20°C to 60°C and pressure up to 10 bars. The data were correlated with Dubinin-Astakhov (D-A) equation corresponding adsorption/desorption process. The present results are compared with the results performed by other researchers.

The main remarkable points can be summarized as follows:

1) The adsorption capacity per kg of adsorbent increases rapidly with increasing relative pressure at the beginning of the adsorption process.

2) The maximum adsorption capacity uptake is 1.92 kg/kg at 20°C after 1200 s.

3) The maximum value of adsorption capacity decreases to 1.31 kg/kg with the increase of adsorbent temperature at 60°C.

4) The maximum adsorption capacity increases about 9.62% compared with the other researches', because the heat transfer area increases in bed at 25°C.

5) The isosteric heat of adsorption varied from 120 to 340 kJ/kg for adsorption capacities ranging from 0.2 to 1.8 kg/kg.

6) The accurate adsorption isotherms and heat of adsorption data of R-134a on granular activated carbon are essential in designing thermally driven pressured bed ad-

sorption chillers based on assorted pair.

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Nomenclature

a : Constant, [-]
 b : Constant, [-]
COP: Coefficient of Performance, [-]
 E : Characteristic energy, J/mol
 h_{fg} : latent heat, kJ/kg
 n : Exponential constant, [-]
 P : pressure, bar
 Q_{st} : Isosteric heat adsorption, kJ/kgw
 R : Universal gas constant, mol/K
 T : Temperature, K
 T_c : Critical temperature for R-134a, K
 X : Adsorption mass capacity, $\text{kg}_{\text{R134a}}/\text{kg}_{\text{carbon}}$
 X_o : Maximum adsorption mass capacity, $\text{kg}_{\text{R134a}}/\text{kg}_{\text{carbon}}$