Adsorption isotherms and kinetics of HFC410A onto activated carbons

Ahmed A. Askalany^{a,*}, Bidyut B. Saha^{b,c,*}, Ibrahim M. Ismail^d

^aMechanical Engineering Department, Faculty of Industrial Education, Sohag University,
Sohag, 82524, Egypt

^bInterdisciplinary Graduate School of Engineering Sciences, Kyushu University, 6-1 Kasuga-
Koen, Kasuga-Shi, Fukuoka 816-8580, Japan

^cInternational Institute for Carbon-Neutral Energy Research (WPI-I2CNER), Kyushu
University

^dMechanical Engineering Department, Faculty of Engineering, Assiut University, Assiut
71515, Egypt

*Corresponding Authors, Tel: +20-862370572, E-mail: ahmed_askalany3@yahoo.com (AAA),
Tel: +81-92-583-7903, E-mail: saha.baran.bidyut.213@m.kyushu-u.ac.jp (BBS).

Abstract

Experimental and theoretical adsorption isotherms and kinetics data of HFC410A onto activated carbons are reported. Activated carbon powder of type Maxsorb III and activated carbon fiber of type A-20 have been used as adsorbents. An experimental apparatus has been designed and built and experimental investigation has been carried out at five adsorption temperatures in the range of 10-50°C. Experimental data has been fitted with Tóth and Dubinin-Astakhov (D-A) adsorption isotherm models and linear driving force (LDF) adsorption kinetics model. A range of adsorption parameters such as isosteric heat of adsorption, adsorption uptake and temperature dependence and Henry's law coefficients are

evaluated. A correlation between Henry's law coefficients and isosteric heat of adsorption at limiting uptake is analyzed.

Keywords; Activated carbon, adsorption, heat of adsorption, HFC410A

Nomenclature

A	surface area, m^2
b	Adsorption affinity, kPa^{-1}
b_0	adsorption affinity at infinite temperature, kPa^{-1}
C	adsorption uptake on mass basis, kg/kg
C_0	maximum adsorption uptake on mass basis, kg/kg
D_s	surface diffusion coefficient, m^2/s
D_{s0}	pre-exponential coefficient, m^2/s
E	characteristic energy, kJ/kg
E_a	activation energy, kJ/kg
F_0	constant
h_a	adsorbed phase enthalpy, kJ/kg
h_{ag}	vapor-adsorption phase enthalpy difference, kJ/kg
h_{fg}	heat of vaporization, kJ/kg
h_g	vapor phase enthalpy, kJ/kg
H_{st}	isosteric heat of adsorption, kJ/kg
H_{st0}	heat of adsorption at limiting relative loading, kJ/kg
k_H	Henry's law coefficients, kPa^{-1}
m	mass, kg
n	structural heterogeneity parameter
P	pressure, kPa

P_s	saturation pressure, kPa
R	universal gas constant, kJ/kmole.K
R_{gas}	gas constant, kJ/kg.K
R_p	particle radius, m
z	micro pore distribution parameter
T	temperature, K
t	time, s
V	volume, m ³
v_a	adsorbed phase volume, m ³ /kg
W_0	total pore volume, m ³ /kg
α_g	thermal expansion coefficient of gas phase, K ⁻¹
ρ	density, kg/m ³
Subscript	
ads	adsorbent
ads	adsorbent
ads cell	adsorption cell
f	final
in	initial
j	number of measurement
load	load cell
pore	pore
sol	solid
tube	tube
void	void

1. Introduction

As global warming becoming an increasingly serious issue, the existing high GWP (global warming potential) based refrigerants must be disposed safely before replacing those by natural refrigerants or relatively lower GWP based refrigerants. Adsorption technology is considered one of the most effective ways to capture and store the residual gases [1-4].

The adsorption isotherms and kinetics of several greenhouse gases such as HFC134A [5], HFC507A [6], HFC32 [7] and CO₂ [8] have been investigated experimentally and theoretically. However, enough data are not available in open literature on the adsorption isotherms and kinetics of HFC410A on to highly porous activated carbon powder (ACP) and activated carbon fiber (ACF).

HFC410A is a near-azeotropic mixture of difluoromethane (CH₂F₂, called HFC32) and pentafluoroethane (CHF₂CF₃, called HFC125) of equal mass fraction. It will be phased out within several years according to the IPCC 4th assessment report [9]. It may be replaced by new low GWP based refrigerants such as CO₂, HFO1234ze(E) [8, 10]. Table 1 shows the ODP, GWP and normal boiling temperature of several HFCs and HFOs.

HFC410A has been used as a refrigerant in old fashion air conditioning systems. Adsorption data on HFC410A are expected to be beneficial for possible thermal compression, separation of the mixtures at the end of the life cycle and capture of the mixture to replace it. Loh et al. [11] studied the adsorption isotherms of HFC410A onto ACP of type Maxsorb III. The presented data at that study focused only on adsorption uptake and heat of adsorption of Maxsorb III/HFC410A.

In addition to the adsorption uptakes and heat of adsorption of activated carbon powder of type Maxsorb III/HFC410A, this study presents the adsorption kinetics and other important parameters such as Henry's law coefficients and heat of adsorption at limiting

relative loading. Moreover, the adsorption isotherms, heat of adsorption, Henry's law coefficients, relation between kH and H_{st0} and adsorption kinetics of activated carbon fiber of type A-20/HFC410A have been presented. These data are essential in designing of adsorption heat pump and storage systems.

2. Experimental apparatus

2.1 Description

The experimental apparatus as shown in Fig. 1 consists mainly of an adsorption cell and a load cell settled inside two different water baths. Two water circulators with an external plumbing are conducted to attain homogeneous temperatures inside the water baths. Seven valves are installed at different locations of the experimental apparatus in order to control the flow of adsorbate. The adsorption and load cells are made of stainless steel (SUS 304) with 2.5 liter volume for each cell. A copper tube coil with a diameter of 1/8 inch has been fabricated inside the adsorption cell to improve heat transfer (heating or cooling) of the adsorption cell. The copper coil is connected to an individual water circulator with heater and cooler. A circular mesh has been installed inside the adsorption cell to prevent migration of activated carbon particles. A Coriolis gas flow meter has been located between the adsorption and load cells to measure flow rate of the adsorbate. The experimental apparatus has been connected to a vacuum pump which has been used for evacuation purposes. Series of type K thermocouples with an accuracy of $\pm 0.1^{\circ}\text{C}$ are attached at different locations all over the test rig to detect temperatures. Pressure transducers are used to measure the pressures of the adsorption and load cells and the evaporator with an accuracy of $\pm 0.25\%$ of the full scale reading.

2.2 Procedure

Before starting the experimental runs, the adsorbent are prepared by heating it up to 100°C for six hours to ensure no moisture content. The adsorbent has been assembled inside the adsorption cell after weighting it by using ± 0.05 g accuracy weighting balance. The volume of adsorption cell, load cell and connecting tubes are determined by filling water method. To start adsorption process, the whole system has been evacuated until the pressure become as low as 25 Pa inside the adsorption and load cells. The load cell is then loaded by HFC410A to a certain pressure where the adsorption process is managed for different progressive pressures. Pressure inside the load cell has not allowed exceeding the saturation pressure at the desired adsorption temperature in order to prevent condensation of the HFC410A. After obtaining stable conditions of initial pressure and temperature at the load cell, the adsorption process starts by opening valves between load and adsorption cells. After equilibrium, the valves are closed. This process is repeated many times by raising the pressure of the load cell every time at same adsorption temperature.

The experimental adsorption process is repeated for various temperatures in the range of 10-50°C. The maximum adsorption temperature has been decided to be less than the critical temperature of the refrigerant which is 70.17°C in case of HFC410A.

2.3 Materials

Activated carbon powder (ACP) of type Maxsorb III and activated carbon fiber (ACF) of type A-20 have been used as adsorbents in the present study. Table 2 shows the specifications of the used adsorbents [12, 13].

3. Data extraction

The collected data of the [present](#) experimental work are mainly the pressure and temperature of the cells and the mass flow rate of the adsorbate. However, the most important data is the uptake amount (adsorption capacity) of the refrigerant by adsorbent which has been determined by the volumetric method. In order to determine the adsorbed amount of refrigerant at certain adsorption temperature, equations from 1 to 4 are used.

$$m_j = m_{j-1} + \Delta m_{load,j} - \Delta m_{void,j} \quad (1)$$

$$\Delta m_{load,j} = (\rho_{in} - \rho_f)_{load} (V_{load} + V_{tube}) \quad (2)$$

$$\Delta m_{void,j} = (\rho_{ads,j} - \rho_{ads,j-1})_{ads\ cell} (V_{void}) \quad (3)$$

$$V_{void} = V_{ads} - \left(\frac{m_{ads}}{\rho_{sol}} + V_{pore} \right) \quad (4)$$

4. Theoretical models

Many theoretical adsorption isotherms and kinetics models have been driven to simulate and predict the behavior of a certain adsorption/refrigeration pair. In order to determine the adjacent parameters of the different adsorption isotherms and kinetics models, the experimental results have been fitted with Tóth model and Dubinin-Astakhov (D-A) model with volume correction for adsorption isotherms and linear driving force (LDF) model for adsorption kinetics.

4.1 Tóth isotherm

This model can be expressed in Eq. 5 which is shown below [14].

$$\frac{C}{C_o} = \frac{bP}{[1+(bP)^t]^{\frac{1}{Z}}} \quad (5)$$

$$b = b_o e^{\left(\frac{H_{st}}{RT}\right)} \quad (6)$$

4.2.1 D-A model with volume correction

The D-A with volume correction is shown in Eq. 7, below [15].

$$C = \frac{w_o}{v_a} \exp \left\{ - \left[\frac{R_{gas}T}{E} \ln \left(\frac{P_s}{P} \right) \right]^n \right\} \quad (7)$$

The absolute adsorption C is derived from the measured data. The adsorbed phase volume (v_a) is calculated using the law of rectilinear diameters approach proposed by Srinivasan et al. [16].

$$v_a = \left[A - BT - \frac{1}{v_g} \right]^{-1} \quad (8)$$

v_g is the gas phase specific volume at the equilibrium pressure and temperature. p_s and v_g are calculated using REFPROP [17].

Considering the uncertainties in the measurement of all physical quantities such as temperature, pressure, mass of adsorbent, volumes of cells and effect of plumb line volume, the estimated uncertainty in the whole range of data are below $\pm 5\%$.

4.2.2 Isosteric heat of Adsorption

Isosteric heat of adsorption of an adsorbent/adsorbate pair is an essential variable for designing a practical gas storage systems or adsorption cooling systems. This section presents evaluation of the isosteric heat of adsorption of HFC410A onto ACP and ACF type adsorbents.

Isosteric heat of adsorption is traditionally expressed as a function of concentration as its dependence on temperature is relatively weak [18, 19]. Clausius-Clapeyron equation is commonly used to estimate heat of adsorption assuming ideal gas at constant concentration as given by Eq. 9.

$$H_{st} = \frac{-R\partial \ln P}{\partial(1/T)} \quad (9)$$

The analysis of the isosteric heat of adsorption starts using the D-A equation which could be rewritten in the form of Eq. 10.

$$\ln P = \ln P_s - \frac{E}{RT} \ln \left(\frac{W_0}{c v_a} \right)^{\frac{1}{n}} \quad (10)$$

$$h_{fg} = \left. \frac{-R\partial \ln P_s}{\partial(1/T)} \right|_C \quad (11)$$

$$H_{st} = h_{fg} + E \left[\ln \left(\frac{W_0}{c v_a} \right) \right]^{\frac{1}{n}} + \frac{ET\alpha}{n} \left[\frac{\ln(W_0)}{c v_a} \right]^{\frac{1-n}{n}} \quad (12)$$

$$\alpha = v_a \left(B - \frac{\alpha_g}{v_g} \right) \quad (13)$$

The thermal expansion coefficient of the gas phase (α_g) and the heat of vaporization (h_{fg}) are obtained from REFPROP.

4.3 LDF model

One of the widely used models in simulating the kinetics of the adsorption process is the LDF model which is presented below by Eq. 14.

$$\frac{c-c_{in}}{c_0-c_{in}} = 1 - \exp[-k_s a_v (t - t_{in})] \quad (14)$$

$$k_s a_v = F_0 \frac{D_s}{R_p^2} \quad (15)$$

Ruthven [20] reported that, during the initial phase of adsorption, surface diffusion coefficient follows the uptake behavior of a semi-infinite medium of any particle shape and can be expressed as,

$$\frac{c}{c_0} = \frac{2A}{V} \sqrt{\frac{D_s t}{\pi}} \quad (16)$$

A plot of relative uptake and square root of time yields with a slope of $\frac{2A}{V} \sqrt{\frac{D_s}{\pi}}$. So that the values of the surface diffusion could be extracted which are expressed as [21]:

$$D_s = D_{s0} \exp\left(\frac{-E_a}{R_g T}\right) \quad (17)$$

Which can be arranged as,

$$\ln(D_s) = \ln(D_{s0}) - \frac{E_a}{R_g T} \quad (18)$$

By plotting $\ln(D_s)$ against $1/T$, one can get the numerical values of activation energy, E_a in kJ/kg and the pre-exponential coefficient, D_{s0} in m^2/s . This plot is known as Arrhenius plot. The slope yields $-E_a/R_g$ where R_g is the gas constant in kJ/kg and the intercept provides D_{s0} [22].

5. Results and discussion

Figures 2 and 3 show the adsorption uptakes of ACP/HFC410A and ACF/HFC410A adsorption pairs, respectively. These figures show the adsorption capacity in kg/kg on the basis of the adsorption pressure at various adsorption temperatures ranging from 10°C to 50°C. The solid lines in the figures are indications for the D-A with volume correction and the dashed lines are indications for the Tóth equation, while the symbols are indications for the experimental results. Both Figs. 2 and 3 show that D-A and Tóth equations agree well with the present experimental adsorption isotherms data. However, the best agreement between the experimental and theoretical results could be seen with the D-A model. These figures indicate that the adsorption capacity of HFC410A on to ACP is 1.6 times higher than that of the ACF. This is because the relatively higher surface area and pore volume of ACP, which are about 1.6 times of those of the ACF.

Figure 4 shows the difference between experimental and calculated adsorption uptakes of HFC410A onto ACP and ACF. This figure shows that both theoretical isotherm models fit the experimental data within error range of $\pm 5\%$.

Table 3 gives Tóth isotherm parameters for ACP/HFC410A and ACF/HFC410A which have been obtained from the fitting between experimental and theoretical data. Table 4 introduces the fitting parameters of D-A equation with volume correction. The parameters A and B have been determined to be 0 and 0.0015 respectively for both ACP/HFC410A and ACF/HFC410A.

Figure 5 shows the maximum adsorption capacity based on Tóth equation for HFC410A, HFC32 and CO₂ onto ACP and ACF adsorbents [7, 8]. This figure indicates that ACP/HFC410A could achieve the highest adsorption capacity comparing to the other adsorption pairs.

Figure 6 and Figure 7 show the isosteric heats of adsorption which are obtained from the D-A equation at various adsorption temperatures for ACP/HFC410A and ACF/HFC410A respectively. The isosteric heat of adsorption has a reverse relationship with the temperature with respect to the relative capacity. These figures show that the isosteric heat of adsorption decreases by increasing the adsorption capacity, which means increasing either the adsorption capacity or the adsorption temperature, decreases the isosteric heat of both ACP/HFC410A and ACF/HFC410A adsorption pairs.

A logarithmic relation between H_{st} and Cv_a/W_0 is found to describe their interdependence along each isotherm mathematically which can be expressed as:

$$H_{st} = H_{st0} + \beta \ln \left(\frac{Cv_a}{W_0} \right) \quad (19)$$

The temperature dependences of H_{st0} and β are plotted in Fig. 8. A polynomial equation of second order describes the temperature dependence of H_{st0} while β seems to be fairly independent of temperature. Hence Eq. 19 is generalized as follows:

$$H_{st} = A_1 T^2 + A_2 T + A_3 + \beta \ln \left(\frac{Cv_a}{W_0} \right) \quad (20)$$

The numerical values of A_1 , A_2 , A_3 , β are listed in Table 5. Difference between the vapor phase enthalpy (h_g) and adsorbed phase enthalpy (h_a) has been calculated. However h_a has been determined using REFPROP based on the adsorbed phase volume (v_a). Figure 8 shows the temperature dependence of vapor-adsorption phase enthalpy difference (h_{ag}). It has been found that the ACP/HFC410A and ACF/HFC410A has same values for h_{ag} . A polynomial equation of second order is found to describe the temperature dependence of h_{ag} as below;

$$h_{ag} = -0.0143xT^2 + 6.5975xT - 526.03 \quad (21)$$

From Eq. 12 it can be seen that the heat of adsorption at limiting relative loading ($Cv_a/W_0=1$) is H_{st0} . It may be recalled that D-A isotherm does not have a Henry's law regime [23]. On the other hand, the Tóth isotherm does not depict a limiting pressure. Akkimaradi et al. [24] and Wang et al. [25] linked these two isotherms through Henry's law coefficient derived from Tóth isotherm and the isosteric heat of adsorption derived from D-A isotherm at limiting loading condition. Tóth equation yields the temperature dependence of the Henry's law coefficients straight away.

$$k_H = \lim_{p \rightarrow 0} \left. \frac{\partial C}{\partial p} \right|_T = C_0 b_0 \exp\left(\frac{H_{st}}{RT}\right) \quad (22)$$

Figure 9 shows the relation between k_H and $1/T$ based on Eq. 17. Figure 10 shows an increasing trend of H_{st0} with respect to k_H for both ACP/HFC410A and ACF/HFC410A pairs.

Empirical cubic equations relating H_{st0} and k_H are given below for ACP/HFC410A and ACF/HFC410A, respectively.

$$H_{st0} = 7 \times 10^8 (k_H)^3 - 2 \times 10^7 (k_H)^2 + 117902 (k_H) - 161.3 \quad (23)$$

$$H_{st0} = 10^9 (k_H)^3 - 2 \times 10^7 (k_H)^2 + 111125 (k_H) - 124.19 \quad (24)$$

Mathematical description of isotherms derived from experimental data together with other supplementary properties such as isosteric heat of adsorption and Henry's law coefficient extracted from those isotherms would be the primary information required for designing of adsorption storage system.

Figures 11 and 12 show the experimental adsorption capacity fitting by LDF model at different adsorption temperatures for ACP/HFC410A, and ACF/ HFC410A, respectively. These figures show relatively good agreement between the experimental data and the LDF model results. Table 6 shows the activation energy and pre-exponential coefficient values for ACP/HFC410A and ACF/HFC410A pairs.

6. Conclusions

The adsorption isotherms and kinetics of HFC410A onto ACP and ACF have been studied. An apparatus has been designed and built in order to conduct the experimental investigations. The maximum adsorption capacity of the ACP is found to be 1.6 times higher than that of ACF at same adsorption temperature. Both investigated adsorption isotherms (Tóth, D-A) models and kinetics (LDF) models have been found to fit the experimental data within $\pm 5\%$ deviations. [The isosteric heat of adsorption data for ACP/HFC410A and ACF/HFC410A pairs have been determined based on D-A equation.](#) Heats of adsorption at limiting relative loading and Henry's law coefficient have been determined for the studied adsorbent/refrigerant pairs. Empirical equations for the relation between temperature and heats of adsorption at limiting relative loading have been presented.

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Table captions:

Table 1. ODP, GWP and boiling temperature of some HFCs refrigerants.

Table 2. Specifications of the adsorbents.

Table 3. Fitting parameters of Tóth equation.

Table 4. Fitting parameters of D-A equation.

Table 5. Fitting parameters for combined temperature and loading dependence heat of adsorption.

Table 6. Activation energy and pre-exponential coefficient for ACP/HFC410A and ACF/410A.

Table 1. ODP, GWP and boiling temperature of some HFCs refrigerants.

Refrigerant	ODP	GWP	Normal Boiling Temp.°C
HFC32	0	675	-52
HFC134a	0	1430	-26.3
HFC152a	0	124	-25
HFC407a	0	2107	-45.2
HFO1234 yf	0	4	-30
HFO1234 ze(E)	0	6	-19
HFC410A	0	2088	-51.6
HFC507A	0	3985	-46.7
HFC123	0.02	77	-27.6
HFC124	0.022	609	-12
HFC502	0.249	4657	-45

Table 2. Specifications of the adsorbents.

Parameter	ACP Maxsorb III	ACF A-20
Mass, kg	50.27×10^{-3}	50.55×10^{-3}
Pore volume, m^3/kg	1.7×10^{-3}	1.03×10^{-3}
Solid density, kg/m^3	2200	800
Surface area, m^2/kg	3140×10^3	2000×10^3
Particle diameter, m	72×10^{-6}	21.6×10^{-6}

Table 3. Fitting parameters of Tóth equation.

Parameters	ACP/HFC410A	ACF/HFC410A
C_0 , kg/kg	4.18	1.68
b_0 , kPa ⁻¹	1.4x10 ⁻⁶	1.84x10 ⁻⁶
z	0.68	0.83
H_{st} , kJ/kg	237	249

Table 4. Fitting parameters of D-A equation.

Parameters	ACP/HFC410A	ACF/HFC410A
$W_0, \text{cm}^3/\text{g}$	5.96	3.25
$E, \text{kJ/kg}$	59.6	72.5
n	1.17	1.43

Table 5. Fitting parameters for combined temperature and loading dependence heat of adsorption.

Parameter	ACP	ACF
A_1 , kJ/kg.K	- 0.019	- 0.008
A_2 , kJ/kg.K	10.052	2.951
A_3 , kJ/kg	-1172.2	-23.792
β	59	56

Table 6. Activation energy and pre-exponential coefficient for ACP/HFC410A and ACF/410A.

Parameter	ACP	ACF
$E_a, kJ/kg$	16	85.4
$D_{s0}, m^2/s$	1.3×10^{-13}	2.8×10^{-13}

Figure captions:

Fig. 1. Schematic diagram of the experimental apparatus for adsorption characterization.

Fig. 2. Fitting of Tóth and D-A with volume correction models with experimental data for ACP/HFC410A.

Fig. 3. Fitting of Tóth and D-A with volume correction models with experimental data for ACF/HFC410A.

Fig. 4. Comparison of adsorption uptake deviations between experimental uptake and calculated uptakes.

Fig. 5. Maximum adsorption capacity based on Tóth equation for six different adsorption pairs.

Fig. 6. Isothermic heat of adsorption at different adsorption temperatures for ACP/HFC410A.

Fig. 7. Isothermic heat of adsorption at different adsorption temperatures for ACF/HFC410A.

Fig. 8. Temperature dependence of fitting parameters of isothermic heat of adsorption in Eq 15.

Fig. 9. Henry's coefficient as a function of $1/T$ for ACP/HFC410A and ACF/HFC410A.

Fig. 10. Relation between k_H and H_{st0} .

Fig. 11. Fitting of LDF model with experimental adsorption kinetics at different adsorption temperatures for ACP/ HFC410A.

Fig. 12 Fitting of LDF model with experimental adsorption kinetics at different adsorption temperatures for ACF/ HFC410A.

P: Pressure transducer, T: Thermocouple, F.M: Flow meter

Fig. 1. Schematic diagram of the experimental apparatus for adsorption characterization.

Fig. 2. Fitting of Tóth and D-A with volume correction models with experimental data for ACP/HFC410A.

Legend: O- 10°C, $\square$ - 20°C, $\square$ - 30°C, $\diamond$ - 40°C, x- 50°C, solid line- D-A equation, dashed lines- Tóth equation.

Fig. 3. Fitting of Tóth and D-A with volume correction models with experimental data for ACF/HFC410A.

Legend: O- 10°C, $\square$ - 20°C, $\square$ - 30°C, $\diamond$ - 40°C, x- 50°C.

Fig. 4. Comparison of adsorption uptake deviations between experimental uptake and calculated uptakes.

Legend: ACP- open symbols; ACF- filled symbols; O- Tóth equation; Δ - D-A equation.

Fig. 5. Maximum adsorption capacity based on Tóth equation for six different adsorption pairs.

Fig. 6. Isosteric heat of adsorption at different adsorption temperatures for ACP/HFC410A.

Fig. 7. Isosteric heat of adsorption at different adsorption temperatures for ACF/HFC410A.

Fig. 8. Temperature dependence of fitting parameters of isosteric heat of adsorption in Eq. 15.

Legend: ACP- open symbols; ACF- Filled symbols; O- H_{st0} ; Δ - β , $\square$ - h_{ag}

Fig. 9. Henry's law coefficient for ACP/HFC410A and ACF/ HFC410A.

Legend: ACP- open symbols; ACF- filled symbols.

Fig. 10. Relation between k_H and H_{st0} .

Legend: ACP- open symbols; ACF- filled symbols.

Fig. 11. Fitting of LDF model with experimental adsorption kinetics at different adsorption temperatures for ACP/HFC410A.

Fig. 12 Fitting of LDF model with experimental adsorption kinetics at different adsorption temperatures for ACF/HFC410A.