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**NOVEL METHOD USING DUBININ-ASTAKHOV THEORY IN SORPTION
REACTOR DESIGN FOR REFRIGERATION AND HEAT PUMP**

APPLICATIONS

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ABSTRACT - This paper presents new methodology that is based on a single given adsorbent-refrigerant pair characteristic (such as activated carbon 208C) leading to the characterization of the same adsorbent (activated carbon 208C) with various refrigerants such as Water, Methanol, Ethanol, R723 (azeotropic mixture of 60% Ammonia and 40% Dimethyl Ether), Dimethyl Ether (DME) and Carbon Dioxide (R744). Overall, the results obtained with both Methanol and Carbon Dioxide (R744) show that the new method predicts the refrigerant uptake with a marginal difference (less than 5%) compared to standard method that heavily depends on experimental data. For example with methanol, the standard method produces a maximum uptake (x_o) of 0.3676 kg methanol/kg carbon while the new method predicts 0.3740 kg methanol/kg carbon; with CO₂ both standard and new methods predict 0.3242 kg CO₂/kg carbon and 0.3190 kg CO₂/kg carbon respectively. The results exploitation of this method led to rapid prediction of key performance indicators of adsorption system utilizing compacted activated carbon 208C-R723 refrigerant pair for ice making, air conditioning and heat pump applications.

Keywords: Adsorption, Ammonia (R717), Ammonia blend (R723), Refrigeration, Heat pump, COP, Dubinin-Astakhov

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NOMENCLATURE

<i>A</i>	Refrigerant characteristic coefficient (1/K)
<i>B</i>	Refrigerant constant characteristic
<i>COP</i>	Coefficient of performance
Cu90Ni10	Copper alloy (90% Copper and 10% Nickel)
DME	Dimethyl Ether
<i>E</i>	Adsorption potential energy (J/mol)
<i>k</i>	Dubinin coefficient
<i>M</i>	Molar mass (g/mol)
<i>n</i>	Dubinin coefficient
<i>Q</i>	Heat (J/kg)
<i>P</i>	Pressure (Pa or bar)
<i>R</i>	Gas constant (J/kg K) or (J/mol K)
R717	Ammonia refrigerant
R723	Ammonia blend refrigerant (60% R717 / 40% DME)
RMSD	Root Mean Square Deviation
<i>v</i>	Specific volume (m ³ /kg)
<i>V</i>	Molar volume (m ³ /mol)
SEE	Standard Estimated Error
<i>T</i>	Temperature (K or °C)
<i>x</i>	Concentration (kg Refrigerant /kg Carbon)
Greek letters	
β	Affinity coefficient
γ	Surface Tension (N/m)
Δ	Difference, variation
ρ	Density (kg/m ³)
Ω	Parachor (m ^{11/4} N ^{1/4} /mol)

Subscripts

<i>C</i>	Condensing, condensation
<i>E</i>	Evaporating, evaporation
<i>G</i>	Generating
<i>in</i>	Input
<i>L</i>	Liquid phase
<i>o</i>	Maximum, Under saturation, Reference
<i>rec</i>	Recovery, Recovering
<i>reg</i>	Regeneration
<i>reject</i>	Rejection, Rejecting
<i>s</i>	specific
<i>sat</i>	Under saturation conditions
<i>u</i>	Universal

1. INTRODUCTION

The design of adsorption reactors for refrigeration and heat pumps applications generally requires the mapping of refrigerant specific uptake against operating conditions mainly temperatures and pressures. A large number of experimental data for the pair studied (adsorbent-refrigerant) are often needed in order to identify the three key parameters of a modified expression of Dubinin-Astakhov equation: x_o (maximum uptake of refrigerant by the adsorbent – kg refrigerant per kg of adsorbent), k (energetic affinity characteristic of adsorbent-refrigerant pair) and n (characteristic of adsorbent micro-pores size distributions) [1-3]. Furthermore this lengthy mapping process that is useful to estimate the potential cooling or heating capacity or COP (coefficient of performance) is always specific to a given adsorbent-refrigerant. This means for instance that with the same adsorbent, each refrigerant will require its experimental test for

mapping which is not cost effective as soon as you would like to screen many refrigerants [4]. Preliminary method of exploring ways of minimizing the number test has already been carried out and some results were presented in 2014 International Sorption Heat Pump Conference [5]. Since then, and on the light of conference presentation feedback, further work was done in providing more clarity and the validity of the method across a large spectrum of pressures (from under atmospheric pressure up to 74 bar absolute). The novel methodology proposed in this paper is based on a single given adsorbent-refrigerant pair map (activated carbon 208C – Ammonia pair) leading to the map of the same adsorbent (Activated Carbon 208C) with various refrigerants such as Water (R718), Methanol, Ethanol, R723 (azeotropic mixture of 60% Ammonia and 40% Dimethyl Ether), Dimethyl Ether (DME) and Carbon Dioxide (R744). The method will be validated by comparing predicted simulation results with standard modified Dubinin-Astakhov equation established with experimental data. Furthermore, results exploitation of this method will lead to a rapid prediction of key indicators performance of adsorption system utilizing compacted activated carbon 208C-R723 refrigerant pair for ice making, air conditioning and heat pump applications.

2. METHODOLOGY DESCRIPTION

The method proposed is articulated around a reference adsorbent-refrigerant pair as activated carbon 208C-Ammonia that was initially fully tested in order to provide the adsorption characteristics. For this specific reference pair, experimental porosity tests were carried out under the following operating conditions: the condensing temperatures varied from 20°C to 50°C ; the evaporating temperature ranges from -20°C to 20°C and the bed temperature range from 35°C to

200°C. The Ammonia refrigerant uptakes were measured during each experimental test and the data were fitted to a modified form of the Dubinin-Astakhov (D-A) equation [2]:

$$x = x_o \exp \left[-k \left(\frac{T}{T_{sat}} - 1 \right)^n \right] \quad (1)$$

where: x is the ammonia concentration (kg ammonia/kg carbon); T is the carbon temperature (K); x_o is the ammonia concentration under saturation conditions (kg ammonia/kg carbon); T_{sat} is the saturation temperature corresponding to the gas pressure (K); k is defined as the energetic affinity characteristic of adsorbent-refrigerant pair and n is the characteristic of adsorbent micro-pores size distributions [3].

x_o , k and n are also commonly called Dubinin coefficients. With activated carbon 208C-ammonia which is our reference pair: $x_o = 0.3077$ kg ammonia/kg carbon; $k=4.439$ and $n=1.187$ [6]. From now on, the adsorption characterization of activated carbon 208C with a refrigerant different than Ammonia consists of three steps:

a) Evaluation of n : The n value provided by the unique test carried out is an intrinsic characteristic of the adsorbent since it mainly depends on the current activated carbon micro-pores size distributions. It is therefore considered constant given by the experimental tests carried with activated carbon 208C and ammonia refrigerant [6]:

$$n = 1.187 \quad (2)$$

b) Evaluation of x_o : The adsorbed refrigerant is always assumed to be in a liquid form located in the adsorbent micro-pores [7]. Therefore the maximum uptake of refrigerant by the adsorbent (x_o) is calculated from the specific volume of micro-pores which is often provided by the adsorbent manufacturer (for activated carbon 208C, $v_s = 0.500$ cm³/g) and the refrigerant liquid density at normal pressure condition (atmospheric pressure) (ρ_L):

$$x_o = \rho_L V_s \quad (3)$$

c) Evaluation of k:

In fact the original Dubinin-Astakhov (D-A) equation is

$$x = x_o \exp \left[- \left(\frac{R_u A}{\beta E_o} \right)^n \left(\ln \frac{P_{sat}}{P} \right)^n \right] \quad (4)$$

where: x is the ammonia concentration (kg ammonia/kg carbon); x_o is the ammonia concentration under saturation conditions (kg ammonia/kg carbon); P_{sat} is the saturation pressure corresponding to adsorbed phase within the micro-pores (bar); P is the system pressure (bar); A is the slope of saturated adsorbate line on Clapeyron diagram; R_u is the universal gas constant ($R_u = 8.3144$ J/mol K); E_o is the reference adsorption potential energy of adsorbent (J/mol); β gas affinity coefficient and n is the characteristic of adsorbent micro-pores size distributions.

The relation between saturation pressures (P_{sat}) and temperatures (T_{sat}) as commonly used on Clapeyron diagram is:

$$\ln P_{sat} = B - \frac{A}{T_{sat}} \quad (5)$$

where: A and B are constants.

By combining equation (4) with equation (5), Dubinin-Astakhov (D-A) equation could also be written as:

$$x = x_o \exp \left[- \left(\frac{R_u A}{\beta E_o} \right)^n \left(\frac{T}{T_{sat}} - 1 \right)^n \right] \quad (6)$$

From both equations (1) and (6), the energetic affinity characteristic of adsorbent-refrigerant pair (k) is identified as:

$$k = \left(\frac{AR_u}{\beta E_o} \right)^n \quad (7)$$

where: A is the slope of saturated adsorbate line on Clapeyron diagram; R_u is the universal gas constant ($R_u = 8.3144$ J/mol K); E_o is the adsorption potential energy of adsorbent (J/mol); β gas affinity coefficient and n is the characteristic of adsorbent micro-pores size distributions.

The adsorption potential energy of adsorbent (E_o) is often experimentally estimated with Benzene as the reference refrigerant. However with the current method and by neglecting any effect of thermal expansion of adsorbate phase, E_o is evaluated from test of activated carbon with Ammonia by using the following equation:

$$E_o = \frac{AR_u}{\beta k^{1/n}} \quad (8)$$

The gas affinity coefficients are widely available in the literature and could also be calculated from the ratio of Parachors [8]:

$$\beta = \frac{\Omega}{\Omega_o} \quad (9)$$

where the Parachor (Ω) is defined as:

$$\Omega = V\gamma^{1/4} \quad (10)$$

Where V is the gas molar volume (m^3/mol) and γ is gas surface tension (N/m). Ω_o is defined as the Parachor of the reference gas (Benzene).

By combining both equations (9) and (10), the gas affinity coefficient could also be written as:

$$\beta = \frac{V}{V_o} \left(\frac{\gamma}{\gamma_o} \right)^{1/4} \quad (11)$$

Where V_o and γ_o are defined as molar volume (m^3/mol) and surface tension (N/m) of the reference gas (Benzene) respectively.

Since the gas molar volume is defined as,

$$V = \frac{M}{\rho} \quad (12)$$

where: M is the gas molar mass (kg/kmol) and ρ is the gas density at boiling temperature (kg/m^3),

$$\beta = \frac{M}{M_o} \frac{\rho_o}{\rho} \left(\frac{\gamma}{\gamma_o} \right)^{1/4} \quad (13)$$

where: M_o and ρ_o are the molar mass (kg/kmol) of and the gas density (kg/m^3) of reference gas (Benzene) respectively.

3. RESULTS AND ANALYSIS

The key characteristics of all refrigerants under investigation, namely Ammonia (R717), Ammonia Blend (R723 - Azeotropic mixture of 60% Ammonia and 40% Dimethyl Ether), Benzene, Carbon Dioxide or CO_2 (R744), Dimethyl Ether (DME), Methanol, Ethanol and Water (R718), used to test the proposed method are presented in **Table 1**. All refrigerants thermo-physical properties required for calculations are extracted for literature [9, 10]. All gas affinity coefficients were calculated using Equation (13) with the exception of Carbon Dioxide refrigerant where the value of $\beta=0.35$ is taken from the literature [11, 12]. Furthermore, due to the supercritical nature of R744 (CO_2) which has a saturation pressure window in liquid form ranging from 5.2 bar (triple point) to 73.6 bar (critical point), it is therefore impossible to consider a liquid form at atmospheric pressure. However, Carbon Dioxide in adsorbed form is often considered as superheated liquid [13]. We have therefore assumed a minimum superheat of

about 2°C at around critical pressure (73 bar): the estimated value is $\rho_{L\text{CO}_2} = 638 \text{ kg/m}^3$ [14]. The estimated value of the reference adsorption potential energy from test of activated carbon 208C with Ammonia is:

$$E_o = 20.566 \text{ kJ/mol} \quad (14)$$

The values of Dubinin coefficients using the new technique are summarized in **Table 2**. The current coefficients are compared mainly with activated carbon 208C with both Methanol and CO₂ corresponding to sub-atmospheric pressure and high pressure refrigerants respectively. The results are shown in **Figure 1**: the Standard Estimated Errors (SEEs) are about $\pm 0.006 \text{ kg/kg}$ carbon for Carbon Dioxide (R744) and $\pm 0.011 \text{ kg/kg}$ carbon for Methanol. Those differences between predictions of the current proposed method and those from the standard Dubinin method are considered marginal since the typical uptake swing (Δx) during the desorption phase for adsorption refrigeration and heat pump applications ranges between 0.15 and 0.25 kg/kg. Therefore the maximum error on the estimation of the cooling production or heating capacities is about 4% while taking on board the SEE on carbon-Methanol pair for standard Dubinin method ($\pm 0.0045 \text{ kg/kg}$ carbon) as shown in **Table 3**. Overall the new method could predict the refrigerant concentration within a Root Mean Square Deviation (RMSD) of about $\pm 1\%$ compared to the standard method. **Figure 2** shows the refrigerant concentration variation function of operating conditions reflected by $(T/T_{\text{sat}}-1)$: it leads to the following general observations regardless the application:

- Both R744 (CO₂) and RE170 (DME) have low uptake ability and very strong bond with carbon adsorbent and often operate with high driving temperature as 200°C or above.

This observation is well in line with experimental evidence [15, 16].

- Methanol, Ethanol, R723, R717 (Ammonia) are classified as average uptake ability and average bond with carbon adsorbent and will operate with average driving temperature (75°C-150°C). It will be recommended not to exceed 150°C driving temperature in practice with both Methanol and Ethanol in order to avoid refrigerant thermal stability leading chemical decomposition.
- R718 (water) has high uptake ability with weak bond with carbon and will require driving temperature below 100°C.

4. RESULTS EXPLOITATION

As example of exploitation of method described, the reactor performance are predicted using a thermodynamic model already developed and as described in **Figure 3 [6]** and which requires amendment of refrigerant properties mainly. The key performance indicators of an adsorption system, namely both cooling and heat capacities and COPs are estimated. Those estimated performance are carried out with compacted activated carbon 208C-R723 refrigerant pair under typical operating conditions for three applications (for ice making, air conditioning and heat pump): condensing temperature $T_C = 35^\circ\text{C}$, evaporating temperature $T_E = -5^\circ\text{C}$ and initial bed temperature $T_1 = 35^\circ\text{C}$ for ice making; $T_C = 35^\circ\text{C}$, $T_E = 10^\circ\text{C}$ and $T_1 = 35^\circ\text{C}$ for air conditioning and $T_C = 40^\circ\text{C}$, $T_E = 5^\circ\text{C}$ and $T_1 = 40^\circ\text{C}$ for heat pump. The driving temperature varies from 80°C to 200°C. For each application, single bed, 2 beds and infinite number of beds (ideal) configurations are investigated. For more than 2 beds, the cycle is regenerative (with heat recovery) with a cross beds temperature difference ΔT of 10 K (typical practical value). For all applications, the density of compacted activated carbon 208C is 770 kg/m³.

4. 1. Ice making application ($T_C = 35^\circ\text{C}$, $T_E = -5^\circ\text{C}$ and $T_1 = 35^\circ\text{C}$)

Figure 4 shows different performance indicators of the system. The heat input (Q_{in}) of each bed is calculated by integration of the effective specific heat ($\Delta Q/\Delta T$) along the process path from the initial temperature T_1 to the generating temperature T_3 (200°C for illustration) as described by Meunier [18]. Furthermore both heat rejected (Q_{reject}) and regenerated (Q_{reg}) could also be estimated with the same method. The COP, the specific cooling energy and cooling density increase with the driving temperature (T_3) as expected. Regardless the bed configurations and driving temperatures, overall, the COP is about 30% to 50% less than the COP with compacted activated carbon 208C-R717 refrigerant pair [6]: for example with 2-beds configuration and a driving temperature of 200°C, the current pair has a COP of about 0.40 against 0.58 with compacted activated carbon 208C-R717 refrigerant pair. Similarly for the same driving temperature (200°C) with the current pair the cooling density is 102.3 MJ/m³ against 131.6 MJ/m³ for previous pair with R717 (corresponding to a specific cooling of about 132.8 kJ/kg against about 170.8 kJ/kg). This lower performance is mainly due to the combination of two factors: the latent heat of R723 is about 28% lower than the latent heat of R717 and the bond of R723 refrigerant with activated carbon 208C is higher compared to the bond with R717 refrigerant therefore requires more heat input for the desorption process (about 12% more).

4. 2. Air conditioning application ($T_C = 35^\circ C$, $T_E = 10^\circ C$ and $T_1 = 35^\circ C$)

Figure 5 shows that the system performance for air conditioning application is similar to ice making application in respect of performance trend with almost the same proportion of low COP and cooling density compared to R717 refrigerant: COP of 0.49 against 0.73 and cooling density is 132.6 MJ/m³ against 172.9 MJ/m³ for 2-beds configuration at 200°C driving temperature.

4. 3. Heat pump application ($T_C = 40^\circ C$, $T_E = 5^\circ C$ and $T_1 = 40^\circ C$)

If both lower latent heat of R723 and higher bond of R723 with activated carbon 208C compared to R717 are highly detrimental for cooling applications (ice making and air conditioning), it is less so with heat pump application. In fact, the requirement of high heat input for desorption process will also imply high heat of adsorption which is useful in heat pump application (**Figure 6**). It means for a regenerative cycle both heat recovered (Q_{rec}) and latent heat from the condensation. As consequence, regardless the bed configurations and driving temperatures, the COP is about 7% to 20% less than the COP with compacted activated carbon 208C-R717 refrigerant pair [6]: for example with 2-beds configuration and a driving temperature of 200°C, the current pair has a COP of about 1.40 against 1.62 with compacted activated carbon 208C-R717 refrigerant pair ; the heating density is 373.2 MJ/m³ against 367.6 MJ/m³ for previous pair with R717 (corresponding to a specific heating of about 484.7 kJ/kg against about 477.4 kJ/kg). This exploitation of the new method of characterization of adsorbent-refrigerant pair has shown the potential of R723 refrigerant for adsorption in refrigeration and heat pump applications. The R717 refrigerant has better performance than R723 when paired with activated carbon 208C for adsorption refrigeration and heat pump applications. However, since R723 is compatible with refrigeration copper alloy (Cu90Ni10) [19], this offers a trade-off between the cost of both Cu90Ni10 and stainless steel 316 (which compatible with R717), the bed overall heat transfer performance and COP.

5. CONCLUSIONS

A novel methodology that is based on a single given adsorbent-refrigerant pair characteristic (example activated carbon 208C–Ammonia pair) leading to the characterization of the same adsorbent (activated carbon 208C) has been described. The new method is tested with Methanol, Ethanol, R723 (azeotropic mixture of 60% Ammonia and 40% Dimethyl Ether) and R744

(Carbon Dioxide). Overall, with exception of R744 (Carbon Dioxide), the results obtained shows a marginal difference (less than 5%) compared to standard method that heavily depends on experimental data. For example with methanol, the standard method produces a maximum uptake (x_o) of 0.3676 kg methanol/kg carbon while the new method predicts 0.3740 kg methanol/kg carbon; with CO₂ both standard and new methods predict 0.3242 kg CO₂/kg carbon and 0.3190 kg CO₂/kg carbon respectively. The exploitation of the proposed method leads to a quick and cost effective prediction of key performance indicators of adsorption system utilizing compacted activated carbon 208C-R723 refrigerant pair for ice making, air conditioning and heat pump applications. The proposed methodology also offers the possibility to characterize an adsorbent using less hazardous gases such as Nitrogen (N₂) or Helium (He) and rapidly screen its potential with a range refrigerants or the desired initial refrigerant.

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Refrigerant	M (g/mol)	T _{Boiling} (°C)	A	B	ρ (kg/m ³)	$\gamma \times 10^3$ (N/m)	β
Ammonia (R717)	17.03	-33.3	2823.40	11.749	682	44.85	0.325
Ammonia Blend (R723)	22.77	-36.4	2689.60	11.300	708	22.10	0.339
Benzene	78.11	80.1			814	21.19	1
Carbon dioxide (R744)	44.01	N/A	1988.34	10.833	638*	N/A	0.350 [9, 10]
Dimethyl Ether (RE170)	46.07	-24.8	2625.75	10.582	735	19.3	0.638
Ethanol	46.01	78.5	5040.30	14.302	757	17.30	0.602
Methanol	32.04	64.6	4631.10	13.600	748	18.97	0.434
Water (R718)	18.02	100	8934.60	36.900	959	58.91	0.253

Table 1: Refrigerants properties (* Density of superheated liquid taken at 73 bar)

Refrigerants	R718 (Water)	Methanol	Ethanol	RE170 (DME)	R723	R744 (CO2)
x_o (kg/kg)	0.4795	0.3740	0.3785	0.3675	0.3540	0.3340
k	23.4723	5.6764	4.2518	1.8297	3.9928	2.7906
n	1.187	1.187	1.187	1.187	1.187	1.187

Table 2: Dubinin coefficients from the proposed method

<i>Refrigerants</i>	Methanol			R744 (CO2)		
	Standard	New	<i>Difference</i>	Standard [15]	New	<i>Difference</i>
x_o (kg/kg)	0.3676	0.3740	<i>0.0065</i>	0.3242	0.3190	<i>-0.0052</i>
k	6.3147	5.6764	<i>-0.6383</i>	2.5135	2.6831	<i>0.1696</i>
n	1.180	1.187	<i>0.007</i>	1.1602	1.187	<i>-0.0268</i>
SEE (kg/kg)	± 0.0045	N/A	<i>N/A</i>	± 0.0087	N/A	<i>N/A</i>

Table 3: Methods comparison

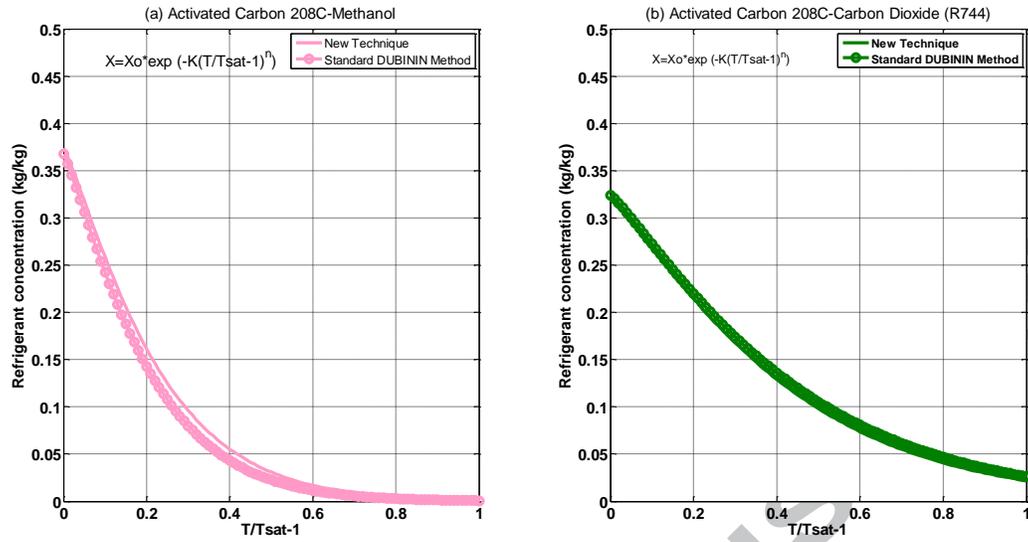


Figure 1: Comparison between the two methods: standard Dubinin method and new technique

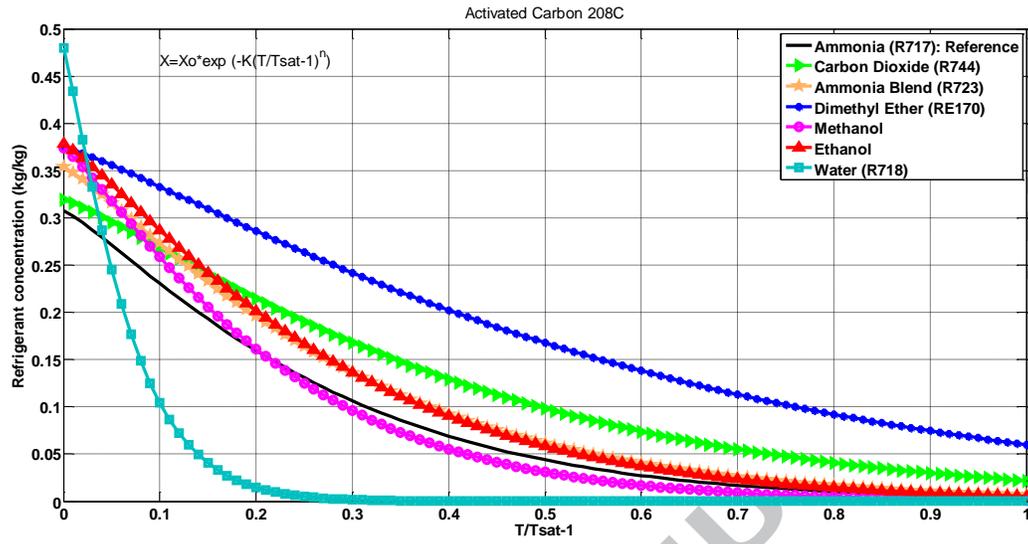


Figure 2: Refrigerants concentration with Activated Carbon 208C with the proposed method

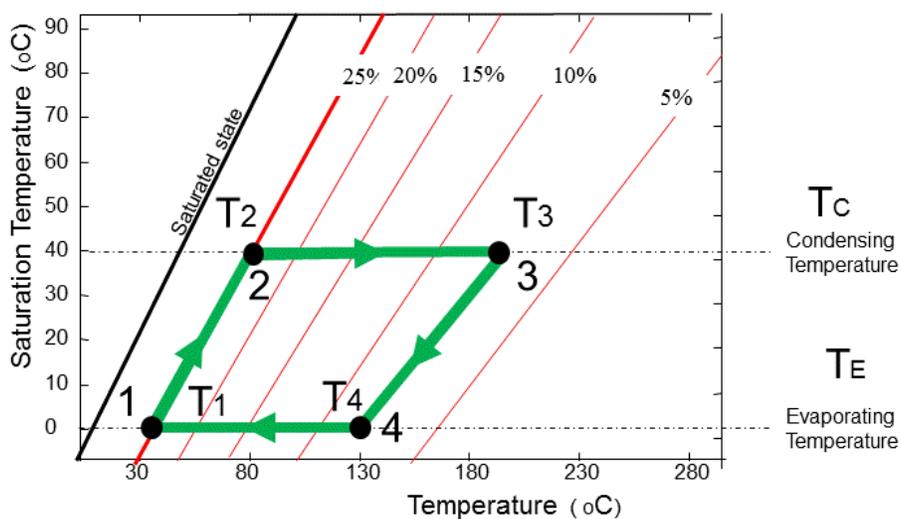


Figure 3: Adsorption thermodynamic cycle. 1 to 2 and 2 to 3: Isosteric and isobaric heat input respectively (desorption process). 3 to 4 and 4 to 1: Isosteric and isobaric cooling (rejected heat) respectively (adsorption process) [6]

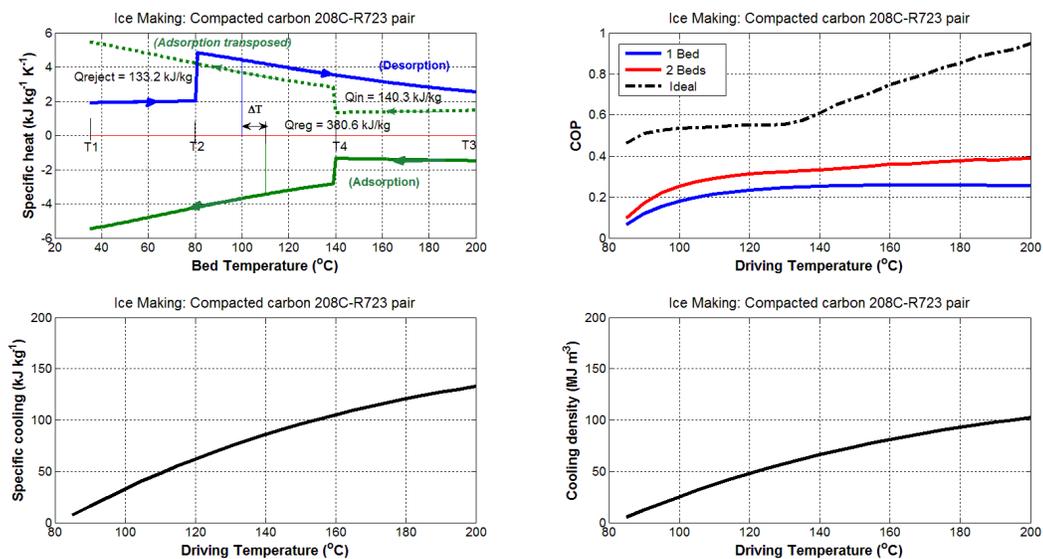


Figure 4: Ice making adsorption system key performance indicators

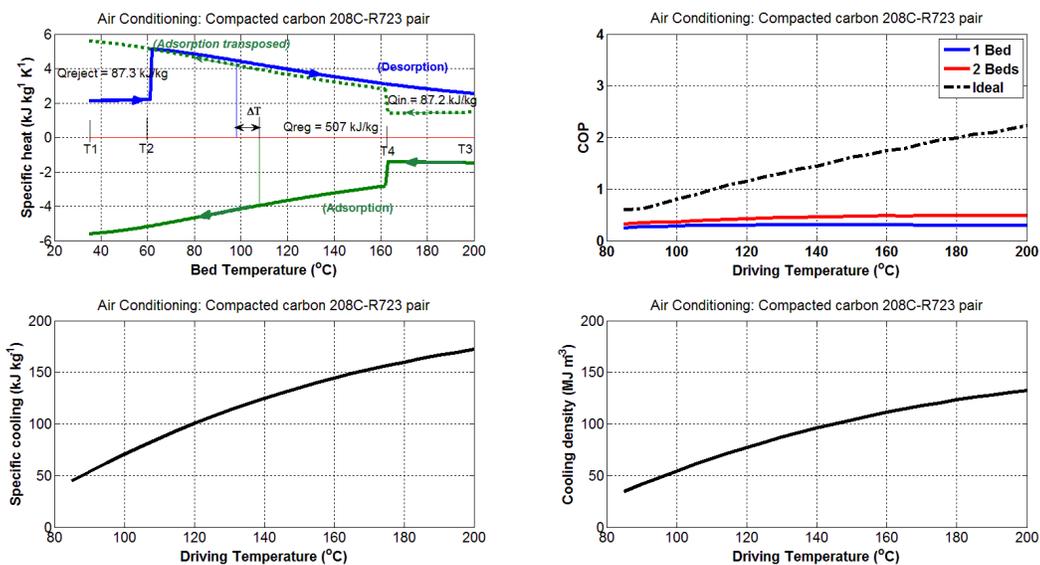


Figure 5: Air conditioning adsorption system key performance indicators

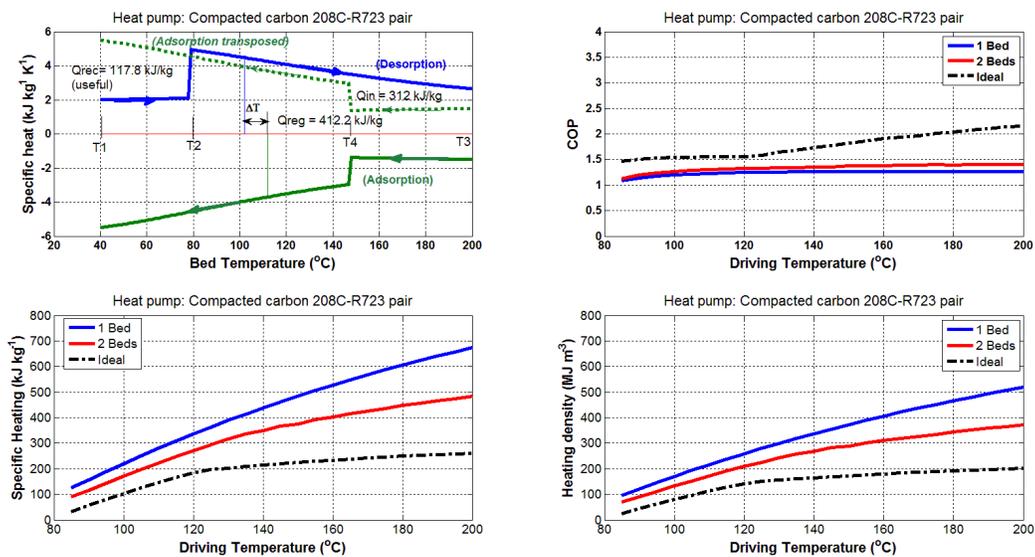


Figure 6: Heat pump adsorption system key performance indicators

HIGHLIGHTS

- A method of characterizing adsorbent-refrigerant pair is described.
- The methodology is based on Dubinin-Astakhov theory.
- Activated Carbon-Ammonia pair leads to same adsorbent with other refrigerants.
- The method proposed predicts pair characteristics within $\pm 5\%$ estimated error.
- The method is cost effective in predicting key performance indicators.