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Kutub Uddin

Kyushu University, Japan, k-uddin@phase.cm.kyushu-u.ac.jp

Ibrahim I. El-Sharkawy

ielsharkawy@phase.cm.kyushu-u.ac.jp

Takahiko Miyazaki

Kyushu University, Japan, miyazaki.takahiko.735@m.kyushu-u.ac.jp

Bidyut Baran Saha

Kyushu University, Japan, saha.baran.bidyut.213@m.kyushu-u.ac.jp

Shigeru Koyama

Kyushu University, Japan, koyama@phase.cm.kyushu-u.ac.jp

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Thermodynamic Analysis of Adsorption Refrigeration Cycles Using Parent and Surface Treated Maxsorb III/Ethanol Pairs

Kutub UDDIN^{1,6*}, Ibrahim I. EL-SHARKAWY^{2,6}, Takahiko MIYAZAKI^{3,6},
Bidyut Baran SAHA^{4,6}, Shigeru KOYAMA^{5,6}

¹Interdisciplinary graduate school of engineering sciences, Kyushu University,
6-1 Kasuga-Koen, Kasuga-shi, Fukuoka 816-8580, Japan
E-mail: k-uddin@phase.cm.kyushu-u.ac.jp

²Faculty of Engineering Sciences, Kyushu University,
6-1 Kasuga-Koen, Kasuga-shi, Fukuoka 816-8580, Japan
Mechanical Power Engineering Department, Faculty of Engineering
Mansoura University, El-Mansoura 35516, Egypt
E-mail: ielsharkawy@phase.cm.kyushu-u.ac.jp

³Faculty of Engineering Sciences, Kyushu University,
6-1 Kasuga-Koen, Kasuga-shi, Fukuoka 816-8580, Japan
E-mail: miyazaki.takahiko.735@m.kyushu-u.ac.jp

⁴Interdisciplinary graduate school of engineering sciences, Kyushu University,
6-1 Kasuga-Koen, Kasuga-shi, Fukuoka 816-8580, Japan
E-mail: saha.baran.bidyut.213@m.kyushu-u.ac.jp

⁵Faculty of Engineering Sciences, Kyushu University,
6-1 Kasuga-Koen, Kasuga-shi, Fukuoka 816-8580, Japan
E-mail: koyama@phase.cm.kyushu-u.ac.jp

⁶International Institute for Carbon-Neutral Energy Research (WPI-I2CNER),
Kyushu University, 744 Motoooka, Nishi-ku, Fukuoka 819-0395, Japan

*Corresponding Author, E-mail: k-uddin@phase.cm.kyushu-u.ac.jp

ABSTRACT

Adsorption equilibrium uptake of environment friendly refrigerant ethanol onto highly porous activated carbon based adsorbents has been experimentally investigated by using a magnetic suspension balance adsorption measurement unit (MSB-VG-S2). Adsorbents used in the present study are parent Maxsorb III, H₂ and KOH-H₂ surface treated Maxsorb III. Experiments have been conducted over adsorption temperatures range from 30 to 70°C and evaporation temperatures between -6 and 65 °C. The Dubinin-Radushkevich and Dubinin-Astakhov adsorption isotherm models have been used to correlate adsorption isotherm data and to plot the pressure-temperature-concentration (P-T-W) diagrams of the assorted pairs. Isothermic heat of adsorption is estimated using the Clausius–Clapeyron equation. In the present study, the performance of adsorption refrigeration cycles using activated carbons/ethanol pairs has also been investigated employing a time-independent mathematical model. Results are compared with other adsorbent/refrigerant pairs found in the open literatures. Theoretical analysis show that the H₂-treated Maxsorb III/ethanol adsorption refrigeration cycle can achieve coefficient of performance (COP) of 0.51 and specific cooling effect of about 374 kJ/kg at the evaporator temperature of -5 °C in combination with heat source and heat sink temperatures of 100 and 30 °C, respectively.

1. INTRODUCTION

Thermally driven adsorption refrigeration and heat pump systems got considerable attention nowadays due to its manufacturing simplicity and environment friendly adsorbent/refrigerant pairs. The research on this area intensified after the imposition of international restrictions on the production and use of CFCs (chlorofluoro-carbons) and HCFCs (hydrochlorofluoro-carbons), which are the commonly used refrigerants for cooling applications. These gases are identified as the major contributors to deplete the ozone layer around the globe (Miller, 1929; Aittomaki and Hakonen, 1986; Critoph, 1989; Meunier, 1993; Cacciola and Restuccia, 1994). The adsorption cooling and heat pump systems could utilize low temperature waste heat or renewable energy sources. The working pairs of adsorption cooling and heat pump are mainly dominated by silica gel/water (Yanagi *et al.*, 1992), zeolite/water (Rothmeyer *et al.*, 1983), activated carbon/ammonia (Critoph, 1989), activated carbon/methanol (Wang *et al.*, 2003) and activated carbon fiber (ACF)/ammonia (Vasiliev *et al.*, 2001) pairs.

Many researchers evaluated the performance of adsorption cooling and heat pump systems based on working pairs, system design and methodology. A transient simulation model for adsorption cooling system using silica gel/water pair powered by renewable energy was investigated (Sakoda and Suzuki, 1984). Saha *et al.* (1995) developed a cycle simulation program to investigate the influence of operating conditions on cooling output and COP of a single-stage silica gel/water adsorption chiller and found hot water temperature 50°C is only viable when cooling water temperature of less than 25°C. Boelman *et al.* (1995) was continued to study parametrically the influence of thermal capacitance and heat exchanger UA-values on cooling capacity for the silica gel/water system. Saha *et al.* (2003) also evaluated the performance of low temperature waste heat driven multi-bed silica gel/water adsorption chiller and showed the system can work even if the heat source temperature is 60 °C. Saha *et al.* (2007) presented the transient modelling for a two-bed, activated carbon fiber (ACF)/ethanol adsorption chiller using heat sources of temperature between 60 and 95 °C along with a coolant at 30°C. Miyazaki *et al.* (2010) evaluated the performance of innovative dual evaporator type three-bed adsorption chiller for cooling application and found significant improvement of system performance over two-bed single stage chiller. Uddin *et al.* (2013) analytically investigated the performance of an ideal adsorption-compression hybrid system and discussed the energy saving potential for the proposed system comparing the conventional system under same operating condition.

The key information to design the thermally driven system is the adsorption characteristics of adsorbent/refrigerant pair which is measured experimentally. Chua *et al.* (2002) measured the adsorption characteristic of silica gel/water system at temperature range from 25 to 65 °C and pressure 0.5 to 7 kPa using volumetric technique. Esteves *et al.* (2008) measured adsorption isotherm of natural gas and biogas components on activated carbon using both open- and closed-loop gravimetry over the pressure and temperature ranges of 0-9 MPa and 0-52 °C, respectively. El-Sharkawy *et al.* (2006) measured the adsorption characteristics of activated carbon fibers/ethanol pair at evaporation temperature 10 and 15°C with varying adsorption temperatures from 11 to 60 °C. Saha *et al.* (2009) measured the adsorption characteristics and heat of adsorption of activated carbon/R-134a pair at temperature range of 5-70 °C and pressure up to 1.2MPa using volumetric method. El-Sharkawy *et al.* (2008) also investigated the adsorption equilibrium of activated carbon/ethanol pair for solar powered adsorption cooling applications using thermogravimetric analyzer. However, it is proven that gravimetric method employing magnetic suspension balance provides high accuracy of adsorption equilibrium and kinetic data.

Water as refrigerant is commonly used effectively in the air-conditioning applications. However, for refrigeration applications that require relatively lower temperatures, methanol has been used before but reservations exist on its toxicity. On the other hand, the utilization of ethanol in adsorption cooling systems has couple of advantages, such as, ethanol is environmental friendly, non-toxic and it has a relatively high vapor pressure even at low temperature levels and its low freezing temperature (Cui *et al.*, 2005).

In the present study, adsorption equilibrium of highly porous Maxsorb III/ethanol, H₂ treated Maxsorb III/ethanol and KOH-H₂ treated Maxsorb III/ethanol pairs are measured experimentally using magnetic suspension adsorption measurement unit at evaporation temperature ranges from -6 to 65 °C and adsorption temperature 30 to 70 °C for a possible use in adsorption refrigeration system application. The isotherms data are correlated using the D-R and D-A equations from which P-T-W diagrams are plotted. The performance of a time-independent adsorption refrigeration cycle has been investigated and compared with other adsorbent refrigerant pairs that available on the open literatures.

2. EXPERIMENT

2.1 Materials

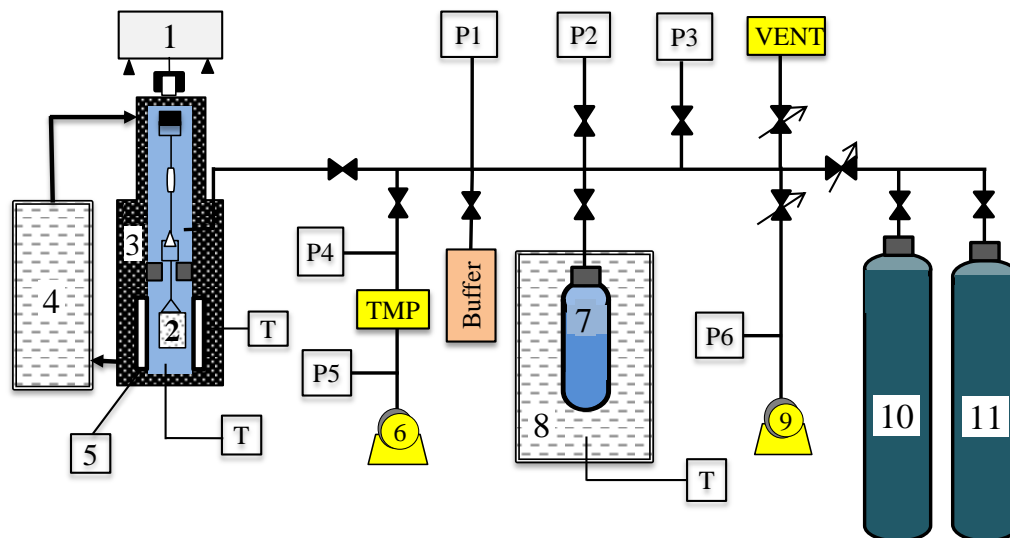
Activated carbon powder, namely Maxsorb III, with surface area is $3045 \text{ m}^2/\text{g}$ and micropore volume is $1.7 \text{ cm}^3/\text{g}$, is treated with H_2 and KOH with maintaining pore structure of the treated sample as similar as possible. H_2 treated Maxsorb III is prepared by placing Maxsorb III in a reduction environmental condition ($\text{Ar}/\text{H}_2=8/2$ (v/v)) at 600°C for 24 h. After that, potassium hydroxide (KOH) treatment was applied to H_2 treated Maxsorb III at different weight ratios to prepare the $\text{KOH}-\text{H}_2$ treated Maxsorb III and the mixture is heat treated at $600-750^\circ\text{C}$ for 1h under nitrogen flow. Finally the mixture is washed with HCl to adjust pH about 7. The aim of surface treatment is to clarify the influence of the surface functionalities on adsorption behavior of ethanol molecules in carbon micropores. Details of treatment can be found elsewhere (Kil *et al.*, 2013). The elemental composition of the sample is summarized in Table1. The refrigerant has been used in the present study is ethanol with purity 99.5%. Thermophysical properties of ethanol were evaluated using Refprop version 9 (Lemmon *et al.*, 2010).

Table 1: Thermophysical properties of activated carbon based adsorbents.

Adsorbents	Elemental compositions					Porosity		
	C [%]	H [%]	N [%]	O [%]	Ash [%]	Surface area [m^2/g]	Micropore volume [cm^3/g]	Pore width [nm]
Maxsorb III	95.13	0.14	0.25	4.35	0.13	3045	1.70	1.11
H_2 treated Maxsorb III	97.91	0.22	0.12	1.75	-	3029	1.73	1.15
$\text{KOH}-\text{H}_2$ treated Maxsorb III	89.15	0.27	0.08	10.46	-	2992	1.65	1.11

2.2 Experimental procedure

A magnetic suspension adsorption measurement unit is used to measure the adsorption characteristics of ethanol onto surface treated activated carbons. Using the multi-step technique the experiment was conducted within evaporation temperature between -14 and 65°C and adsorption temperature 20 and 70°C . In each step adsorption temperature was kept constant and evaporation temperature increased step by step until reaching relative pressure of about 0.9. The sorbent was first heated at 120°C for 4 hours under vacuum condition of $3 \times 10^{-4} \text{ Pa}$ to remove any adsorbed gas inside the sample. As the weight measurement method is influenced by the buoyancy force so the effect of buoyancy is considered during measurements inserting inert gas. The schematic diagram of the experimental apparatus is shown in Figure1.



1. Magnetic suspension balance unit, 2. Sample cell, 3. Circulation oil jacket, 4, 8. Isothermal oil bath, 5. Sheathed heater, 6, 9. Rotary and Diaphragm pump, respectively, 7. Refrigerant pool, 10. Nitrogen, 11. Helium, TMP- Terbo-molecular pump, T- Thermocouple. P- Pressure gauge.

Figure 1: Schematic diagram of the experimental set up.

An isothermal air bath is used to avoid the condensation inside the connecting tube. A detailed description of the experimental procedure can be found elsewhere (Uddin *et al.* 2014).

3. RESULTS AND DISCUSSION

3.1 Adsorption Isotherm

The Dubinin - Radushkevich (D-R) and Dubinin - Astakhov (D-A) equations are widely used to fit the equilibrium uptake data of vapors onto carbon based adsorbents. Equation (1) and Equation (2) represents the D-R and D-A equations, respectively.

$$W = W_0 \exp \left[- \left\{ \frac{A}{E} \right\}^2 \right] \quad (1)$$

$$W = W_0 \exp \left[- \left\{ \frac{A}{E} \right\}^n \right] \quad (2)$$

Where A is the adsorption potential that can be estimated as in Equation (3);

$$A = R_g T_{ad} \ln \left(\frac{P_{ad}}{P_{eva}} \right) \quad (3)$$

Here W stands for the equilibrium uptake (kg/kg) for the adsorbent/refrigerant pair, W_0 defines the maximum adsorption capacity (kg/kg). E is the adsorption characteristic parameter (kJ/kg), T_{ad} is the adsorption temperature (K), P_{ad} defines the saturation pressure (kPa) of refrigerant at adsorption temperature and P_{eva} is the equilibrium pressure (kPa). The exponential parameter n gives the best fitting of $\ln(W)$ versus A^n plot. The numerical values of fitting parameters using the above correlations are furnished in Table 2.

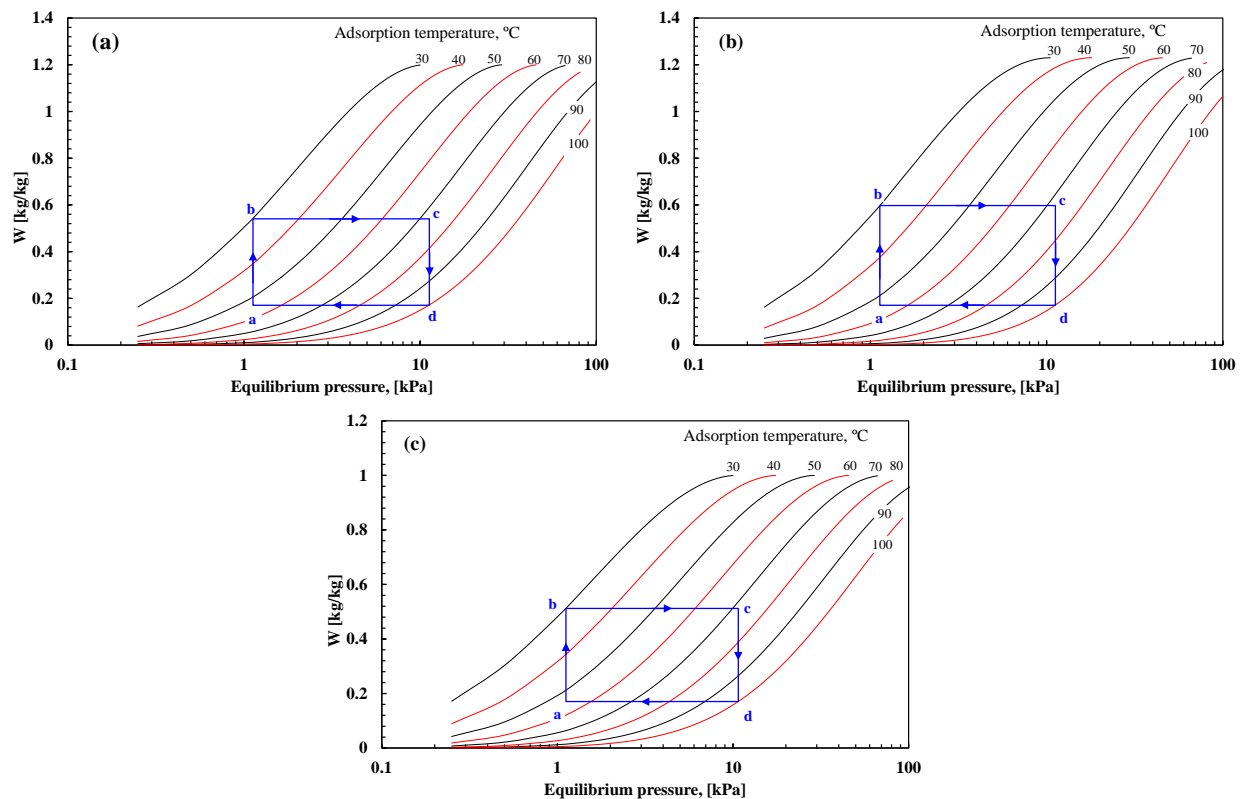


Figure 2: Adsorption isotherms of (a) Maxsorb III/ethanol, (b) H₂ treated Maxsorb III/ethanol and (c) KOH-H₂ treated Maxsorb III/ethanol pair as predicted by equilibrium isotherm equation.

It is found that the maximum adsorption uptake for H₂ treated Maxsorb III is higher than the other two studied adsorbents (Table 2), which follows the order of surface area and micro pore volume of the sample (Table 1). Employing the isotherm model with the numerical values from Table 2, the adsorption isotherms of Maxsorb III/ethanol, H₂ treated Maxsorb III and KOH-H₂ treated Maxsorb III/ethanol pairs are predicted and presented in Figures 2(a)-2(c), respectively. Ideal adsorption refrigeration cycles working at evaporation temperature -5°C, adsorption temperature 30°C and desorption temperature 100°C have been superimposed on these figures.

Table 2: Isotherm fitting parameters of assorted adsorbent/refrigerant pairs

Adsorbents	W_0 [kg/kg]	n [-]	E [kJ/kg]
Maxsorb III	1.20	1.8	139
H ₂ treated Maxsorb III	1.23	2.0	138
KOH-H ₂ treated Maxsorb III	1.00	1.9	152

The isosteric heat of adsorption is estimated using Clausius–Clapeyron equation. The average values of isosteric heat of adsorption are found to be 1032, 1034, 1035 kJ/kg for Maxsorb III/ethanol, H₂ treated Maxsorb III/ethanol and KOH-H₂ treated Maxsorb III/ethanol pairs, respectively.

3.2 Thermodynamic cycle

Figure 3 shows the schematic diagrams of a basic adsorption system. The system is mainly consists of an evaporator, a condenser and an adsorber/desorber heat exchangers (sorption element). The condenser and the evaporator are connected through a throttling valve or a bending capillary tube for maintaining the pressure difference. Here the basic cycle is assumed to be an ideal and as consequence, the pressure drops as well as the heat loss are neglected. The P-T-W diagram shows the relationship between the equilibrium pressure, adsorbent temperature and adsorption capacity at equilibrium state conditions which makes it possible to estimate the performance of adsorption cycle. Figures 4(a)-4(c) show the P-T-W diagrams of Maxsorb III/ethanol, H₂ treated Maxsorb III/ethanol and KOH-H₂ treated Maxsorb III/ethanol pairs, respectively. The ideal cycle for the same adsorbent-refrigerant pair is superimposed therein. Each cycle consists of two isosteric and two isobaric processes.

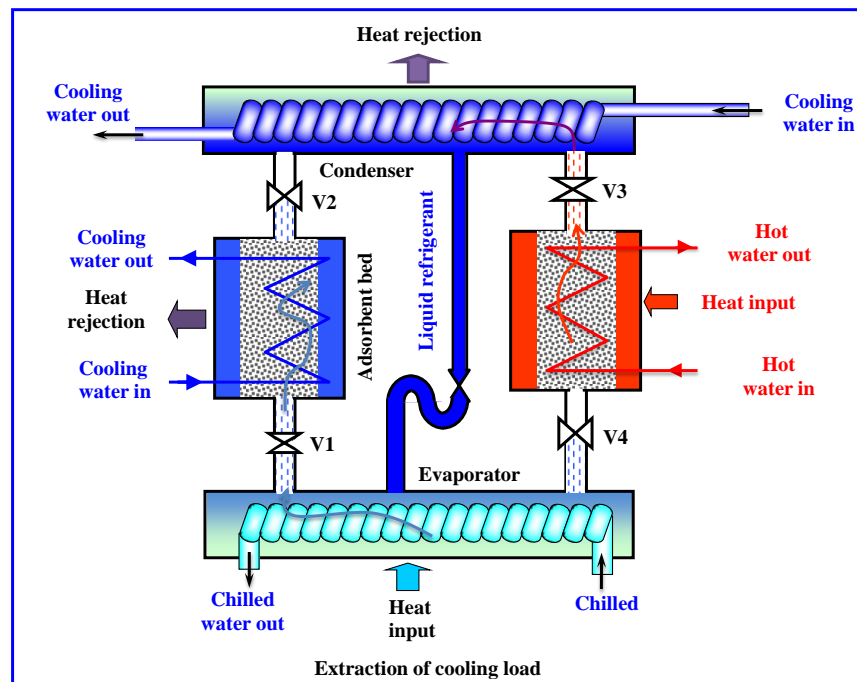


Figure 3: Schematic diagram of ideal adsorption cooling system.

In adsorption process (a→b), the pressure is kept constant at P_{eva} . The refrigerant vapor evaporates in the evaporator picking up its latent heat from the chilled water then, adsorbed by the adsorbent packed in the adsorber via the valve V1 (see Figure 3). The refrigerant concentration in the adsorber increases from W_{min} to W_{max} . In pre-heating (b→c) process, the sorption element is isolated and heated at constant concentration using a high temperature heat source and hence the pressure increases from P_{eva} to P_{con} . In desorption process (c→d), the refrigerant regenerates and goes to the condenser via the valve V3 at pressure P_{con} . The refrigerant concentration on the sorption element decreases from W_{max} to W_{min} . In the pre-cooling process (d→a), the adsorbent bed is cooled at constant concentration which makes the pressure decrease from P_{con} to P_{eva} .

It can also be seen from Figures 4(a)-4(c) that the concentration difference for H₂ treated Maxsorb III/ethanol pair is about 10% higher than Maxsorb III/ethanol pair and 19% higher than that of KOH-H₂ treated Maxsorb III/ethanol pair at the same operating condition, demonstrating the superiority in ice making applications (see Table 3). In addition to uptake difference, the adsorption kinetics of assorted working pair is another essential parameter that needed to be considered during making a dynamic simulation of adsorption system. The adsorption kinetics data for the assorted adsorbent/refrigerant pairs can be found elsewhere (El-Sharkawy *et al.*, 2014).

The model described here is a thermodynamically equilibrium model. This means, all the thermal contributions are calculated based on heat and mass balance provided by the (P-T-W) diagrams.

Cooling effect can be estimated as given in Equation (4),

$$Q_C = M_s (W_{max} - W_{min}) \left[\Delta h_{eva} - \int_{T_{eva}}^{T_{ad}} C_{p,ref} dT \right] \quad (4)$$

Where M_s is the mass of the sample adsorbent, W_{max} and W_{min} is the maximum and minimum sorption uptake, respectively. Δh_{eva} is the vaporization enthalpy and $C_{p,ref}$ is specific heat of refrigerant.

Desorption heat can be calculated as given in Equation (5),

$$Q_{des} = M_s Q_{st} (W_{max} - W_{min}) \quad (5)$$

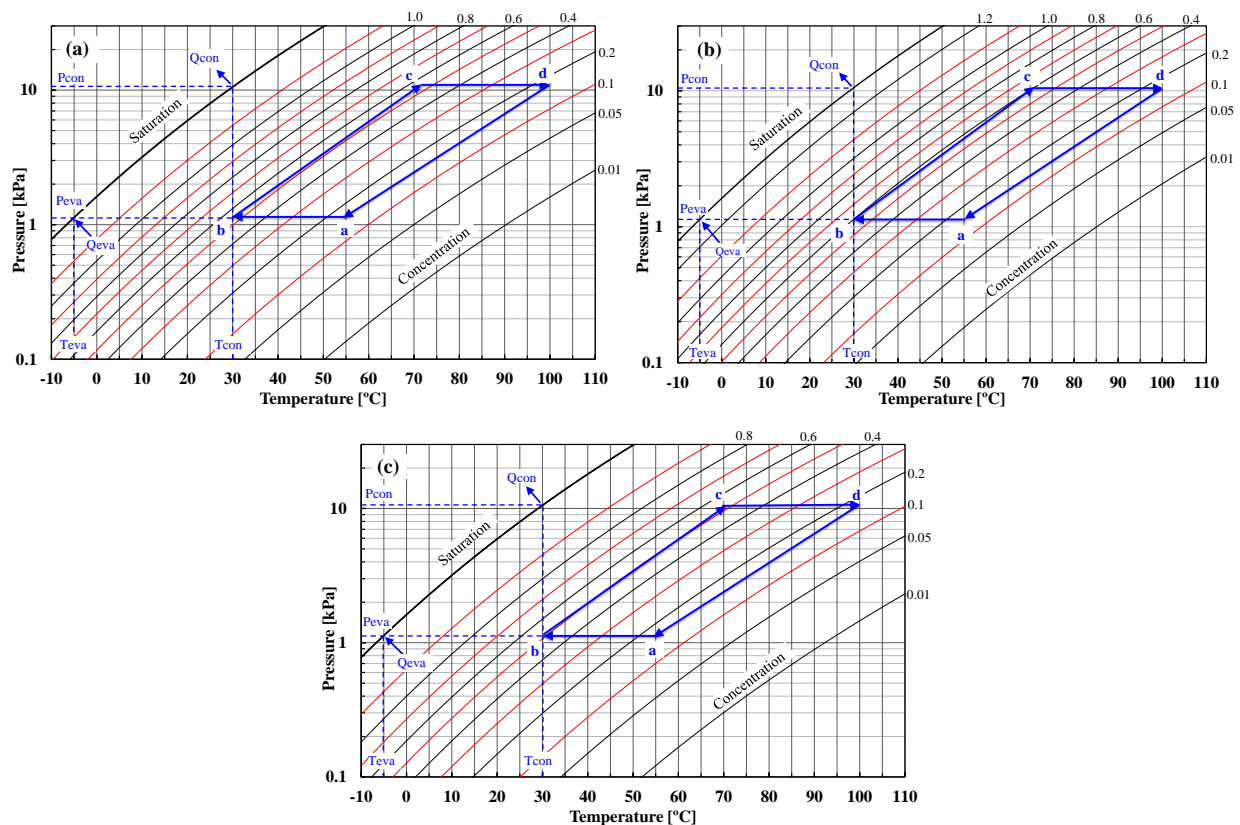


Figure 4: P-T-W diagram of (a) Maxsorb III/ethanol, (b) H₂ treated Maxsorb III/ethanol and (c) KOH-H₂ treated Maxsorb III/ethanol pairs.

$$W_{max} = W_0 \exp \left[- \left\{ \frac{RT_{ad}}{E} \ln \left(\frac{P_{ad}}{P_{eva}} \right) \right\}^n \right] \quad (6)$$

$$W_{min} = W_0 \exp \left[- \left\{ \frac{RT_{des}}{E} \ln \left(\frac{P_{des}}{P_{con}} \right) \right\}^n \right] \quad (7)$$

Total sensible heat is the sum of sensible heat during pre-heating and desorption shown in Equation (8)

$$Q_{sh} = Q_{sh,pr} + Q_{sh,des} \quad (8)$$

Sensible heat during pre-heating can be estimated as in Equation (9),

$$Q_{sh,pr} = M_s \int_{T_b}^{T_c} C_{p,s} dT + M_s W_{max} \int_{T_b}^{T_c} C_{p,ref} dT + M_{bed} \int_{T_b}^{T_c} C_{p,bed} dT \quad (9)$$

Sensible heat during desorption can be estimated as in Equation (10),

$$Q_{sh,des} = M_s \int_{T_c}^{T_d} C_{p,s} dT + M_s \frac{W_{max} + W_{min}}{2} \int_{T_c}^{T_d} C_{p,ref} dT + M_{bed} \int_{T_c}^{T_d} C_{p,bed} dT \quad (10)$$

For simplicity, the thermal capacity of the adsorption bed is considered two times that of thermal capacity of the adsorbent. The specific cooling effect (*SCE*) and the coefficient of performance (*COP*) of the time independent ideal adsorption cycle is calculated using the following Equations (11) and (12), respectively:

$$SCE = (W_{max} - W_{min}) \left[\Delta h_{eva} - \int_{T_{eva}}^{T_{con}} C_{p,ref} dT \right] \quad (11)$$

$$COP = M_s \frac{SCE}{Q_{des} + Q_{sh}} \quad (12)$$

Figure 5(a) Shows the variation of *SCE* with desorption temperature for five different adsorbents/ethanol pairs at evaporation temperature -5°C and adsorption temperature 30°C . The value of *SCE* increases linearly with the increase of desorption temperature. This is due to the concentration difference with increase of regeneration temperature at constant adsorption and evaporation temperature. It can be seen from the Figure 5(a) that the *SCE* values for H_2 treated Maxsorb III is higher than the other studied working pairs. For the sake of comparison, the performance of adsorption cooling cycles employing four assorted pairs are furnished in Table 3. It is found that, at regeneration temperature 100°C along with adsorption and evaporation temperatures of 30 and -5°C , respectively, the H_2 treated Maxsorb III/ethanol pair achieves an *SCE* as high as 374 [kJ/kg] whilst Maxsorb III/ethanol, KOH-H_2 treated Maxsorb III/ethanol and ACF A-20/ethanol pairs provide 90%, 81% and 64% of the *SCE* value of H_2 treated Maxsorb III/ethanol pair, respectively. This is because the H_2 treated Maxsorb III/ethanol pair possess highest concentration difference ($W_{max} - W_{min}$) at the same operating conditions (see Table 3).

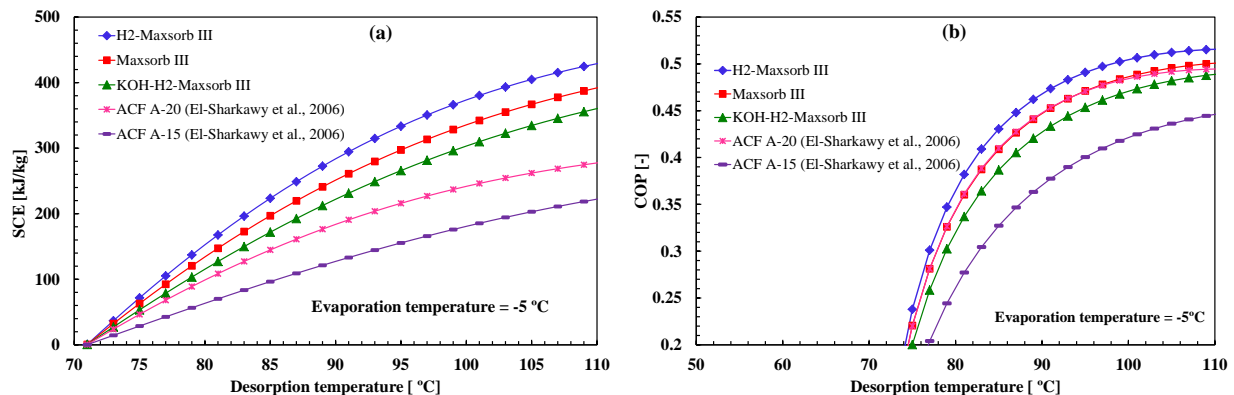


Figure 5: Effect of desorption temperature on (a) specific cooling effect (*SCE*) and (b) Coefficient of performance (*COP*).

Figure 5(b) shows the variation of coefficient of performance with desorption temperature at evaporation temperature $-5\text{ }^{\circ}\text{C}$. It can be noticed that the COP increases sharply only when the desorption temperature is below $100\text{ }^{\circ}\text{C}$. After that there is no significant change in COP even though SCE increases. This is happened due to the requirement of heat input becomes significantly large when the temperature difference between heat source and heat sink becomes higher than $100\text{ }^{\circ}\text{C}$. As for the ice making application where the evaporation temperature is kept constant at $-5\text{ }^{\circ}\text{C}$, the cycle should be driven by relatively higher regeneration temperature to produce sensible cooling production.

Table 3: Adsorption uptake and cycle performance of the assorted adsorbents using ethanol for ice making applications; adsorption and evaporation temperatures are 30 and -5°C , respectively.

Desorption temperature [$^{\circ}\text{C}$]		Maxsorb III	H_2 treated Maxsorb III	KOH-H_2 treated Maxsorb III	ACF A-20
90	W_{\max} [kg/kg]	0.543	0.561	0.516	0.363
	W_{\min} [kg/kg]	0.254	0.235	0.261	0.152
	SCE [kJ/kg]	251.2	283.7	222.0	183.7
	COP [-]	0.447	0.468	0.427	0.448
100	W_{\max} [kg/kg]	0.543	0.561	0.516	0.363
	W_{\min} [kg/kg]	0.157	0.132	0.168	0.085
	SCE [kJ/kg]	335.5	373.5	303.2	241.7
	COP [-]	0.486	0.505	0.471	0.484
110	W_{\max} [kg/kg]	0.543	0.561	0.516	0.363
	W_{\min} [kg/kg]	0.092	0.068	0.102	0.044
	SCE [kJ/kg]	391.9	428.9	360.4	277.5
	COP [-]	0.501	0.516	0.489	0.495

4. CONCLUSIONS

Adsorption equilibriums of H_2 treated Maxsorb III, Maxsorb III and KOH-H_2 treated Maxsorb III with ethanol have been measured experimentally using a magnetic suspension adsorption measurement unit. Experimental results show that, the maximum adsorption capacity of H_2 treated Maxsorb III/ethanol pair is about 1.23 kg/kg whilst the Maxsorb III and KOH-H_2 treated Maxsorb III/ethanol show maximum adsorption capacities of 1.2 and 1.0 kg/kg , respectively. The Dubinin-Radushkevich and Dubinin-Astakhov equations are used to correlate the equilibrium uptake data and to plot P-T-W diagrams of assorted pairs. Thermodynamic analysis shows that the H_2 treated Maxsorb III/ethanol adsorption cycle can achieve a COP as high as 0.51 with an evaporation temperature -5°C , regeneration temperature $100\text{ }^{\circ}\text{C}$ along with a coolant at $30\text{ }^{\circ}\text{C}$ whereas the KOH-H_2 treated Maxsorb III/ethanol pair can achieve COP 0.47 . For the sake of comparison the change of SCE and COP with desorption temperatures for the five pairs have been studied. It is also found that SCE of ideal cycle using H_2 treated Maxsorb III/ethanol pair is superior to that of other studied adsorbents/ethanol pair.

NOMENCLATURE

A	adsorption potential [kJ/kg]	Q_{des}	desorption Heat [J/kg]
COP	coefficient of performance	Q_{st}	isosteric heat [J/kg]
$C_{p,\text{ref}}$	specific heat of refrigerant [J/kg K]	Q_{sh}	sensible heat [J/kg]
$C_{p,s}$	specific heat of adsorbent [J/kg K]	$Q_{\text{sh,pr}}$	sensible hat during pre-heating [J/kg]
$C_{p,\text{bed}}$	specific heat of bed [J/kg K]	$Q_{\text{sh,des}}$	sensible heat during desorption [J/kg]
E	adsorption characteristic parameter [kJ/kg]	R_g	gas constant [kJ/kg K]
Δh_{eva}	evaporation heat [J/kg]	SCE	specific cooling effect [J/kg]
M_{bed}	mass of bed [kg]	T_{ad}	adsorption temperature [K]
M_s	mass of adsorbent [kg]	T_{des}	desorption temperature [K]
P_{ad}	saturation pressure of refrigerant at adsorption temperature [kPa]	W	equilibrium uptake [kg/kg]
P_{eva}	equilibrium pressure of refrigerant [kPa]	W_0	maximum adsorption capacity [kg/kg]
P_{con}	condensation pressure [kPa]	W_{\max}	maximum uptake [kg/kg]
P_{des}	desorption pressure [kPa]	W_{\min}	minimum uptake [kg/kg]

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