An investigation of the ammonia adsorption performance on different adsorbents for cooling applications

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Ammonia (NH_3) is a common gas and has been widely investigated on adsorption for numerous purposes. Ammonia is commonly employed as a refrigerant in the refrigeration systems to obtain temperatures below 263 K. Adsorption systems powered by low-temperature heat sources that utilize ammonia as working fluid work at positive pressures, therefore, their manufacturing and usage are much easier, and they require less maintenance, comparing with vacuum systems. In this study, ammonia adsorption performance of various adsorbents is investigated numerically for cooling applications. The adsorbents considered in this study are zeolite, alumina, silica gel and activated carbon. The adsorption performance of each adsorbent-adsorbate pair under various operating conditions is compared. Among the adsorbate-adsorbent pairs investigated in this study, ammonia and activated carbon gave the best performance values in all cases. According to the results of the study, most important parameters that effects COP, Q_r and SCP of an adsorption cooling system are the adsorbent type, cooling source temperature, heating source temperature and evaporation temperature at the evaporator, respectively. It is shown that increasing evaporator temperature and heating source temperature results an increase in COP, Q_r and SCP values of the adsorption system, whereas, for the case of cooling source temperature, highest values of COP, SCP and Q_r are obtained at the lowest cooling fluid inlet temperature.

Keywords: Ammonia, adsorption cooling, adsorbate-adsorbent pair, positive pressure, COP, SCP

INTRODUCTION

Global energy demand is growing rapidly. Cooling and refrigeration demand constitutes a large part of global energy consumption. For this reason, especially during the summer season, a rapid increase can be observed in the amount of electricity consumption. Thermally driven cooling technologies such as absorption and adsorption refrigeration technologies can serve as an alternative to conventional refrigeration cycles [1]. Adsorption refrigeration systems can be considered as environmentally friendly since they require low-grade heat sources such as solar energy or waste heat [2]. In comparison with absorption cooling systems, an adsorption system can be driven by heat sources in a large temperature range, while absorption systems require heat sources with higher temperatures [3],[4]. And last but not least, an adsorption system can be designed and manufactured simply when compared with an absorption system [5]. In addition to their numerous advantages, adsorption cooling systems also have some drawbacks including (a) lower COP when compared with absorption technology and conventional cooling cycles, (b) they have relatively high initial investment costs, as a result, they cannot compete with other cooling technologies

commercially [6]. Many researchers have conducted works on developing adsorption cooling technology in order to overcome the drawbacks that restrict them from being competitive in the related market [7].

Adsorption is a reversible physical phenomenon, realised with an interaction between adsorbent (solid) surfaces and an adsorbate (fluid vapour), driven by cohesive forces [7]. The physical adsorption process of the gas occurs mainly within the pores and surface of the solid adsorbent [8]. The adsorbed amount and concentration of refrigerant in the pores are strongly dependent on pressure and temperature variations as well as the operating conditions of the system [9]. The design of an adsorption refrigeration system requires the knowledge of adsorption characteristics of the employed adsorbent-adsorbate pair when the temperature and pressure are varying [10].

The isosteric heat of adsorption is a combined specific property of an adsorbent/adsorbate pair. The equilibrium adsorption properties at several adsorbent temperatures and adsorption chamber adsorbate pressures were studied for a wide range of pairs by several researchers [5-11].

The adsorption characteristics of every adsorbent-adsorbate pair are usually quite different than each other. Detailed specification about the adsorption isotherms of the assorted

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adsorbent/adsorbate pair as well as the isosteric heat of adsorption must be known in order to design an adsorption based cooling system. Detailed literature reviews on adsorption working pairs for refrigeration/cooling applications can be found in the references [1, 3, 4, 5, 14].

Basic adsorption cooling system consists of one adsorption bed, expansion valve, evaporator and condenser. Impermanent cooling production nature of a one-bed system is not convenient as cooling cannot be produced when the bed is undergoing desorption [12]. Multiple bed systems which have at least two adsorber beds are more convenient and practical to use. Using more beds increase COP values, however, the system becomes more complex, as a result of adding more installation elements such as pipes and valves [13].

Adsorption pair which is a vital part of the adsorption refrigeration cycle consists of adsorbent and refrigerant [14]. Refrigerant or adsorbate should have the following properties: [1, 2, 10, 14].

• Small molecular size to allow it to be adsorbed by the adsorbent.

• Non-toxic, non-corrosive and non-flammable.

• Large latent heat per volume

• Good thermal stability

Unfortunately, none of the fluids fulfils completely these requirements. Among the fluids appropriate to use in the adsorption systems, ammonia, methanol and water all have relatively high latent heats, however water cannot be used for freezing purposes as its freezing temperature is 273.15 K and methanol is flammable [1, 14]. Although, ammonia is toxic, it has advantages to other two as it can be used at positive pressures. In addition to that, it has no effect to ozone layer as well as no global warming potential. This makes ammonia a suitable and favoured adsorbate for adsorption cooling cycles.

Ammonia is a colourless, diffusive gas at atmospheric conditions. The ammonia molecule is basic and polar in nature, as a result, several types of adsorbents can be utilized to examine their ammonia adsorption efficiencies, including zeolites, alumina, activated carbon, silica [15]. Ammonia has a relatively high latent heat, about 1365 kJ/kg at 243 K.

In order to choose an appropriate adsorbent some of the important considerations are: [10, 14]

• Low cost, widely available

• Non-toxic and non- corrosive.

• Desorption of most of the adsorbate when exposed to thermal energy

• The ability to change capacity with the variation of temperature

• Excellent compatibility with the refrigerant.

Since adsorbent used for adsorption system affects the system performance significantly, it is very important to understand the characteristics of an adsorbent. Silica gel, zeolite, alumina and activated carbon are the most common adsorbents that are used in adsorption cooling cycles. These are appropriate to the adsorption of the ammonia vapour.

Silica gel is a natural mineral purified and processed into the granular or beaded, vitreous, porous form of silicon dioxide (SiO_2) produced synthetically from silicate and sulfuric acid [5]. Silica gel can be considered as one of the widely used low temperature working adsorbent, which can be employed in the cooling systems driven by the relatively low-temperature heat sources in which the temperature is lower than 373 K [10, 14,15].

Zeolite is a type of alumina silicate crystal composed of alkali or alkali soil [4]. Due to their low cost and availability, zeolites gained a significant interest as an adsorbent. Natural zeolites also gained a remarkable interest due to their valuable properties such as ion-exchange capability [1]. About 150 types of zeolites can be artificially synthesized, and they are named by one letter or a group of letters, such as type A, type X, type Y, type ZSM, etc. [1, 4].

Activated alumina is aluminium oxide in the form of very porous spheres. Since it is a highly porous material, it is a great adsorbent for adsorption cycles.

Activated carbons are made by pyrolizing and carbonising source materials, such as coal, lignite, wood, nut shells and synthetic polymers, at high temperatures (1000 to 1100 K) [1, 4, 13, 14]. Activated carbons can be found in many different forms such as powders, granulated, microporous, molecular sieves and carbon fibres. It is widely used for adsorption applications.

Due to its zero global warming and ozone depletion potential, ammonia has been considered as being one of the most efficient refrigerants. However, there are not enough studies in the literature concerning adsorption of ammonia. A number of studies such as refs. [13-17] have been performed on the adsorption cooling systems employing the pair of activated carbon and ammonia. Only a few study considered silica gel-ammonia pair as an adsorbent-adsorbate pair [16, 17]. On the other hand, to the best of our knowledge, there is not any published research subject to a comparative evaluation of the adsorption cooling system with ammonia as adsorbate fluid onto the

adsorbent solid including the silica gel, zeolite, alumina and activated carbon.

Since conducting experimental investigation of adsorption systems is complicated and expensive, it is much easier and convenient to simulate the process numerically. As a result, in this study, a mathematical modelling method for adsorption cooling systems proposed by Kilic [18] is used, validated and applied for ammonia and four different adsorbent pairs. With the used mathematical method, adsorption cooling cycle characteristics of any adsorbent-refrigerant pair can be easily estimated.

In this study, silica gel, zeolite, alumina and activated carbon are chosen as adsorbents and ammonia is considered as the adsorbate. Then, the performance of these four different adsorbent-adsorbate pairs for an adsorption refrigeration / cooling application with different working conditions are investigated and compared with each other. Main contributions of the present study into the current literature can be explained with two-fold as: firstly, it presents a more realistic modelling approach by the use of real fluid properties in the calculations, and secondly, four different adsorbents are used for the adsorption cooling and refrigeration system with the ammonia employed as working fluid. The presented results can be used on the design of an adsorption based cooling cycle systems in which ammonia is employed as adsorbate with the one of the adsorbents among silica gel, zeolite, alumina and activated carbon.

MATHEMATICAL MODELLING

The adsorption cooling systems are similar to the known mechanical vapour compression systems and the system components such as the evaporator, condenser and expansion valve are the same. The main difference is that the thermal compressor takes the place of the mechanical compressor. Therefore, an adsorption cooling/refrigeration system can be driven only by heat energy so it does not need electrical energy to perform. Details of the working principle of the thermal compressor operating according to the adsorption refrigeration cycle are given in the other study by Kilic [18]. In that study, the mathematical model of the two-bed adsorption cooling system and the calculation procedure were given in details. Therefore, the important equations and parameters related to the present work is introduced in this section.

Adsorption isotherms

It has become a common approach that the use of Dubinin–Astakhov (D–A) model for the calculation of the adsorbate uptake value on the adsorbent as a function of temperature and pressure in the adsorption bed [19]. Dubinin–Astakhov (D–A) equation may be written in the form given as follow:

$$W = W_0 \, exp \left\{ - \left[\frac{RT}{E} \, ln \left(\frac{p_s}{p} \right) \right]^n \right\} \qquad (1)$$

with

$$W = X v_a \quad \text{and} \quad W_0 = X_0 v_0 \qquad (2)$$

In Eq.(1), *E* may be expressed as the specific characteristic energy of the assorted adsorbent-adsorbate pair which may be evaluated from the experimental measurements. *R* is the gas constant. Pressure is represented by p, and suffix s is referring to the saturation state. Temperature is represented by *T*. The parameter n is a power constant which results the best fitting of the experimental isotherms. The quantity *X* represents the specific adsorbed mass of adsorbate (kg of adsorbate per kg of adsorbent), and v_a is the specific volume of the adsorbed phase, which is given by

$$v_a = v_b \, exp \left(\Omega(T - T_b) \right) \qquad (3)$$

where v_b is the saturated liquid specific volume at the normal boiling point, *b* represents the van der Waals volume, and Ω is given as:

$$\Omega = \ln(b/v_b) / \ (T_c - T_b) \qquad (4)$$

T is the temperature. The critical and normal boiling point temperatures of the refrigerant are represented by suffixes c and b, respectively. The parameter v_0 can be obtained by using Eq.(2) at T = 273.15 K. Table 1 shows the properties and parameters of the adsorbates used in the present study.

Table 1. Properties and parameters of the adsorbents used in this study [16, 17]

	Unit	Zeolite13X	Alumina	Silicagel	AC-LM127
ρ	kg/m³	647	735	445	750
c_p	kJ/kgK	0.96	0.880	0.920	0.900
*ΔH_{ads}	kJ/kg	1901	1741	1731	1676
W_0	m³/kg	0.000399	0.000159	0.000235	0.000577
X_0	kg/kg	0.251	0.100	0.148	0.363
E	kJ/kg	804.0	587.2	558.9	487.1
n		0.98	0.844	1.04	0.95
E_a	kJ/kg	2466	2466	2466	2466

Calculated values at T=303.15 K in the present study.

Adsorption kinetics

Adsorption and desorption rates of the adsorbate onto adsorbent can be calculated by the use of Linear Driving Force (LDF) approach. Adsorption kinetics (dX/dt) equation is given in Eq.(5).

$$\frac{dX}{dt} = F_o \frac{D_o}{r_p^2} exp\left(-\frac{E_a}{RT}\right)(X_{eq} - X) \tag{5}$$

where X_{eq} is the equilibrium concentration at the given pressure and temperature; F_o is a constant characteristic of adsorbent's shape. The equilibrium concentration is calculated using the Dubinin-Astakhov equation given in Equation 1. X is the instantaneous concentration of the adsorbent bed. The values of the adsorption kinetic parameters are given in Table 1 and Table 2.

Adsorption heat

For adsorption of fluids below their thermodynamic critical point, its magnitude is larger than the heat of vaporization of the adsorbate, which has a strong temperature dependence. As a result, the difference between the adsorption heat and the vaporization heat is a property of relevance in the design of adsorption refrigeration systems. Moreover, the gas phase of the adsorbent is not ideal, during the adsorption of the adsorbate molecules onto the assorted adsorbent is affected by the pressure and temperature changes. Therefore, the heat of adsorption is calculated by using the following Eq.(6) as suggested by El-Sharkawy et al. [20].

$$\Delta H_{ads} = h_{fg} + (E)[ln(W_0/W)]^{1/n} + (ET\Omega/n)[ln(W_0/W)]^{((1-n))/n} \tag{6}$$

Table 2. Properties of the ammonia and the parameters used in the present study

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Properies	Values	Unit
MW	17.03	kg/kmol
T_b	239.8	K
T_c	405.4	K
R	0.4882	kJ/kg K
P_c	11333	kPa
ρ_c	225.00	kg/m^3
b	0.002183	m^3
v_b	0.001467	m^3/kg
v_0	0.001589	m^3/kg
Ω	0.002402	
*h_{fg}	1145	kJ/kg
D_o	2.54E-4	m^2/s
F_o	15	
R_p	1.7E-4	m
$M_{bm}c_{p,bm}$	14	kJ/K
M_s	10	kg

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*Values at T=303.15 K.

System performance parameters

Adsorption cooling system performance is commonly defined by its specific cooling power (SCP) in W/kg adsorbent and the coefficient of performance (COP). The two parameters are the most important data among the technical specifications of such products. COP and SCP are expressed by Eq. (7) and Eq. (8) respectively.

$$COP = \frac{Q_{eva}}{Q_{heat}} \tag{7}$$

$$SCP = \frac{Q_{eva}}{t_{cycle}M_s} \tag{8}$$

Where cooling capacity of the evaporator (Q_{eva}) and heat load of the system (Q_{heat}) in a cycle can be calculated by Eq.9 and Eq.10, respectively.

$$Q_{eva} = \oint_0^{t_{cycle}} \frac{dm_{eva,out}}{dt}\left(h_{exv,o} - h_{eva,sv}\right)dt \tag{9}$$

$$Q_{heat} = \oint_0^{t_{cycle}} \dot{Q}_{heat}dt \tag{10}$$

It should be mentioned that the electrical power consumption of chilled water, cold water and hot water pumps are relatively small so can be neglected.

Validation of the mathematical model

Several studies have been already reported about the adsorption cooling systems for the use of ammonia-activated carbon pair. Tamainot-Telto et al. [17] conducted a theoretical simulation work by the use of ammonia-activated carbon pair. In order to validate present mathematical model computations performed according to their parameters and working conditions used in their study. LM127 type activated carbon is chosen as the adsorbent. The mass of the metal bed material (M_{bm}) used in adsorption bed is ignored in this calculations. The cooling water inlet temperature ($T_{cool,in}$) to the bed and the condenser temperature (T_{cond}) is equal to 308.15 K. Two different evaporator temperature (T_{eva}) are considered as 268.15 K and 283.15 K. Fig.1 shows the comparisons between the present predicted results and the findings of Tamainot-Telto et al. [17] for the heating fluid inlet temperature in the range of 355.15 to 403.15 K.

It can be seen that the predicted COP values are mainly in a good agreement with the data of Tamainot-Telto et al. [17]. As the temperature increases from 355.15 to 403.15 K, there is a slight increase at COP values for both evaporator temperatures.
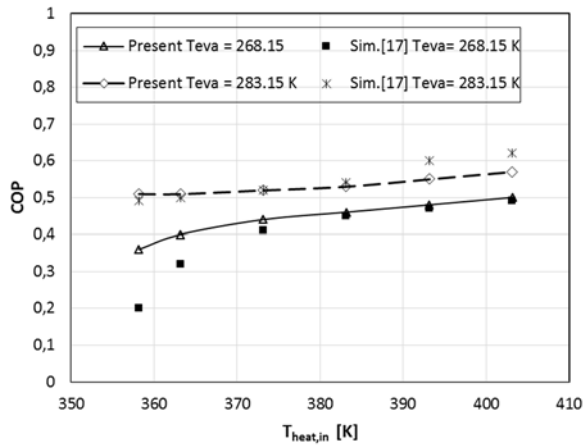
Fig.1. Comparisons of the predicted COP values of ammonia-activated carbon (LM127) pair with the simulation of Ref.[17] for the different heating fluid inlet temperatures.($T_{cool,in}$=T_{cond}=308.15K)

Increasing evaporator temperature considerably increases COP values in both sets of results. The maximum differences of the predicted COP values appear at lower heating temperature values for the case of low evaporator temperature. Although there is a very good agreement for the case of high evaporator temperature. This is probably resulted by the difference between their model and the present model. Main differences between the models are the form of D-A equation for the pair adsorption heat calculations and fluid properties calculations. In the present model, the properties of the fluid are obtained for real fluid properties, the adsorption heat is not constant. Even though there are differences between the models, the results show very good agreements. Hence, the mathematical model can be accepted as validated.

RESULTS AND DISCUSSION

The aim of the present study is to perform a comparative work on the adsorption cooling system using ammonia with different adsorbents. Four different widely used adsorbents (silica gel, zeolite (13X), alumina and activated carbon (LM127)) are chosen. Two-bed adsorption cooling system as described in previous sections are used in the calculations. The mass of the adsorbent (M_s) used in each bed is taken as 10 kg in the calculations. In the computations, adsorption/desorption time is taken as 600 seconds, precooling/preheating time is taken as 50 seconds. Therefore, the cycle time is taken as 1300 seconds. Computations are performed at the same working conditions for the four different adsorbent-adsorbate pairs. Properties and

parameters of the adsorbents and adsorbate used in this study are given in Tab.1 and Tab.2, respectively.

Table 3. Working conditions used in the calculations

Case	T_{eva} [K]	$T_{heat,in}$ [K]	$T_{cool,in}$ [K]	$T_{chill,in}$ [K]
1	268.15	**343.15 – 403.15**	303.15	287.15
2	**253.15-283.15**	363.15	303.15	287.15
3	268.15	363.15	**293.15-313.15**	287.15
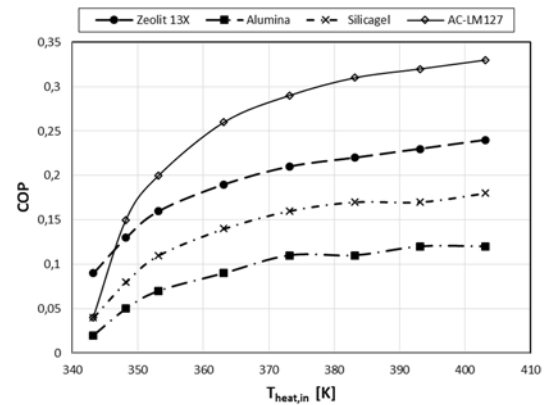
Fig.2. Comparisons of COP for the different heating and cooling fluid inlet temperatures. (T_{eva}=268.15 K; $T_{cool,in}$ = 303.15 K)
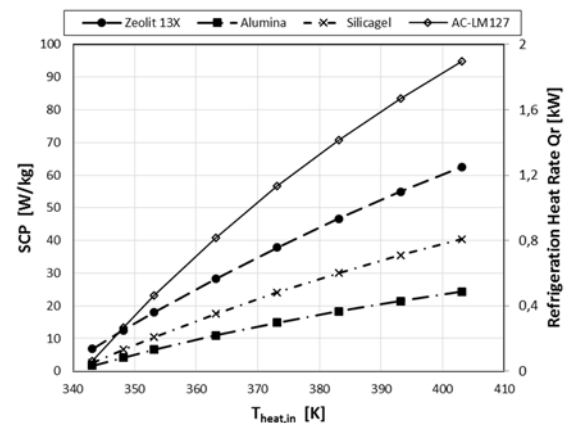
Fig.3. Comparisons of SCP and refrigeration heat rate for the different heating fluid inlet temperatures. (T_{eva}=268.15 K; $T_{cool,in}$ = 303.15 K)

Other temperature settings are applied as follow:
Condenser temperature $T_{cond} = T_{cool,in} + 3$ K ;
Evaporator temperature $T_{eva} = T_{chill,\,o} - 3$ K;
Bed maximum temperature $T_{bed,max} = T_{heat,in} - 2$ K ;
Bed minimum temperature $T_{bed,min} = T_{cool,in} + 2$ K .

Computations are performed for three different set of working conditions as given in Tab.3. Fig.2 shows the computed results of the coefficient of performance (COP) for Case 1. It can be seen that COP increases with rising heating source temperature for all the pairs. This may be explained

as when the heating temperature rises, more adsorbate circulating in the system with increasing desorption rate from the bed. Active carbon and alumina shows the best and worst COP values, respectively, among the pairs. For all of the pairs, there is a significant drop in the COP under the heating source temperature of 363 K.
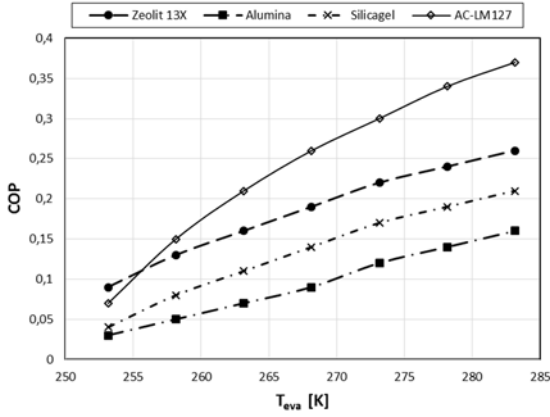

Fig.4. Comparisons of COP for the different evaporator temperatures.(T_{heat}=363 K; $T_{cool,in}$ =303 K)

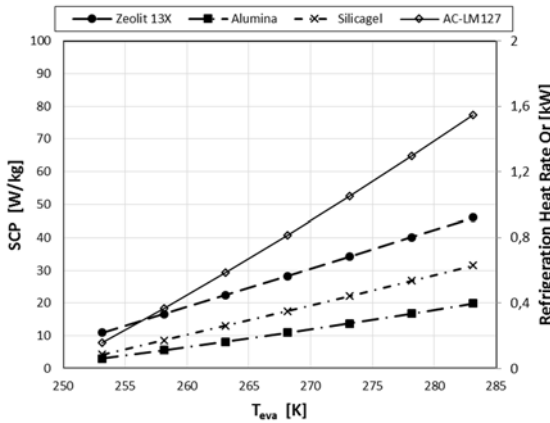**Fig.5.** Comparisons of SCP and refrigeration heat rate for the different evaporator temperatures. (T_{heat}=363 K; $T_{cool,in}$ = 303 K)

Fig.3 shows the computed results of the specific cooling power (SCP) and refrigeration heat rate (Q_r). For the all working fluids, SCP and Q_r values rise almost linearly with the heating temperature. Active carbon pair has the highest values of SCP and Q_r, 95 W/kg and 1.9 kW, respectively, at the heating temperature of 403.15 K. It may be explained as more fluid mass is desorbed from the bed with rising heating source temperature, due to this more working fluid circulates in the system, and the cooling power rises accordingly. Zeolite, silica gel and alumina pairs show less performance compared to the active carbon in descending order respectively.
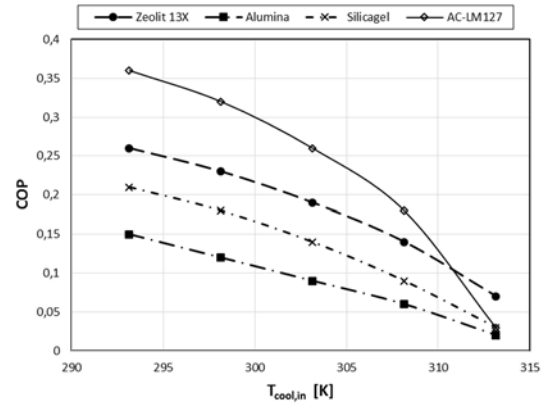

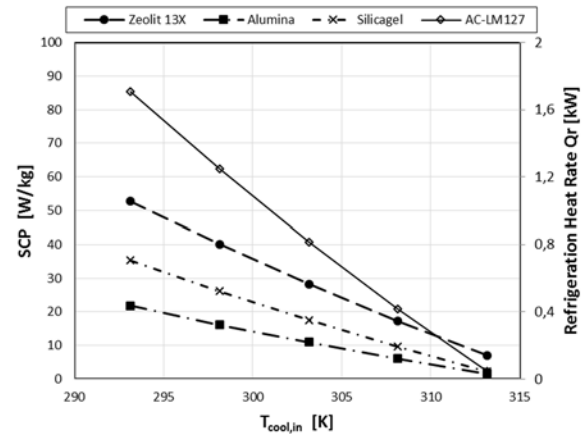**Fig.6.** Comparisons of COP for the different cooling fluid inlet temperatures.(T_{heat}=363 K; T_{eva} =268 K)

Fig.7. Comparisons of SCP and refrigeration heat rate for the different cooling fluid inlet temperatures. (T_{heat}=363 K; T_{eva} =268 K)

Comparing COP and SCP for the working fluids when the evaporator temperature changing as Case 2, it can be seen at Figs.(4, 5) that the highest COP (=0.38) and SCP (=79 W/kg) are obtained for the activated carbon at T_{eva} = 283.15 K, and the lowest ones are found for the alumina (as 0.03 and 3 W/kg, respectively) at T_{eva} = 253.15. Refrigeration heat rate also shows a similar trend, and its maximum values are 1.58, 0.92, 0.51 and 0.4 at T_{eva} = 283.15 K for active carbon, zeolite, silica gel and alumina, respectively. In general, increasing evaporator temperature result an increase in COP, SCP and Q_r values.

The results for Case 3 presented in Figs.(6, 7). The change of the cooling fluid inlet temperature on the performance parameters shows a reverse effect with the evaporator temperature. Highest values of COP, SCP and Q_r are obtained at the lowest cooling fluid inlet temperature, $T_{cool,in}$ = 293.15 K. This may be explained as decreasing cooling fluid temperature drops the minimum bed temperature and also

decrease condensation temperature and pressure at the condenser. Therefore, more adsorbate can be adsorbed at the bed with a lower adsorption temperature, and more adsorbate can be desorbed at the bed with lower condenser pressure. The performance parameters order is similar to the Case 1 and 2.

Considering the adsorption heat values (at 303.15K) given at Tab.1, zeolite 13X has the largest one (1901 kJ/kg), alumina and silica gel have almost same (1741 and 1731 kJ/kg, respectively), activated carbon has the smallest value (as 1676 kJ/kg).

CONCLUSIONS

This study presents a comparative study on a two bed adsorption cooling system with a working fluid ammonia as adsorbate and four different widely used adsorbents as zeolite 13X, alumina, silica gel and activated carbon (LM127). These four adsorbent-adsorbate pairs and various cycle operating conditions are investigated at the heating source temperature range from 343.15 to 403.15 K.

The effects of the different evaporator and cooling source temperatures are also evaluated and compared. The performance indicators of COP, SCP and refrigeration heat rate (Q_r) are investigated and compared for the same working conditions. Among the adsorbent-adsorbate pairs considered in this study, the best performance values are obtained for the activated carbon and ammonia pair. In the investigated range of working conditions, the maximum values of COP, SCP and Q_r for the pair with activated carbon are 0.38, 88 W/kg and 1.8 kW, respectively. The zeolite-ammonia pair shows similar behaviour to the activated carbon pair with about 28% and 34% less performance in terms of COP and SCP, respectively.

It is shown that the parameters that have the largest effect on COP and SCP are, in decreasing order, adsorbent type, cooling source temperature, heating source temperature and evaporation temperature at the evaporator. These results are valid for all working pairs. In general, increasing evaporation temperature and decreasing adsorption bed temperature during the adsorption process increases COP, SCP and Q_r for all working pairs.

It should be noted that the calculations are performed based on a basic adsorption cycle with a two-bed system. The selection of the adsorbent type has a great effect on the performance of the system. Hence, higher performances may be obtained with the selection of more efficient adsorbent for ammonia. Moreover, the system performances may be increased by the use of more advanced cycles in which new or advanced techniques, such as heat and mass recovery, applied.

NOMENCLATURE

b-   the van der Waals volume, m³;
c_p-   specific heat, kJ/kgK;
COP – coefficient of performance, - ;
D_0 –   surface diffusion coefficient, m²/s;
E –   characteristic energy, kJ/kg;
E_a –   activation energy, kJ/kg;
F_o -   a constant characteristic of adsorbent's shape.
h_{fg}-   vaporization enthalpy, kJ/kg;
ΔH_{ads}- adsorption heat, kJ/kg;
M -   mass, kg;
MW- molecular weight of fluid, kg/kmol;
n-   exponential constant, -.
p-   pressure, kPa;
p_s-   saturation pressure, kPa;
R –   gas constant, kJ/kgK;
SCP – specific cooling power, W/kg;
Q -   heat, kJ;
Q_r -   refrigeration heat rate, kW;
$\dot{Q}$ -   heat rate, kW;
t -   time, s;
t_{cycle}- total cycle time, s;
T -   temperature, K;
X –   adsorption capacity on mass basis, kg/kg;
v_a –   adsorbed phase specific volume, m³/kg;
v_b –   saturated liquid specific volume at normal boiling temperature, m³/kg;

Subscripts

a-   adsorbed phase;
ad-   adsorption;
b-   boiling point;
bed- adsorber bed;
bm- bed material;
c-   critical point;
cyc- cycle;
cond- condenser;
eva- evaporator;
f-   fluid;
fg-   phase change from liquid to gas;
in-   inlet;
out- outlet;
min- minimum;
max- maximum;
s-   sorbent;
s-   saturation;
l-   liquid phase;
v-   vapour phase.

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