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Adsorption Heat Pump Using an Innovative Coupling Refrigeration Cycle

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Abstract. An adsorption heat pump system using a new binary coupling adsorptive cycle is developed and tested. Experimental results show that the COP of the binary coupling cycle is higher than that of a pure zeolite-water system. The system operating pressure is found to be moderate (close to the ambient pressure) when a proper concentration of ammonia is used in the system. The moderate operating pressure of the new coupling cycle results in low leakage to the system, achieving long life and the light design of the system vessels. The use of the new binary adsorption cycle greatly improves the feasibility and reliability of the adsorption heat pumps, providing essential benefits for the industrialisation of adsorption heat pumps.

Keywords: adsorption heat pump, binary coupling, thermodynamics and operating pressure

1. Introduction

Recently, the possible applications of solid adsorption heat pumps have been greatly improved (Pons et al., 1999). Most efforts to develop solid adsorption heat pump technology for industrialisation and commercialisation had previously focused on single component adsorbate, the heat transfer enhancement in the adsorber, or cascading multi-adsorber cycles to fully utilise inner energy. Tamainot-Telto and Critoph (2001) made use of monolithic carbon for sorption refrigeration and heat pump applications. Tsilingiris (2000) showed the heat transfer analysis of low thermal conductivity solar energy absorbers. Sward et al. (2000) proposed an adsorption heat pump model for simulation of the thermal wave process with local equilibrium. The high conductivity of a polymer was utilised to coat the zeolite in order to improve the thermal conductivity of the fixed bed (regenerator) and decrease the thermal contact resistance between the adsorbent and adsorber surfaces (Hu et al., 1997; Zhu et al., 1998; Wang et al., 1999). Stitou et al. (2000) showed the results of development and comparison of advanced cascading cycles coupling a solid/gas thermo-chemical process and a liquid/gas absorption process. Qu (2001) described a study on heat and mass recovery in adsorption refrigeration cycles. Lai (2000) proposed an enhanced adsorption cycle operated by periodic reversal forced convection. Zhu (1998) found that the thermal decomposition of methanol (adsorbate) in solar-powered solid adsorption refrigeration systems results in the decrease of COP. For the single component adsorptive in the adsorption heat pump, the system pressure is too low (high vacuum) when some working media, such as water with zeolite, are used. It is difficult to keep the system operating in a high vacuum for a long time. On the other hand, the operating pressure is too high when other working media, such as NH_3 with zeolite, are used. This would result in the equipment being designed to be heavier and more costly. This paper presents the coupling cycle process of a solid adsorption with liquid absorption heat pump system (an ADAB system), which uses a binary adsorptive in the closed cycle, allowing the operating pressure to be near ambient. An intermittent binary adsorptive cycle system has been developed to improve the COP and the life of system. An experimental unit is built to test the performance of the ADAB system and compare its performance with that of pure adsorptive systems. Experiments are conducted to study the characteristics of the closed binary adsorption cycle and the effect of ammonia concentrations on the system operating pressure.

2. Thermodynamic Description of the Innovative Cycle

In an adsorption heat pump system using a single component adsorptive, the pressure of the system is controlled by the saturation pressures of the adsorbate at the evaporating and condensing temperatures respectively. The pressure of the system swings from the saturation pressure of the adsorbate evaporating temperature to the saturation pressure of adsorbate dew point (condensing) temperature. In the binary component cycle, the pressure can also be adjusted by the saturation pressure of the mixture vapour. A closed binary adsorption cycle is developed based on this principle. The operating pressure of the closed binary adsorption cycle can be adjusted by the concentration of the mixture.

A new coupling cycle system has been developed using the binary components (NH_3 and H_2O) as the working media. The system consists of Zeolite, NH_3 and H_2O as working media to form a closed coupling cycle process of a solid adsorption heat pump system supplemented with a liquid absorption process (ADAB system). The working media (NH_3 and H_2O) are a good pair of absorption refrigerants in absorption refrigeration (Fernández-Seara and Vázquez, 2001), and are adsorbates (i.e. refrigerants) when working together with solid adsorbent (zeolite), so that the system could adjust the concentration of ammonia, allowing the system to operate at moderate pressure (i.e. near ambient pressure).

The differences between the cyclic behaviours of the single and binary components, briefly, are as follows. The Clapeyron sketch diagrams (Meunier et al., 1996) of the intermittent cycles are presented (Fig. 1) to analyse and compare the difference between the pressures of single and binary adsorption cycles.

In the adsorption phase *c*-*d*-*a*, the working medium evaporates, picking up the ambient heat. At same time, the adsorbent releases the heat of the adsorption process. In the regenerative phase *a*-*b*-*c*, the working medium is first changed to a vapour from the adsorbent by heating, and finally becomes a liquid in the condenser. In a cycle process, the driving force for the working medium flow is the pressure difference between P_c and P_{ev}.

As shown in Fig. 1(A), the operating pressure of the water-zeolite system operates between 0.6 kPa and 4.2 kPa at high vacuum in a refrigeration cycle, which is

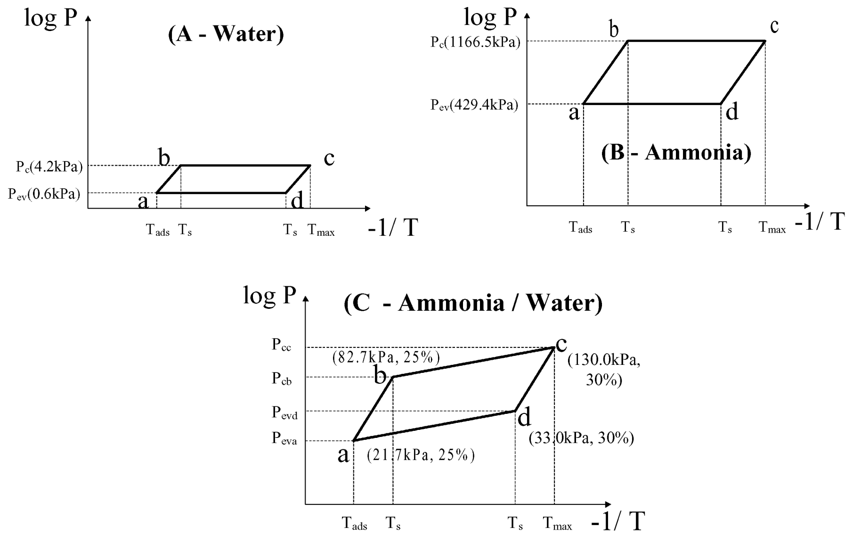

Figure 1. Clapeyron ideal cycle sketch diagram of single and binary mixture.

0.59–4.1% of the ambient pressure (101.3 kPa). However, in Fig. 1(B), the operating pressure of the NH_3-zeolite system operates between 429 kPa and 1166 kPa, which is 4–11 times the ambient pressure. The needs of high vacuum or high pressure usually result in a heavier design of the vessels of the system, system leakage, high maintenance and manufacturing costs, and even poor refrigeration performance. For example, an adsorption refrigerator will lose a 30–50% coefficient of performance (COP) if the pressure of the air leaked into the system of active carbon-CH_3OH is over 0.3 kPa (0.3% of ambient pressure) (Zhu et al., 1998).

In Fig. 1(C), a closed binary adsorptive cycle is presented referring to the property data given by Zhang and Zhao (1987). From *d* to *a*, the concentration of ammonia changes from 30% to 25%, and the evaporating pressure (P_{ev}) changes from 33.0 kPa to 21.7 kPa when the ammonia evaporates at 0°C. From *b* to *c*, the concentration of ammonia changes from 25% to 30% and the condensing pressure (P_c) changes from 82.7 kPa to 130.0 kPa when the desorption ammonia vapour is condensed and absorpted at 30°C. Therefore, the system operating pressure is from 21.7 kPa to 130.0 kPa, which is equivalent to 21.4% to 128% of the ambient pressure and close to the ambient pressure.

3. The Experimental Unit and Mathematical Model

An experimental unit of an intermittent binary adsorptive cycle system is developed and tested. The performance and characteristics of the ADAB system are compared with those of the pure adsorptive systems.

3.1. Experimental Unit and Measurement

The experimental unit of the ADAB system is shown in Fig. 2. It consists of three vessels (#3, #8, #11) made of glass for observing the processes of the cycles. During the desorption process, vessel #3, filled with zeolite, is used as the adsorber. It is connected to the evaporator (#11) and the condenser (#8). During the adsorption process, vessel #3 is used as a regenerator.

The main performance data of the ADAB system are temperatures, such as evaporating temperature, condensing temperature, adsorption temperature, desorption (regeneration) temperature and oil bath temperature, pressure, capacity of ammonia, and concentration of ammonia.

A high-speed computerized data acquisition system, with hardware and software from HP, records the temperatures measured by the thermocouples. The concentration of ammonia is calculated using the measured density according to the relation between its concentration and density. A dosimeter is put in the evaporator to measure the ammonia density.

3.2. The Performance of the ADAB System and Mathematical Model Analysis

The correlation of the binary adsorptive equilibrium is established (Szostak, 1992; Peng, 2000) according to the experimental results by the vacuum weight method in a closed system using zeolite and different concentrations of ammonia. The coefficient of performance (COP) of an adsorptive cooling system is defined as

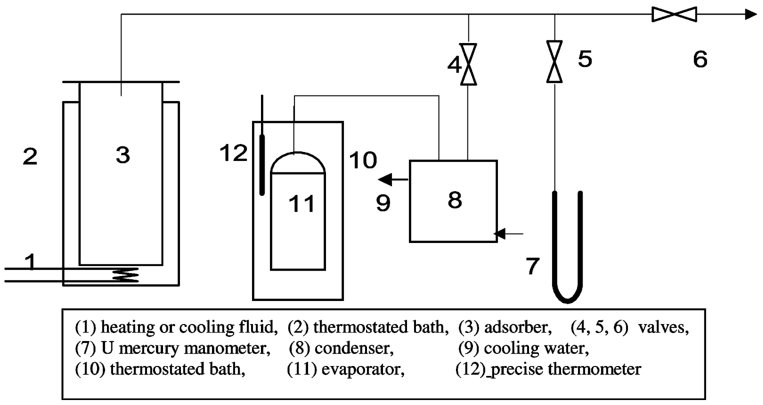

Figure 2. Schematic of the experimental unit.

the heat load in the evaporator per unit heat input to the generator, as illustrated in Eq. (1).

$$\mathrm{COP} = Q_{\mathrm{ev}}/Q_g \tag{1}$$

For the liquid phase of the unidea binary mixture (Prausnitz et al., 2000; ASHRAE, 1985), the enthalpy and entropy of the NH_3 vapour can be calculated by the improved R-K equation. The enthalpy of the NH_3-H_2O solution is calculated by the formula shown in Eq. (2).

$$h = X\left(\int_{T_0}^{T} C_{\mathrm{PNH_3}}\,dT + h^0_{\mathrm{NH_3}}\right) + (1+X)\left(\int_{T_0}^{T} C_{\mathrm{PH_2O}}\,dT + h^0_{\mathrm{H_2O}}\right) + \Delta h_{\mathrm{mix}}, \tag{2}$$

where Δh_{mix} is a function of temperature and concentration, which can be obtained using the Gibbs function shown in Eq. (3).

$$\Delta h_{\mathrm{mix}} = -RT^2\left[X\frac{\partial}{\partial T}(\ln \gamma_{\mathrm{NH_3}}) + (1-X)\frac{\partial}{\partial T}(\ln \gamma_{\mathrm{H_2O}})\right], \tag{3}$$

where $\gamma_{\mathrm{NH_3}}$, $\gamma_{\mathrm{H_2O}}$ are determined by the UNIQAC model (Pruasnitz, 1980). The differential terms can be assumed as constants due to very low changes in the liquid specific capacity and heat expansion in the cycle.

To simplify the calculation of COP, only the NH_3 vapour is taken into account, and the water vapour is neglected as its proportion in the vapour mixture is very low. Mass transfer limitation between the liquid-vapour interface and the solid-vapour interface is not considered. Therefore, Q_e is the actual evaporating refrigeration capacity, which can be calculated using Eq. (4).

$$Q_e = (L_{\mathrm{A_3}} - L_{\mathrm{A_2}})h_{\mathrm{mix}}, \tag{4}$$

where $L_{\mathrm{A_3}}$ and $L_{\mathrm{A_2}}$ are the masses of ammonia in the absorber at the end of desorption and at the end of adsorption respectively during the cycles when equilibrium has been established. h_{mix} is the evaporating enthalpy of ammonia. Because the proportion of pure water in the effective adsorptive capacity is very small, Eq. (4) can be rewritten as Eq. (5).

$$Q_e = (A_{\mathrm{N_3}} - A_{\mathrm{N_2}})h_{\mathrm{NH_3}}, \tag{5}$$

where $A_{\mathrm{N_3}}$ and $A_{\mathrm{N_2}}$ are the masses of pure ammonia in the absorber at the end of desorption and at the end of adsorption respectively. $h_{\mathrm{NH_3}}$ is the evaporating enthalpy of the NH_3 liquid. $A_{\mathrm{N_3}}$ and $A_{\mathrm{N_2}}$ can be calculated according to the volume, density, and mass concentration of ammonia. $h_{\mathrm{NH_3}}$ can be calculated by Eq. (6).

$$h_{\mathrm{NH_3}}(T) = h_{\mathrm{NH_3}}(239.7\,\mathrm{K})\left(\frac{1 - T/406.15}{1 - 239.7/406.15}\right)^n, \tag{6}$$

where $n = 0.321$ and Q_g is the regeneration heat, which involves the desorption heat and the sensible heat of the adsorbent. The calculation of Q_g is shown in Eq. (7).

$$Q_g = W_Z C p_Z \Delta t + Q_{\mathrm{des}} + (L_{\mathrm{A_3}} - L_{\mathrm{A_2}})Cp_{\mathrm{mix}}\Delta t, \tag{7}$$

where

$$\Delta t = T_d - T_a, \tag{8}$$

where T_d is the desorption temperature and T_a is the adsorption temperature. W_z and C_{Pz} are the mass and specific heat of zeolite respectively. $L_{\mathrm{A_1}}$ is the mass of ammonia at the beginning of the first cycle. $C_{P\mathrm{mix}}$ is the specific heat of the ammonia.

Because the proportion of pure water in the efficient adsorptive capacity is small, Eq. (8) can be rewritten as Eq. (9).

$$Q_g = W_Z C p_Z \Delta t + Q_{\mathrm{des}} + (A_{\mathrm{N_1}} - A_{\mathrm{N_2}})Cp_{\mathrm{NH_3}}\Delta t, \tag{9}$$

where $A_{\mathrm{N_1}}$ is the mass of NH_3 liquid at the beginning of the first cycle. $C_{\mathrm{PNH_3}}$ is the specific heat of the NH_3 liquid. In practice, a software package was developed for calculating the COP in different test conditions.

4. Experimental Results and Discussion

In the study, various tests were conducted to find out the ammonia concentration best suited to the new coupling cycle. The results of experiments using pure water, three concentrations of NH_3/water mixture, and one concentration of NH_3/water mixture with different initial liquid volumes in the evaporator are presented for comparison and analysis. Very good agreement between the thermodynamic description and the experimental results was observed when the experiments reached the equilibrium state.

4.1. *The Operation Process and Operating Pressure*

Figure 3 is the Clapeyron sketch diagram of the experimental cycle. The pressure of the ADAB system changed as the temperature of the adsorber changed. The curve with circus points represents a (non-realistic) cycle, using the regeneration temperature of 130°C, at equilibrium when the experiment was conducted very slowly over a long period. The curve with triangular points is a realistic experimental cycle when the experiment was conducted at a normal speed at the same operating temperatures. In the experimental cycle, the equilibrium points are *a* (30°C, 13.3 kPa), *b* (60°C, 53.2 kPa), *c* (130°C, 79.7 kPa) and *d* (85°C, 22.1 kPa). The points *e* and *f* of a normal cycle are in conditions (60°C, 10 kPa) and (100°C, 91 kPa) respectively.

It is worth to mention that the operation of the ADAB system was based on the Zeolite adsorbing NH_3 vapour in the adsorption process and H_2O absorbing NH_3 vapour in the desorption process. The vapour in the system is the mixture of NH_3 and H_2O from NH_3/water mixture. However, due to the different physical characteristic between NH_3 and H_2O (Zhang and Zhao, 1987), the vapour mixture is almost pure NH_3. For example, when NH_3/water mixture of 30% (weight) ammonia concentration evaporates at 0°C during at the adsorption process, the total pressure of mixture vapour is 33.0 kPa (P_{ev}) while the pressure of NH_3 vapour is 32.7 kPa, which is 99% of the total pressure. Therefore, ammonia distiller in the ADAB absorption refrigerators is no longer required and there is no significant limitation on vapour phase diffusion.

The pressure of the normal experimental cycle varied between 10 kPa (*e*) and 95 kPa (*f*), and the pressure equilibrium experimental cycle varied between 13 kPa (*a*) and 79.7 kPa (*c*). The pressure of the ADAB system was always higher than that of pure water systems. The pressure of the ADAB system was between 10 kPa and 93 kPa with the best concentration found. The pressure of the pure water systems was between 0.6 kPa and 5 kPa. The pressure of the ADAB system is 20 times that of the pure water system. We observed that the rate of evaporative vapour is high, so that the temperature of the ammonia drops rapidly to zero from ambient temperature during the first run of the system. There are a lot of bubbles in ammonia liquid, rising rapidly from the bottom to the top in the evaporator during the adsorption process.

During the tests, obvious changes of system operating pressure were observed when different concentrations of ammonia were used in the evaporator. When the concentration of ammonia is very low, the characteristics of the ADAB system are very close to those of the pure water/zeolite system. We note that the condensing pressure of the pure NH_3 vapour in other systems is over 1300 kPa, 10 times that in the ADAB system. The absorption process adopted as the condensing process of the ADAB system offers important advantages as far as the system operating pressure is concerned.

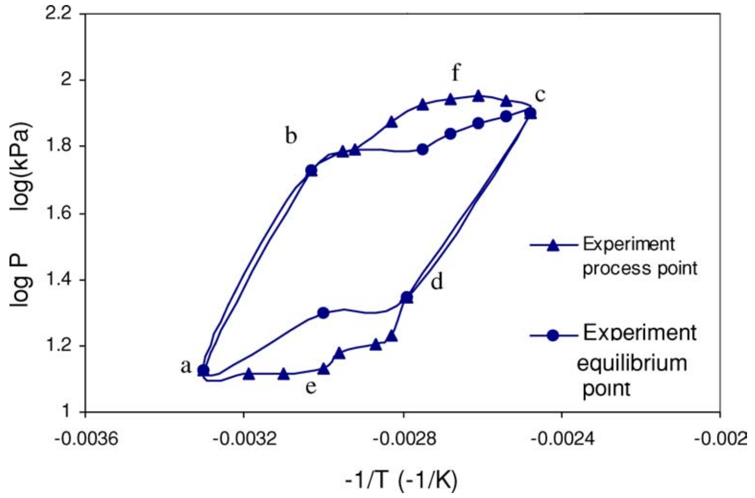

Figure 3. Clapeyron cycle sketch diagram of binary mixture (X_e).

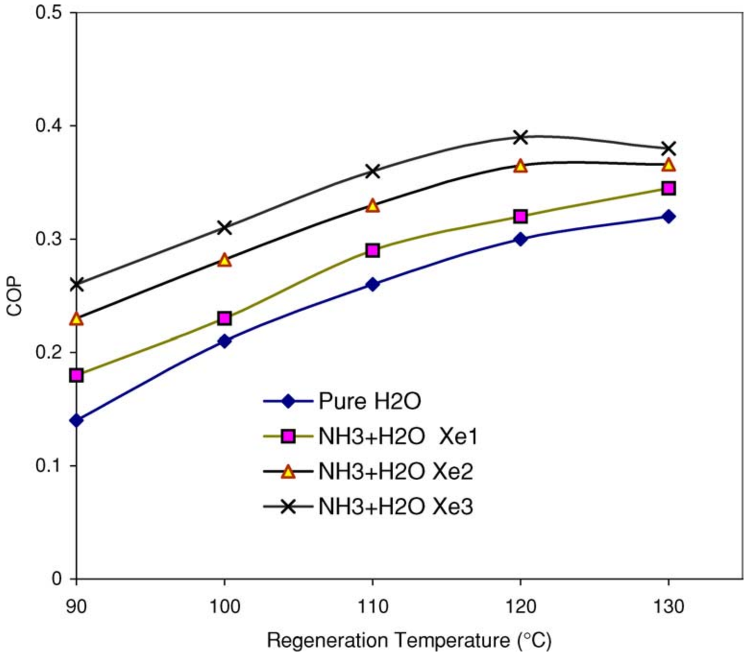

Figure 4. COP vs. regeneration temperature in different concentrations (X) of ammonia.

4.2. *Effects of Regeneration Temperature on COP*

The experimental results of the COP (Figs. 4 and 5) of the ADAB system using different ammonia concentrations (including pure water) under the same working conditions are presented and compared. Figure 4 shows the COP of the system at different regeneration temperatures when ammonia of different concentrations was used. Figure 5 shows the COP of the system at different regeneration temperatures when ammonia of different initial volumes (but the same concentration) was used.

As shown in Figs. 4 and 5, the COP of the binary adsorptive system was more sensitive to the temperature than the pure adsorptive system. The regeneration temperature remained the most important of the operation

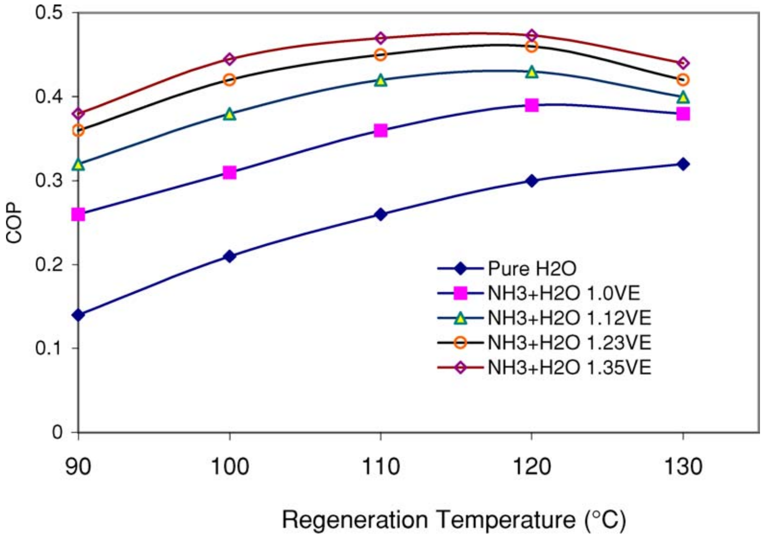

Figure 5. COP vs. regeneration temperature in ammonia (X_e).

parameters in both the binary and the pure component systems. The trend of COP with regeneration temperature is the same in both systems. However, the optimum regeneration temperature, corresponding to the highest COP, in the binary component system was lower than that of the pure water system.

The optimum temperature of the binary component system was about 120°C in the experiment at the recommended ammonia concentration. This shows that the new system is more suited to being driven by low temperature heat sources. When the ammonia concentration used was close to the recommended concentration, the COP increased, just as the regeneration temperature increased when the temperature was below 120°C and dropped when it was over 120°C.

4.3. *Comparison of COP of Pure and Binary Adsorbate Systems*

The COP of the binary component system of different ammonia concentrations is compared with the COP of the pure water system. The COP of the ammonia mixtures was significantly higher than that of pure water, as shown in Fig. 4. Within the tested range of ammonia concentration, the COP of the binary component system at the same regeneration temperature increased when the ammonia concentration increased. The ammonia concentration X_e offers the highest COP, and therefore the concentration of X_e is recommended for the experimental set up.

The operating pressure of the ADAB system increases with the ammonia concentration. The two extreme situations are a pure NH_3 or a pure H_2O with zeolite adsorption system. It was a pure water-zeolite adsorption system if the ammonia concentration was close to zero. It was a pure ammonia-zeolite adsorption system if the ammonia concentration was close to 1. The studies and comparison of these two extreme situations can be found in reference (Best et al., 1987).

4.4. *Effect of Initial Refrigerant Volume in Evaporator on COP*

Another new phenomenon was observed in the experiment. The ammonia volume in the evaporator affects the performance of system. To study such effects, four tests were conducted using four different initial volumes (ratio of volumes: 1:1.12:1.23:1.35) and the same (recommended) ammonia concentration (X_e) in the evaporator. The COP of the ADAB system increased at the same operation conditions when the initial volume of the refrigerant was larger, as shown in Fig. 5. Both the volume and the concentration of the ammonia in the evaporator were reduced during the adsorption process in the ADAB system. When a larger initial volume of ammonia is used in the evaporator, the ammonia concentration is reduced less during the adsorption process, and the pressure of ammonia vapour established at the end of adsorption is higher. This results in the higher adsorption capacity of zeolite and higher COP.

However, experiments show that there was no additional benefit when the initial volume was increased further over a certain limit. The low concentration of ammonia ($NH_3 + H_2O$) after the adsorption process was used to absorb the NH_3 vapour. Since there is always a high concentration of ammonia (around 30°C) left in the condenser at the end of the desorption process, it is moved to the evaporator and has to be cooled down by evaporation to the evaporating temperature (0°C) at the beginning of the adsorption process, resulting in a waste of the chilling capacity. Thus, a further increase of the initial volume will result in lower COP. Therefore, in a practical system, there exists an optimum initial volume that needs further investigation.

4.5. *Comparison Between ADAB System and Traditional Absorption Systems*

In traditional (NH_3/H_2O) absorption systems, the regeneration temperature is over 100°C and there is a large amount of water vapour in the ammonia vapour from the regeneration process. Distillers have to be used to separate the water from the ammonia vapour. However, the experiments show that the ammonia vapour at the outlet of the evaporator of the ABAD system is almost pure NH_3 vapour as a result of the low evaporation temperature (0–10°C). Therefore, the ADAB system does not require a distiller, and thus offers advantages over traditional NH_3/H_2O absorption refrigerators.

5. Conclusion

The new coupling cycle offers obvious advantages. Experimental results show that the COP of the ADAB system is higher than that of the pure water-zeolite system. The ammonia concentration and regeneration temperature affects the COP significantly. From the experiments, it was found that the proper regeneration temperature is about 120°C, while the COP of the experimental ADAB system would be maximized. The vapour evaporated from the ammonia ($NH_3 + H_2O$) liquid is almost pure NH_3vapour due to the large differential pressure between ammonia and water at evaporating temperature. This results in the ADAB system not needing an ammonia distiller, which is essential in traditional NH_3/H_2O absorption systems.

This study also shows that the use of NH_3/H_2O as the refrigerant and zeolite 13X as the adsorbent can achieve preferable operating conditions. The evaporative pressure of the system is around 13.3–26.6 kPa and the condensing pressure is around 50–90 kPa, which avoids the need for a high vacuum in the system. The ADAB system is easy to operate than other single adsorptive systems, due to its moderate operation pressure. This results in the vessels of the system being lighter in weight and causing less leakage into the system.

Nomenclature

A_{N_1}	Initial mass of pure ammonia (kg)
A_{N_2}	Mass of pure ammonia in the absorber when desorption begins (kg)
A_{N_3}	Mass of pure ammonia in the absorber when adsorption ends (kg)
Cp	Specific heat (kJ/kg · K)
Cp_z	Specific heat of zeolite (kJ/kg · K)
Cp_{mix}	Specific heat of ammonia (kJ/kg · K)
Cp_{NH_3}	Specific heat of ammonia (kJ/kg · K)
h_{NH_3}	Evaporating enthalpy of ammonia (kJ/kg)
h_{mix}	Evaporating enthalpy of ammonia (kJ/kg)
L_{a_1}	Initial mass of ammonia (kg)
L_{a_2}	Mass of ammonia in the absorber when desorption ends (kg)
L_{a_3}	Mass of ammonia in the absorber when adsorption ends (kg)
Q_{ev}	Heat of actual evaporating refrigeration capacity (kJ)
Q_g	Ideal regeneration heat, including desorption heat and sensible heat of adsorbent (kJ)
R	Ideal gas constant
T_a	Adsorption temperature (°C)
T_d	Desorption temperature (°C)
T_{ev}	Evaporation temperature (°C)
T_g	Regeneration temperature (°C)
X	Concentration of ammonia (kg/kg)
W_z	Mass of zeolite (kg)
γ	Activity coefficient

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