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ABSTRACT

An intermittent thermochemical sorption refrigeration system consisted of a reactor and an evaporator/condenser was constructed and studied by using strontium chloride/EG (expanded graphite) as composite sorbent and ammonia as refrigerant. Firstly, the sorption characteristic of strontium chloride/EG-ammonia was investigated, and the reaction enthalpy and entropy were obtained experimentally and compared with other previous data. Then, from the view of practical application, the specific cooling power (SCP) and coefficient of performance (COP) of the system were obtained at different operating temperatures. The regeneration process of $SrCl_2$ was driven by high-temperature heat transfer fluid with temperature of $94\pm1.5^{\circ}C$ from an external heat source, which was used to simulate solar collectors. In the evaporation–adsorption process, the evaporation temperature varied from 15 to $-15^{\circ}C$, and heat sink temperature was kept at $25\pm0.5^{\circ}C$. The experimental results showed that the COP increases with increasing evaporation temperature, and its maximum value could reach 0.24 when the evaporation temperature was $15^{\circ}C$, and the corresponding mean SCP was 291.5W/kg.

1. INTRODUCTION

Nowadays, three important issues, including greenhouse gas emissions, global warming and climate change have drawn the worldwide attention, and are urging people to enact the environmental regulations, which focus on how energy is obtained and utilized around the world. The Montreal Protocol (Velders *et al.*, 2007) and the Kyoto Protocol (United Nations, 1997) put restrictions on the production and use of ozone depletion substances such as CFCs' and HFCs', which are widely utilized in the vapor compression refrigeration systems and contribute to the ozone layer depletion and greenhouse gas emissions. Moreover, the electricity consumed by the system during operation increases the depletion of fossil fuels. Air conditioning and refrigeration industry is affected badly by the protocols. As consequence, researchers are required to explore new alternative ways to ease the issues mentioned above (Kiplagat *et al*., 2013).

Solid–gas sorption refrigeration systems have a lower negative environmental impact in comparison with vapor compression refrigeration systems by employing nature friendly fluids (e.g. water, methanol, ammonia, etc.) as refrigerants (Wang and Oliveira, 2006), among which ammonia has been appointed as a substitution of CFCs' and HFCs' by the International Refrigeration Institute (Spinner, 1993). These refrigerants have zero ozone depletion potential (ODP) and zero global warming potential (GWP), which meet the requirements of the Montreal Protocol and Kyoto Protocol. The working principle of a sorption refrigeration system is based on the sorbent's ability to absorb the refrigerant vapor at low temperature and to desorb it at high temperature. The sorption refrigeration machine can be driven by low-grade heat and not use electricity as the main driving energy. The low-grade heat

source could be industrial waste heat, exhaust gases from engines or solar thermal collectors. Therefore, the application of sorption refrigeration technology would also contribute to the efforts to solve the energy scarcity (Li *et al*., 2011). The comprehensive reviews in literatures (Wang and Oliveira, 2006; Meunier, 1998; Li *et al*., 2014) discuss the feasibilities of solid sorption systems and the progress in the development of different solid-gas sorption refrigeration thermodynamic cycle driven by low-grade thermal energy.

The sorption refrigeration machines that use chemical sorbents such as metal chlorides [Iloeje *et al*.,1995; Oliveira *et al*., 2007a; Kiplagat *et al*., 2010; Goetz *et al*., 1997] have been studied. Ammonia is often employed as the refrigerant in the machines. The chemisorption process follows the monovariant characteristic between pressure and temperature in the equilibrium condition. The main advantage of metal chloride working pairs is the larger sorption capacity and volume cooling density, which is about 5-6 times higher than those obtained with physical adsorption working pairs [Wongsuwan *et al*., 2001]. This implies that cooling systems based on thermochemical reaction process can have smaller reactor size and lower manufacturing costs when compared with those based on physical adsorption process. However, Metal chlorides usually suffer the drawbacks of low thermal conductivity and agglomeration, which will make the mass and heat transfer deteriorate and cause low heat addition/removal rates, resulting in long cycle times and low cooling power density. Hence, some porous materials such expanded graphite [Oliveira *et al*., 2009], carbon fibers [Aidoun and Ternan, 2002] and activated carbon [Cacciola *et al*., 1995; Wang *et al*., 2004] are employed as additives to prevent the agglomeration phenomenon so as to improve the mass and heat transfer in reactive salts. Moreover, machines using ammonia as the refrigerant have higher pressure in the system than those using methanol or water, which means better mass transfer.

Solar-powered sorption refrigeration has been regarded as a promising technology due to its distinct advantage of the close coincidence of high peak cooling demands with the maximum available solar thermal energy. Li *et al.* (2012) proposed a solar-powered multimode thermochemical sorption refrigeration system and its working mode could be switched for air-conditioning, refrigeration or deep-freezing according to the available solar insolation. Considering only single stage machines, Duenas *et al*. (2001) and Rivera *et al*.(2007) studied the dynamic behavior of solar heating system of a thermochemical refrigerator based on solid-gas reaction between barium chloride($BaCl_2$) and ammonia, and found that the relative low temperatures of dissociation were between 50°C and 60°C, which could be supplied by flat plate collectors. Moreover, their further research showed that the generation temperature was 53 °C for a condensation temperature of 23 °C, and in the evaporation-adsorption process, the evaporation temperature was between -10 and 0°C. Kiplagat *et al*. (2010) showed that the specific cooling capacity of a consolidated composite based on Lithium chloride and expanded graphite was 1.8-6.7 times higher than the value obtained when activated carbon acted as the sorbent. Oliveira *et al*. (2009) utilized a composite sorbent (NaBr and expanded graphite) in chemisorption air conditioning systems driven by low-grade heat source, and found the SCP and COP of the system is 129W/kg and 0.46 when the heat source temperature, the heat sink temperature and the evaporation temperature was 65°C, 30°C and 15°C, respectively. Li *et al.* (2009) developed a consolidated composite sorbent made of $BaCl_2$ and expanded graphite for solar-powered thermochemical cooling systems, and found the heat transfer and sorption characteristics could be improved significantly by using consolidated composite sorbent. Sanchez *et al*. (2015) studied the effect of the reactor's characteristics on the performance of a chemisorption refrigerator using an $SrCl_2$ composite sorbent with the aid of simulation, and found that it was possible to improve the cooling power and the COP of a chemisorption refrigerator with the same relative increment above their minimum value by adjusting the values of independent variables

By now, the report about the $SrCl_2/NH_3$ chemisorption refrigeration is very little, especially in consideration of the practical application. This paper describes a chemisorption refrigeration system based on $SrCl_2/EG$ composite sorbent and NH_3, which is driven by low-grade heat source. From the view of practical application, the effects of the different evaporation temperatures on the COP and SCP of the system are studied.

2. EXPERIMENT SYSTEM

2.1 System description

A small scale chemisorption experiment device was designed and constructed to operate under a single stage cycle (Figure 1). The device consisted mainly of a thermochemical reactor, a condenser/evaporator, and an expansion valve. The reactor was filled with $SrCl_2$/EG composite sorbent and the condenser/evaporator was used to store liquid NH_3. The auxiliary systems including two thermostatic circulators were set up to supply the heat for regeneration and evaporation process, and take the heat away for sorption and condensation process by pumping the heat transfer fluid with the setting temperature into reactor and condenser/evaporator. For the heat transfer fluid, it is the mixture of ethylene glycol /water by volume ratio of 2:3, and its freezing point is -40°C.
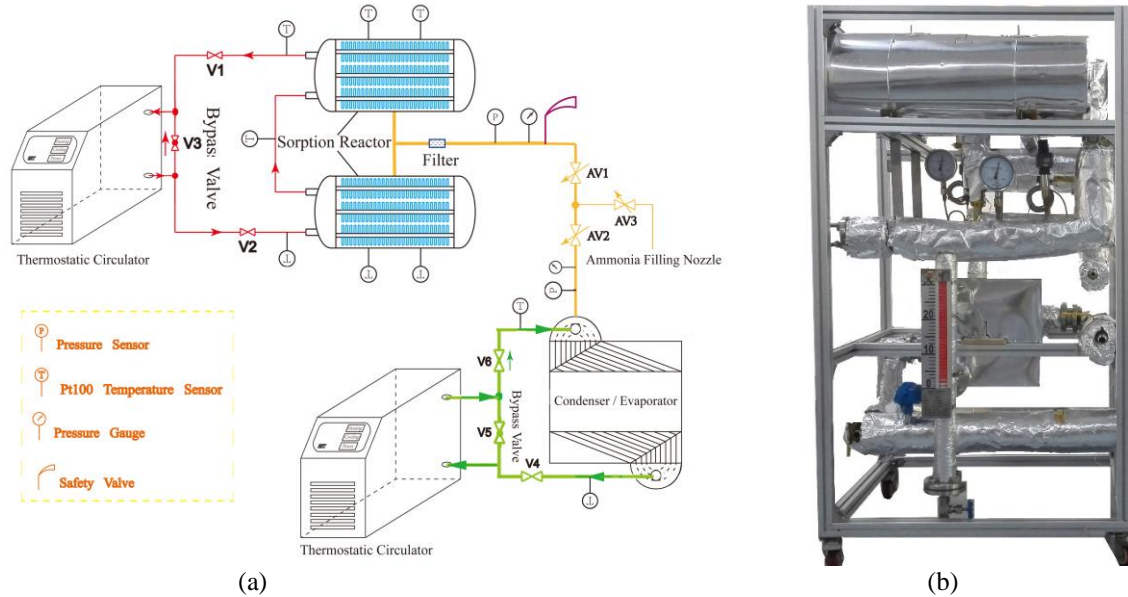

(a)   (b)

Figure 1: Experiment system, (a) the overall schematic of system; (b) the photograph of the main part

The reactor was connected to a condenser/evaporator through stainless steel pipes fitted with flow control valves. Measurement transducers such as temperature and pressure sensors were installed in the system.

2.1.1 Reactor: In the reactor, the decomposition and synthesis processes occur as shown in Equation (1).

$$SrCl_2 \cdot NH_3 + 7NH_3 \underset{\text{decomposition}}{\overset{\text{synthesis}}{\rightleftharpoons}} SrCl_2 \cdot 8NH_3 + \Delta H_r \tag{1}$$
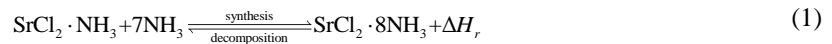
The reactor is mainly consist of 20 finned tubes made of carbon steel which are filled with the composite sorbents and covered by a thin wire mesh to prevent the sorbent from trickling out. Figure 2 shows the photographs of the filled fin tube heat exchanger and reactor without thermal insulation material.
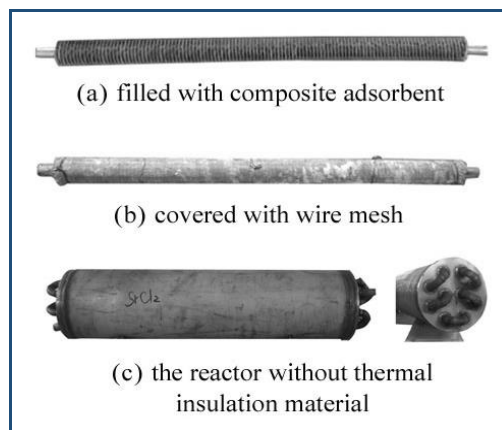

(a) filled with composite adsorbent

(b) covered with wire mesh

(c) the reactor without thermal insulation material

Figure 2: The photographs of fin tube heat exchanger and reactor

The salt was impregnated into expanded graphite powder according to the procedure given by Oliveira *et al*. (2007b), which involves expansion and exfoliation of raw expandable graphite (mesh 80, type KP80, from Qingdao Tianhe

Graphite Co. Ltd., China) at 800 ºC for 2 min and soaking it in a salt solution. Then, the resultant slurry was placed in a drying oven at 120 ºC for 24 h to remove the water. Thereafter, the composite sorbents were manually filled into the spaces between fins and the host tube. Finally, the fin tubes were arranged in a cylindrical steel case with an internal diameter of 151 mm, as shown in Figure 2. Before conducting the experiment, the reactor was dried at temperature between 170 ºC and 200 ºC in vacuum condition for about 1h in order to remove all the moisture absorbed by the salt during the filling process.

Table 1: Construction details of the reactor

Parameters	Value
Number of fin for every finned tube	149
Fin thickness	0.4 mm
Fin length	9.5 mm
Distance between fins	5 mm
Number of heat exchange tube	20
Inner diameter of heat exchange tube	⌀ 10mm
outer diameter of heat exchange tube	⌀ 16mm
length of every heat exchange tube	860mm
Inner diameter of shell	⌀ 151mm
Mass of salt (m_s)	3.73 kg
Mass of expanded graphite (m_{EG})	0.65 kg
Inert material/composite sorbent heat capacity ratio (R_{Cp})	11.8
Thickness of the adsorber case wall	4 mm
Pressure loading capacity of the reactor case wall	3.0 MPa

The heat transfer fluid exchanged heat with the sorbent through the walls of the finned pipes, whereas the refrigerant gas entered and left the sorbent through the pores of expanded graphite and the spare space in the reactor; hence, the steel case always contained some NH_3 during the operation of the system. The detailed parameters of the finned tube heat exchanger and the reactive bed are given in Table 1.

2.1.2 Data measurement and acquisition: In the inlet and outlet of reactor and condenser/evaporator, four thermocouples (PT100) were placed to measure the temperature of heat transfer fluid ($T_{Ad,in}$, $T_{Ad,out}$, $T_{Cd,in}$, $T_{Cd,out}$). In the reactor and condenser/evaporator, a mechanical pressure gauge and a pressure sensor were placed to measure the pressure (P_{Ad}, P_{Cd}). Also, four thermocouples were placed inside the reactor to measure the temperature of reactor ($T_{Ad,1}$, $T_{Ad,2}$, $T_{Ad,3}$, $T_{Ad,4}$), which were inserted into different depth in the radial direction of reactor, and one thermocouple was placed between the two sub-reactors to measure the temperature of heat transfer fluid ($T_{Ad,m}$) in the channel.

Table 2: Specifications of the measuring sensors

Equipment	Specification	Quantity	Accuracy
Temperature sensor	PT100 (-50~450ºC)	9	±0.15 ºC
Pressure sensor	0~4 MPa	2	0.5%
Pressure gauge	0~3.6Mpa	2	2.5%

The data from sensors were captured every 6 seconds and stored in an Agilent 34927A data logger. The specifications of the measuring sensors are shown in Table 2. The mass flow rate of the heat transfer fluid in the reactor and evaporator were measured by weighing the fluid which outflowed at setting time. For every experimental condition, the measurements were conducted for 3 times to obtain the mean value. It was because the effect of temperature on the viscosity of heat transfer fluid would lead to the change of mass flow rate of the heat transfer fluid. Moreover, the heat capacity of heat transfer fluid at different temperature was referred.

2.2. Experimental conditions

In this intermittent system, three independent parameters were controlled during the operation of the machine: (1) the inlet temperature of heat transfer fluid in the evaporator ($T_{Ev,in}$); (2) the inlet temperature of heat transfer fluid in the reactor during the regeneration period ($T_{Ad,in}$); and (3) the heat sink temperature. The time for regeneration process was set for 40 minutes, and the time for generation process 20 minutes. One experimental condition was repeated for 3 times to allow an estimation of the experimental error. The initial condition for every experiment was kept the same, where both the temperatures of reactor and condenser/evaporator were at 25±0.5 °C, and the valve was open.

2.3. Coefficient of performance (COP) and specific cooling power (SCP)
The COP is calculated with Equation (2), whereas the SCP is calculated using Equation (3):

$$COP = \frac{Q_{Ev}}{Q_{Ds}} \tag{2}$$

$$SCP = \frac{Q_{Ev}}{m_S t} \tag{3}$$

Where m_s is the mass of the salt (kg), t is the evaporation time(s), Q_{Ev} and Q_{Ds} are determined from the following equations, respectively:

$$Q_{Ev} = \sum_{i=0}^{200} \dot{m}_{Ev} C_p (T_{Ev,in} - T_{Ev,out})_i \Delta t \tag{4}$$

$$Q_{Ds} = \sum_{i=0}^{400} \dot{m}_{Ad} C_p (T_{Ad,in} - T_{Ad,out})_i \Delta t \tag{5}$$

where $\dot{m}$ is the mass flow rate of the heat transfer fluid, C_p is the specific heat capacity of heat transfer fluid. Δt is equal to 6 seconds.

3. RESULTS AND DISCUSSION

3.1 Determination of reaction enthalpy and entropy
Refer to the Equation (1). The relation between the equilibrium temperature and the pressure, and its stoichiometric coefficients were given in literature (Neveu and Castaing, 1993). In this work, we firstly characterized the equilibrium condition for this reaction, and confirmed the reaction enthalpy and entropy.
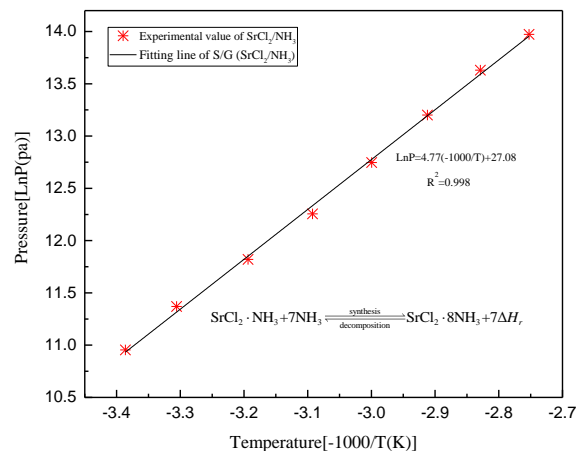

Figure 3: The equilibrium pressure of the reactor at different temperatures

Keep the valve closed, and record the equilibrium pressure of the reactor at different temperature, and then the fitting line could be obtained as Figure 3. It was found that the equilibrium temperature and pressure kept well linear relationship with the value of R^2 approaching to 1. According to the fitting line of S/G ($SrCl_2/NH_3$), we showed the corresponding Equation (6):

$$\ln P = -\frac{4770}{T} + 27.08 \tag{6}$$

$$\ln P = -\frac{\Delta H_r}{RT} + \frac{\Delta S_r}{R} \tag{7}$$

Combined with the gas constant $R = 8.3145 \text{J}/(\text{mol} \cdot \text{K})$, the values of ΔH_r and ΔS_r can be calculated depending on the Clapeyron equation (7). The table 3 lists the value of ΔH_r and ΔS_r obtained in this study and the data from (Neveu and Castaing, 1993), it is found that the biggest difference is just 4.2%, so the results are available.

Table 3: The comparison of reaction enthalpy and entropy

Reactant	ΔH_r (kJ/mol)	$\Delta S_r [\text{kJ}/(\text{mol} \cdot \text{K})]$	Reference
SrCl$_2$/NH$_3$(8-1)	39.66	225.16	Present study
	41.43	228.80	(Neveu and Castaing, 1993)

3.2 The temperature and pressure evaluation curve with time
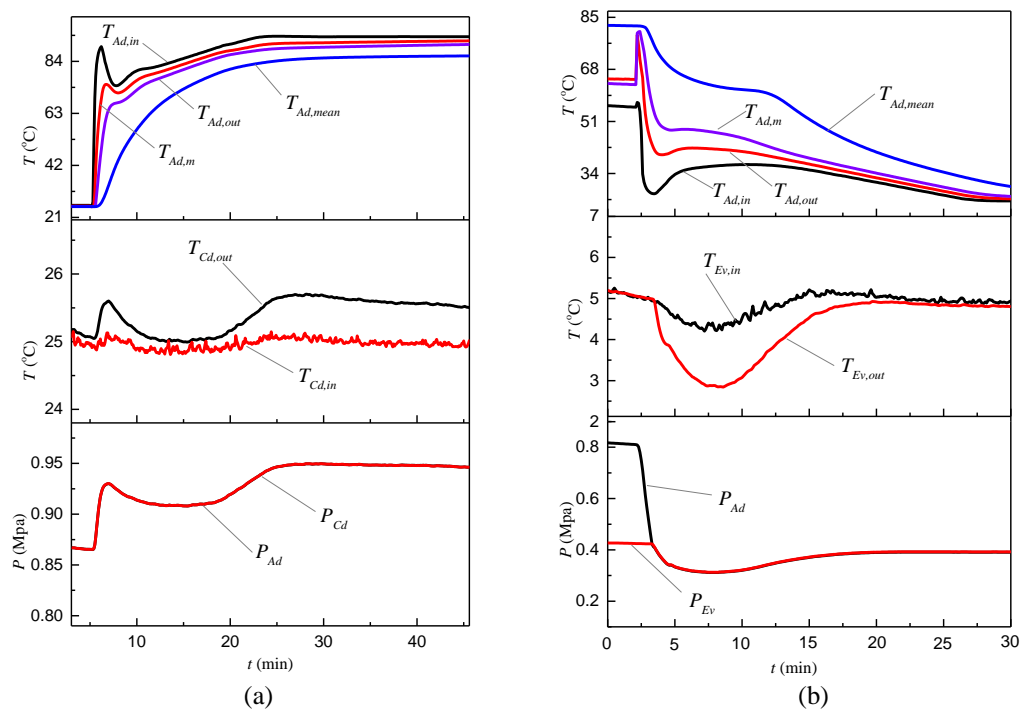

Figure 4: The temperature and pressure profiles of adsorber and condenser/evaporator in
(a) regeneration, and (b) adsorption stage

Figure 4 shows the temperature and pressure profiles of adsorber and condenser/evaporator in the decomposition and synthesis stage. The mean value of $T_{Ad,1}, T_{Ad,2}, T_{Ad,3}$ and $T_{Ad,4}$ was defined as $T_{Ad,mean}$. The operation process was described as follows: Firstly, the reactor and condenser were kept at ambient temperature (25±0.5ºC), and the valve kept open. At the 5th minute, the hot heat transfer fluid with temperature of 94±1.5 ºC was pumped into the reactor to heat the sorbent, at the same time, the heat transfer fluid with ambient temperature was pumped into condenser to keep it at constant temperature. Combined the profiles of temperature with pressure, it could be known that the temperature difference between the inlet and outlet of condenser began to increase obviously at about 20th min, which indicated that desorption or regeneration process took place. The corresponding temperature of reactor was 81ºC. The temperature difference between the inlet and outlet of reactor reached the stable value when temperature of reactor was 85ºC. Moreover, due to the open valve, the pressure value of reactor and condenser kept the same, and showed the same change trend with the outlet temperature of condenser. At the 45th min, the valve was closed, and the regeneration process was finished.

Then, the heat transfer fluid in the high-temperature thermostatic circulator was adjusted into the temperature of 25±0.5°C, and that in the low-temperature thermostatic circulator was adjusted into the temperature of 15±0.5 °C and then pumped into the condenser to cool it until 15±0.5°C. Meanwhile, the adsorber was cooled naturally.

Before the sorption stage, the pressure value in the evaporator was far lower than that in the reactor. In order to prevent the synthesis reaction being too drastic, it was necessary to lower the pressure value in the reactor by cooling it with the heat sink temperature of 25±0.5 °C from the high-temperature thermostatic circulator. Observe the pressure change in the reactor and open the valve until the pressure in the reactor was approaching to that in the evaporator. Then, the evaporation-sorption stage began, and the process lasted about 20 minutes.

3.3 Effect of the condensation temperature on the desorption mass of NH₃

When the heat source temperature is set at 94 °C, for the different condensation temperature of 30, 25, 20, 15, 10 °C, the desorption quantity ($m^{'}$) of NH₃ for per kilometer salt is calculated by the following equation:

$$Q_{Cd} = \sum_{i=0}^{400} \dot{m}_{Cd} \, C_p (T_{Cd,out} - T_{Cd,in})_i \Delta t \tag{8}$$

$$m_{NH_3} = \frac{Q_{Cd}}{\Delta H} \tag{9}$$

$$m^{'} = \frac{m_{NH_3}}{m_s} \tag{10}$$

Where the Q_{Cd} is the condensation heat released in the regeneration-condensation stage, ΔH is the phase change enthalpy of NH₃ at the condensation temperature, and the m_s is the mass of salt in the reactor.
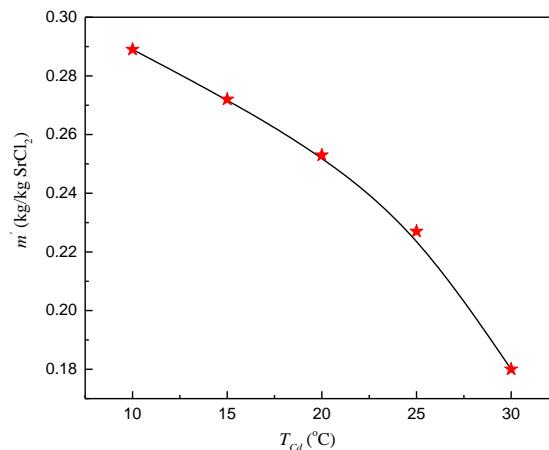

Figure 5: Variation of the desorption mass of NH₃ with the condensation temperature

Figure 5 shows the desorption mass of NH₃ changes with the condensation temperature at heat source temperature of 94±1.5°C. The regeneration mass of NH₃ decreases with increasing the condensation temperature. Depending on the sorption equilibrium relationship of SrCl₂/NH₃ obtained above, the condensation temperatures of 10, 15, 20, 25, and 30 °C are corresponding to the regeneration temperature of 74.6, 78.3, 83.5, 86.5, and 91.3°C. The sorbent needs to be heated to the regeneration temperature firstly, and then the regeneration process will take place. The regeneration process is conducted for 40 minutes, and the heat source temperature is kept at 94 °C, so the lower regeneration temperature corresponding to the condensation temperature will lead to bigger driven force for decomposition reaction, which means more desorption mass of NH₃. In theory, the maximum value of desorption mass of NH₃ is 0.75kg/kg SrCl₂. Here, the maximum value tested is 0.288kg/kg SrCl₂.

3.4 Effect of the evaporation temperature on the COP

Heat source temperature was kept at 94±1.5 °C, the heat sink temperature was kept at 25±0.5°C. For different evaporation temperature of -15,-10,-5, 0, 5, 10, and 15 °C, the COP of system was calculated by equation (2). Figure 6 shows that the COP varies with the evaporation temperature. Due to the constant heat sink temperature, the lower evaporation temperature made the driven force for regeneration reaction smaller, so the evaporation quantity of

liquid NH$_3$ decreased, which contributed to the lower cooling output and the lower COP. The maximum value of COP tested was 0.24 for evaporation temperature of 15 ^{o}C, and the minimum one was 0.13 for evaporation temperature of -15°C. Consider the heat capacity of reactor, including metal and composite sorbent heat capacity, the value was relatively high.

Compared with other researches, the COP was not high, and the one reason could be the large sensible heat load of the reactor, which led to more heat input to reactor in the regeneration stage. Obviously, this problem could be settled by increasing the quantity of salt and decreasing the proportion between the mass of the reactor and the mass of sorbent, which meant increasing the scale of the system. The other reason could be the short time of regeneration, it could be seen that the regeneration process was not enough depending on the outlet temperature evolution of the condenser when the regeneration process was conducted for 40 minutes. Here, we considered the compromise between the COP and SCP
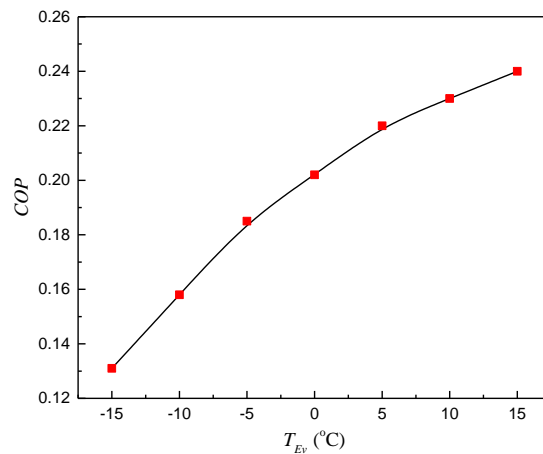

Figure 6: Variation of the COP with the evaporation temperatures

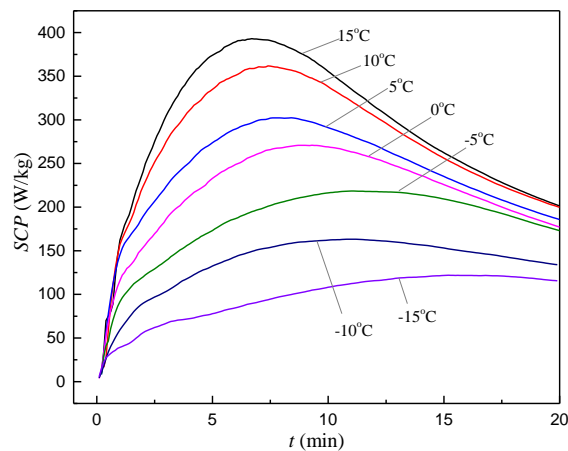### 3.5 Effect of the evaporation temperature on the SCP

Figure 7: The evolution curve of SCP with the time at different evaporation temperatures

The SCP of system was calculated according the equation (3). For different evaporation temperatures, the evolution curves of SCP with the evaporation time were showed in Figure 7, which indicated that for every evaporation temperature, the SCP increased firstly with the evaporation time and then decreased. In the initial stage of evaporation process, the pressure in the reactor decreased fast with its temperature decreasing, so the driven force between the reactor and evaporator increased, and evaporation process was sped up. It would reach the maximum value when it approached to the equilibrium pressure of SrCl$_2$/NH$_3$ corresponding to the evaporation temperature.

With the synthesis reaction going on, the driven force would decrease, so the evaporation rate decreased also. For different evaporation temperatures of -15, -10, -5, 0, 5, 10, and 15 ℃, the maximum values were 393.0, 361.8, 302.3, 270.9, 218.5, 163.2, and 121.9 W/kg, respectively, and the mean value were 291.5, 277.2, 241.0, 217.7, 183.2, 136.9,and 96.8 W/kg, respectively.

4. CONCLUSION

An intermittent thermochemical refrigeration system using strontium chloride ($SrCl_2$)/expanded graphite(EG) as composite sorbent and ammonia as refrigerant was constructed and studied from the view of practical application.

The sorption characteristic of $SrCl_2/NH_3$ was investigated, and the reaction enthalpy and entropy were obtained experimentally, which confirm previous reported data. Experimental results showed that the system could utilize effectively solar energy with temperature about 94 ℃ and output the cooling capacity. The COP obtained varied between 0.13 and 0.24 depending on the evaporation temperature ranging from -15 to 15℃. The mean SCP increased from 98.6 to 291.5W/kg in the same range of evaporation temperature. The value was far higher than that obtained with physical adsorption icemaker using consolidated activated carbon–methanol as working pair in which COP and SCP were 0.125 and 32.6 W/kg, respectively.

Nomenclatures

COP	Coefficient of performance
SCP	specific cooling power
EG	expanded graphite
Dimensional variables	
m	mass (kg)
R	ratio
T	temperature (°C)
P	pressure (Mpa)
C_p	specific heat capacity
ΔH	enthalpy change (kJ/kg) or (kJ/mol)
ΔS	entropy change [kJ/(kg·K)] or [kJ/(mol·K)]
t	time (min)
Subscript	
Ds	desorption
Cd	condenser/condensation
Ev	evaporator/evaporation
Ad	adsorption bed
in	inlet
out	outlet
s	salt
r	reaction

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