Experimental investigation on a $\text{MnCl}_2\text{-CaCl}_2\text{-NH}_3$ thermal energy storage system

L. Jiang, F.Q. Zhu, L.W. Wang, C.Z. Liu, R.Z. Wang*

*Institute of Refrigeration and Cryogenics, Key Laboratory of Power Machinery and Engineering of
Ministry of Education, Shanghai Jiao Tong University, Shanghai, 200240, China*

Abstract

Thermal energy storage (TES) is regarded as one promising technology for renewable energy and waste heat recovery. Among TES technologies, sorption thermal energy storage (STES) has drawn burgeoning attention due to high energy storage density, long-term heat storage capability and flexible working modes. Originating from STES system, resorption thermal energy storage (RTES) system is established and investigated for recovering the heat in this paper. The system is mainly composed of three high temperature salt (HTS) unit beds; three low temperature salt (LTS) unit beds, valves and heat exchange pipes. Working pair of $\text{MnCl}_2\text{-CaCl}_2\text{-NH}_3$ is selected for the RTES system. 4.8 kg and 3.9 kg MnCl_2 and CaCl_2 composite adsorbents are filled in the adsorption bed. Results indicate that the highest thermal storage density is about 1836 kJ/kg when the heat charging and discharging temperature is 155°C and 55°C, respectively. Volume density of heat storage ranges from 144 to 304 kWh/m³. The highest ratio of latent heat to sensible heat is about 1.145 when the discharging temperature is 55°C. The energy efficiency decreases from 97% to 73% when the discharging temperature increases from 55 to 75°C.

Keywords

Sorption; Resorption thermal energy storage; Energy density; Composite adsorbent

* Corresponding author. Tel.: +86-21-34208038; Fax: + 86-21-34206814
Email: lwwang@sjtu.edu.cn (L.W. Wang)

28 **Nomenclature**

ATES	Adsorption thermal energy storage
c	Specific heat (kJ/(kg.°C))
cha	Charging
COP_h	Coefficient of performance for heat storgae
$discha$	Discharging
ENG-TSA	Expanded natural graphite treated with sulphric acid
HSD	Heat storage density
HTS	High temperature salt
LTS	Low temperature salt
m	Mass flowrate (kg/s)
M	Mass (kg)
Q	Heat (J)
RTES	Resorption thermal energy storage
TES	Thermal energy storage
V	Volume (m ³)

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30 **Greek letters**

ε	Ratio of latent heat to sensible heat
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32 **Subscripts**

ad	Adsorbent
c	Condensing
h	Heat
i	Inlet
in	Input
l	Latent heat
m	mass
metal	Metal

o	Outlet
out	Output
sa	Salt
s	Sensible heat
v	Volume
w	Mass flowrate

1. Introduction

Thermal energy storage (TES) is considered as one paramount technology to utilize the renewable energy by weakening the mismatch between supply and demand. Reasonable utilization of renewable energy such as solar energy and geothermal heat inseparably depends on the development of sustainable and economically feasible TES systems. Under this scenario, an increasing number of attentions have been drawn to seek for the possible and suitable solutions to boost TES technology recently^[1].

Generally speaking, common TES technology can be divided into three categories, and they are sensible heat storage, latent heat storage and thermochemical energy storage^[2, 3]. Sensible heat storage is based on the heat exchanging process between energy storage materials such as pebble, concrete and heat exchange fluid such as water and oil^[4, 5]. Latent heat storage relies on the heat charging or discharging during phase change process of the materials such as paraffin, nitrate, etc.^[6]. However, both sensible and latent heat storage take disadvantages of low energy density, long-term and long-time storage and unacceptable energy loss^[7, 8]. Comparably, thermochemical energy storage can overcome these shortcomings, which can make up time-discrepancy, space-discrepancy as well as balancing supply and demand^[9].

As one common thermochemical process, adsorption process has been extensively studied for refrigeration and heat pump system^[10-12]. Similarly, adsorption working process has also been investigated for the possibility to be applied for TES system in recent decades. As for adsorption refrigeration, working pair including adsorbent and adsorbate can also be selected for the application of adsorption thermal energy storage (ATES) systems, and adsorbent means the materials which have the ability to adsorb and adsorbate means working fluid which can be adsorbed. Compared with only heat transfer in conventional sensible and latent heat storage systems, both heat and mass transfer processes

should be considered in ATES systems. Research results indicate that working performance of ATES system is not only dependent on selected materials and system design, but also on its thermodynamic cycle. For materials part, zeolite/H₂O, silica gel/H₂O and chloride/H₂O working pairs have been widely studied. Hauer^[13] reported an successful large-scale open sorption system with zeolite/H₂O working pair, which was installed in a school and connected with the local district heating network in Munich, achieving a heat storage density of 124 kWh/m³. Jae et al.^[14] developed several generations of prototype with silica-gel/H₂O working pair in a series of studies and experimentally achieved a material storage density of 50 kWh/m³. Yu et al.^[15] investigated a novel silica gel-LiCl₂ composite adsorbent for the application of ATES. The novel composite adsorbent could achieve a cold storage density of 108 kWh/m³ and a heat storage density of 163.6 kWh/m³ with 80°C desorption temperature by simulating adsorption properties. After that, to solve highly hygroscopic problem for silica gel-LiCl₂ composite adsorbent as well as to enhance the heat transfer ability, activated carbon (AC) and expanded natural graphite treated with sulfuric acid (ENG-TSA) were added since addition of other elements didn't influence the equilibrium water uptake of LiCl. Results showed that variation of bulk densities from 462 to 820 kg/m³ displayed thermal conductivities from 2.0 to 2.83 W/(mK), improving that of loose-packed AC-LiCl composite by at least 14 times^[16]. For thermodynamic cycle part, Li et al.^[17] analyzed different solid-gas reaction TES cycles for application of direct heat supply, heat upgrade and combined heating and cooling. Theoretical TES performances of different modes are compared, and advantages of different modes are pointed out. Lourdudoss et al.^[18] evaluated the three phase absorption heat pump process for energy storage, which indicated that three phase absorption process enjoyed a higher TES performance.

Like adsorption process, resorption process has also been investigated for refrigeration system gradually, which begins to be applied in the TES technology. Resorption process involves two different adsorbents such as halides. Compared with ATES system, resorption thermal energy storage (RTES) systems are characterized as safety feature and simple structure since there is no ammonia liquid in the system. Li et al.^[19] analyzed working pair MnCl₂-CaCl₂-NH₃ for energy storage and upgrade, and indicated its feasibility and high energy density. Bao et al.^[20] designed and established one resorption system for cold storage and long distance refrigeration. Results pointed out the optimal distance and period for the system.

Nonetheless, there is few research on the experiments for RTES system, especially for that

ammonia is rarely investigated for the application of TES in real system. In this paper, a novel RTES system is established and investigated with improved safety. $\text{MnCl}_2\text{-CaCl}_2\text{-NH}_3$ work pair is selected and ENG-TSA is used as additive to develop composite adsorbent. In order to have an overall understanding of this novel system, energy analysis is used for investigating the performance, and exergy analysis is not utilized since it is much lower than energy efficiency for energy storage system^[21].

2. Working process of RTES cycle

For RTES, there are commonly three modes for different application occasions, which are energy storage, energy upgrade and combined cooling and heating supply. This paper presents direct heat supply of energy storage mode. The working procedures of the RTES cycle can be generally divided into two processes: charging and discharging process. Fig.1 shows schematic diagram of thermal energy storage process. Fig.1a is working mode, and Fig.1b is thermal dynamic process, in which energy storage is between energy upgrade and combined heating and cooling supply. The charging processes consist of desorption process in HTS reactor and adsorption process in LTS reactor (Fig.1a). As Fig.1b shows, when HTS reactor is heated by heat source at point A, ammonia vapor which clings to adsorbent starts to escape as the threshold binding force between the adsorbate and adsorbent is exceeded. Ammonia vapor flows through a pipe and adsorbed by LTS reactor at a lower temperature level (point B). Adsorption heat $Q_{\text{ads-L}}$ is released to the surrounding environmental medium at point B. Compared with sensible and latent heat storage characterized as direct method, RTES cycle is considered as an indirect TES approach. This is mainly because the binding potential is stored in the adsorbent during the charging process. Simultaneously, adsorption heat is released to heat sink. In addition, part of sensible heat has to be brought into the system to heat up the adsorbent and other components. After the charging process, HTS and LTS reactor are separated from each other. When heating or cooling demand is needed, HTS and LTS reactor are connected with each other again. Similarly, discharging process proceeds in a reverse direction: it includes an adsorption process of HTS reactor and desorption process of LTS reactor (Fig.1a). Depending on practical demands, LTS reactor can produce a cooling effect of $Q_{\text{des-L}}$ at point C or HTS reactor can supply a heating effect at point D in Fig.1b.

Fig.1. Schematic diagram of thermal energy storage process. (a) Working mode, (b) thermal dynamic processes.

Fig.2a is the photo of RTES system. Six shell-and-tube type vessels are used as two adsorption reactors, i.e. HTS and LTS reactor. Temperature of HTS adsorption reactor is controlled by the oil tank while LTS adsorption reactor is controlled by ethanol water thermostat. 4.9 kg $MnCl_2$ and 3.8 kg $CaCl_2$ are impregnated in ENG-TSA with a mass ratio of 6:1 to form the composite adsorbent which is squeezed into the gaps of the finned pipes. The heat exchanging fluid flows through the finned pipes inside and refrigerant vapor occupies the rest space inside the shell. ENG-TSA is able to enhance the mass transfer and adsorption effectiveness due to its porous structure^[22]. The main apparatus of RTES system are an oil-water heat exchanger for cooling the HTS and LTS reactors, HTS reactor, an ethanol water thermostat, and LTS reactor. Fig.2b is photo of RTES system with auxiliary equipment, including oil tank, cooling tower, RTES system. Pressure sensors (full scale from 0 to 2.5 MPa with tolerance of $\pm 0.1\%$) are placed at HTS and LTS reactor. Pt100 thermal resistance temperature sensors are used with 0.5% accuracy. The rest of experimental data are monitored and logged by an Agilent 34972A. Based on all the accuracies of measurement sensors, the calculated errors of mass energy density and COP_h (coefficient of performance) are 4.5% and 5.6%, respectively.

Fig.2. Photo of RTES system (a) RTES system, (b) RTES system with auxiliary equipment.

To get the comprehensive understanding of performance of RTES system, different charging and discharging temperature are investigated and compared. Two working processes are involved in the experiment. First, the ammonia valve between HTS reactor and LTS reactor is open during the heat storage process. After that ammonia valve is closed and HTS reactor is cooled by the cooling tower. Then the ammonia valve is open again to supply the heat when HTS reactor reaches discharging temperature. Heat charging temperature of oil ranges from 135 to 155°C with the increment of 5°C. The cooling temperature is controlled as environmental temperature around 30°C. For sorption thermal energy storage, it is meaningful when the discharge temperature is higher than 40 °C. The heat discharging temperature for the experiment ranges from 55°C to 75°C, which can meet the domestic hot water requirement and part of the industrial requirement such printing and dyeing industry.

3. Performance evaluation

The charging heat of RTES system is calculated by:

$$Q_{h,cha} = \int_0^{t_{h,cha}} c_o \cdot m_o \cdot (T_{HTS,i} - T_{HTS,o}) dt \quad (1)$$

where $Q_{h,cha}$ is the charging heat provided by hot oil. $t_{h,cha}$ is charging time. C_o is specific heat of oil. $T_{HTS,i}$ and $T_{HTS,o}$ are inlet and outlet temperature of HTS reactor.

The discharging heat of RTES system is calculated by:

$$Q_{h,discha} = \int_0^{t_{h,discha}} c_o \cdot m_o \cdot (T_{HTS,i} - T_{HTS,o}) dt \quad (2)$$

where $Q_{h,discha}$ is the discharging heat. $t_{h,discha}$ is discharging time.

Heat storage density for both mass density and volume:

$$HSD_m = \frac{Q_{h,out}}{M_{ad}} \quad HSD_v = \frac{Q_{h,out}}{V_{ad}} \quad (3)$$

where HSD_m is mass energy density. HSD_v is volume energy density. M_{ad} is mass of adsorbent. V_{ad} is filling volume of adsorbent.

Heat storage efficiency is defined as:

$$COP_h = \frac{\text{Useful heat production}}{\text{Heat consumption}} = \frac{Q_{h,out}}{Q_{h,in}} = \frac{Q_{s,out} + Q_{l,out}}{Q_{h,in}} \quad (4)$$

where COP_h is heat storage efficiency of the resorption system. $Q_{s,out}$ is the sensible heat output. $Q_{l,out}$ is the latent heat output.

The ratio of latent heat to sensible heat can be expressed as:

$$\varepsilon_r = \frac{Q_{l,out}}{Q_{s,out}} \quad (5)$$

ε_r is the ratio of latent heat to sensible heat.

4. Results and discussions

4.3.1 Temperature and pressure trends

Since temperature and pressure take the same trends of RTES system in the working process, one representative working condition is selected for illustration. Fig.3 shows temperature and pressure variation under the condition of 155°C charging temperature and 55°C discharging temperature. Both inlet and outlet oil temperature of HTS reactors is indicated in Fig.3a. Three processes take around 80 minutes in total. Heating storage process, cooling process and heat supply process take 30, 10, and 40 minutes, respectively. Since cooling tower is used to cool the HTS reactor with oil-water heat exchanger, cooling process is much faster than other two processes. The maximum temperature increment in heat supply process is about 12°C, and average temperature increment is 7.1°C.

For pressure of HTS and LTS reactor shown in Fig.3b, since ammonia valve between HTS reactor and LTS reactor is open during the heat process, pressure variation for LTS and HTS reactor is same, which represents system pressure. Pressure increases sharply with the increasing temperature.

The highest system pressure is about 7 bar. When heating temperature becomes constant, pressure increases slowly. Heat storage process lasts for 30 minutes, after that the ammonia valve is closed. The

HTS reactor is cooled to charging temperature while LTS reactor is controlled as environmental temperature. Temperature difference causes pressure difference. When ammonia valve opens for discharging the heat, pressure of HTS and LTS reactor tends to be same again.

(a)

(b)

Fig.3. Variation of temperature and pressure in heat charging and discharging process (a) Inlet and outlet temperature of HTS reactor, (b) Pressure of HTS and LTS reactor.

As the mentioned above, RTES experiment is mainly involved in charging and discharging process. For different heat charging and discharging temperature, 20 groups of experiments are carried out to investigate the performance of RTES system.

Fig.4a indicates outlet temperature of HTS reactor with different discharging temperature at 155°C heat charging temperature. Results indicate that the maximum temperature increment and discharging time increase with the decrease of discharging temperature. When temperature of LTS reactor is 25°C, the theoretical heat discharging temperature is correspondingly 80°C. If heat discharging temperature become lower, effect of reaction heat is more significant with the larger driven potential. The lowest

200 outlet temperature increment is about 3°C and lasts for 20 minutes when inlet temperature is 75°C. In
201 contrast, the highest outlet increment temperature can reach 15°C and lasts for 40 minutes when the
202 inlet temperature is 50°C. In the early period after opening the ammonia valve, the outlet temperature
203 increases sharply then decreases gradually. This is mainly because the desorption rate of LTS reactor is
204 higher than adsorption rate of HTS reactors, and adsorption reaction of HTS proceeds fast. Fig.4b
205 indicates outlet temperature variation with different heat charging temperature at 55°C discharging
206 temperature. Results indicate that increment of heat charging temperature will be conducive to the
207 desorption process of HTS reactor, which mean more ammonia will be adsorbed by LTS in the
208 charging process. With the same discharging temperature, more cycle adsorption quantity will improve
209 the effect of discharging heat. This phenomenon will be obvious at 135, 140, 145°C heat charging
210 temperature. The outlet temperature increment is about 6-7°C and lasts for less than 25 minutes since the
211 desorption process is not complete at 135 °C heat charging temperature thoroughly. Comparably, at 140
212 and 145°C heat charging temperature, the outlet temperature increment is about 10°C and lasts for more
213 than 30 minutes. Therefore, heat charging temperature will have the large influence on RTES system.
214 For heat charging temperature higher than 145°C, increment of heat discharging temperature and time
215 are relatively close to each other, which indicates the HTS reactor nearly desorbs completely in
216 150-155°C. Higher heat charging temperature couldn't cause more desorbed ammonia. Table 1 shows
217 the largest outlet temperature increment for RTES system at different heat charging and discharging
218 temperature. It will indicate the variation trends more clearly. The increment of outlet temperature of
219 HTS reactor ranges from 3 to 12°C at different working conditions. 140°C heat charging temperature
220 and 65°C discharging temperature will be a critical condition to discriminate the temperature
221 increment.

Fig.4. The outlet temperature increment of HTS reactor for RTES system (a)different heat discharging temperature, (b)different heat charging temperature.

Table 1. The largest outlet temperature increment of HTS reactor for RTES system

The largest outlet temperature increment		Heat charging temp. °C				
		135	140	145	150	155
Discharging temp. °C	75	3	6	6	7	7
	70	5	6	6	9	9
	65	6	8	9	9	9
	60	6	8	10	10	11
	55	7	9	11	12	12

4.3.2 Energy density of RTES system

Heat storage density(HSD) for RTES system is calculated based on the filling mass and volume of adsorbent as shown in Fig.5. Fig.5a shows mass density of heat storage at different heat charging and discharging temperature. The highest thermal storage density is about 1836 kJ/kg when the heat charging and discharging temperature is 155°C and 55°C, respectively. At different heat charging and discharging temperature, mass density of heat storage decreases with the increment of heat discharging temperature and decrease of heat charging temperature. The mass density ranges from 702-1836 kJ/kg. The volume density of heat storage takes the same trends as the mass density at different heat charging and discharging temperature as shown in Fig.5b. In order to keep the consistency of the manuscript, kJ is used for the volume heat storage density though its magnitude will be a little larger. Volume heat

storage density ranges from 416654 to 1095125kJ/m³ at different charging temperature.

Fig.5. Heat energy storage density for RTES system (a)Mass density, (b)Volume density.

4.3.3 Energy efficiency of RTES system

With the same heat charging temperature of 155°C, Fig.6 shows the ratio of latent heat to sensible heat and heat storage efficiency at different discharging temperature from 55 to 75°C. It indicates that ratio of latent heat to sensible heat and heat storage efficiency decrease with the rise of discharging temperature which agrees with the trends of discharging temperature increment as shown in Table.1. The highest ratio of latent heat to sensible heat is about 1.145 when the discharging temperature is 55°C. Comparably, the lowest ratio is about 0.868 when the discharging temperature is 75°C. This is mainly because the discharging temperature has a great influence on the adsorption of HTS reactor and desorption of LTS reactor. The higher the discharging temperature is, the lower temperature drive force between HTS and LTS reactor is. Heat storage efficiency is determined by the discharging and charging heat. The increment of discharging temperature will decrease the sensible heat output, which will lead to the decline of heat storage efficiency when the charging heat is almost same. The energy storage efficiency decreases from 97% to 73% when the discharging temperature increases from 55 to 75°C.

Fig. 6. The ratio of latent heat to sensible heat and heat storage efficiency at different discharging temperature.

With the heat discharging temperature of 55°C, ratio of latent heat to sensible heat and heat storage efficiency at different heat charging temperature is shown as Fig.7. Results show that ratio of latent heat to sensible heat and heat storage efficiency change little when the charging temperature is higher than 145°C. This is because the reaction heat is influenced by the cycle adsorption quantity which is determined by the charging temperature. For the charging temperature higher than 145°C, HTS reactor can almost complete desorption in charging process thoroughly. Since the discharging temperature is same, cycle adsorption quantity changes little. In contrast, for the charging temperature lower than 145°C, ratio of latent heat to sensible heat decreases sharply. This is because HTS reactor only desorbs partly which causes the little ammonia adsorbed by the HTS reactor. The lowest ratio and heat storage efficiency is about 54% and 58%, respectively with 135°C charging temperature and 55°C discharging temperature.

Fig. 7. The ratio of latent/sensible heat and heat storage efficiency at different heat input temperature.

4.3.4 Potential improved performance

For theoretical thermal analysis of RTES system, mass ratio between metal and salts is generally about 1:1 with the same temperature difference. However, for real system the ratio cannot be reached by the constraints of manufacture processing and cost. In this $\text{MnCl}_2\text{-CaCl}_2\text{-NH}_3$ thermal energy storage system, this ratio between metal and salts is as high as 5.9. If mass of whole system is considered to calculate thermal energy density, the results will decrease to the 14.5% of original value. For the better performance, the improvement of the system is investigated with the decrease of ratio between mass of metal and adsorbent from 3 to 5.5:1, which is considered to be a reasonable range. Fig.8 shows mass thermal storage density of RTES system with different ratio between mass of metal and adsorbent. It indicates that the highest mass heat storage density is about 353.8kJ/kg at 155°C heat charging temperature. For 155°C heat charging temperature, the highest heat storage density is able to improve about 30% when compared with the lowest value at 155°C heat charging temperature. For different heat charging temperature and ratio, the highest heat storage density could improve as high as 135% when compared with the lowest value.

Fig.8. Mass thermal storage density of RTES system.

Fig.9 shows ratio of latent heat to sensible heat and heat storage efficiency at different ratio between metal and adsorbent when heat charging temperature is 150°C. It indicates that both heat storage efficiency and ratio of latent heat to sensible heat increase with decrease of ratio between metal

to adsorbent. Ratio of latent heat to sensible heat increases sharply while heat storage efficiency increases slowly. This is mainly because the increment of latent heat will lead to the decrease of sensible heat correspondingly. The total heat output doesn't increase greatly which causes the small increment of heat storage efficiency. It indicates that when the ratio between metal mass and adsorbent mass decreases to 3:1, the highest ratio of latent heat to sensible heat and heat storage efficiency can reach 1.78 and 0.961, respectively. The improvement can be 70% and 8% for ratio of latent heat to sensible heat and heat storage efficiency, which demonstrates the good potential performance of RTES system.

Fig.9. The ratio of latent heat and sensible heat and heat storage efficiency at 150°C charging temperature.

5. Conclusions

A novel resorption energy storage system was investigated and analyzed. Working pair of $\text{MnCl}_2\text{-CaCl}_2\text{-NH}_3$ was selected for the system. Resorption energy storage process was investigated at different heat charging temperature and discharging temperature. Conclusions were yielded as follows:

- (1) The maximum temperature increment and discharging time increase with the decrease of discharging temperature. The outlet temperature increment is about 5°C and lasts for 25 minutes when inlet temperature is 70°C. In contrast, the outlet increment temperature can reach 15°C and last for 40 minutes when the inlet temperature is 50°C. For the heat charging temperature higher than 145°C, heat discharging temperature increment and time lasted are relatively closed.
- (2) The highest thermal storage density is about 1836 kJ/kg when the heat charging and discharging temperature is 155°C and 55°C, respectively. At different heat charging and discharging temperature, mass energy density of heat storage decreases with the increment of heat discharging

temperature and decrease of heat charging temperature. Mass density ranges from 702 to 1836 kJ/kg. Volume energy density of heat storage takes the same trends as the mass energy density at different heat charging and discharging temperature, which ranges from 416654 to 1095125kJ/m³.

(3) The highest ratio of latent heat to sensible heat is about 1.145 when the discharging temperature is 55°C. The energy efficiency decrease from 97% to 73% when the discharging temperature increases from 55 to 75°C. The ratio of latent heat to sensible heat and heat storage efficiency changes little when the charging temperature is higher than 145°C. For the charging temperature lower than 145°C, ratio of latent to sensible heat decrease sharply. The lowest ratio and heat storage efficiency is about 54% and 58%, respectively at 135°C charging temperature and 55 °C discharging temperature.

(4) The system performance has the potential to be improved when the ratio between metal and adsorbent decreases. If the ratio can be decreased to 3:1, ratio of latent heat to sensible heat and heat storage efficiency can reach 1.78 and 0.961, respectively, which improve 70% and 8%, respectively.

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Figure Captions

Fig.1. Schematic diagram of thermal energy storage process.(a) Working modes, (a) thermal
dynamic process.

Fig.2. Photo of cogeneration system (a) Resorption refrigeration part, (b) Cogeneration system
with auxiliary equipment.

Fig.3. Variation of temperature and pressure in energy storage and supply process (a) Temperature,
(b) Pressure.

Fig.4. The outlet temperature increment for RTES system (a)different heat releasing temperature,
(b)different heat source temperature.

Fig.5. Heat energy storage density for RTES system (a)Mass density, (b)Volume density.

Fig.6. The ratio of latent/sensible heat and heat storage efficiency at different discharging

404 temperature.

405 Fig.7. The ratio of latent/sensible heat and heat storage efficiency at different discharging

406 temperature.

407 Fig.8. Mass thermal storage density of RTES system.

408 Fig.9. The ratio of latent/sensible heat and heat storage efficiency at 150°C charging

409 temperature.