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SOLAR ADSORPTIVE CHILLER BASED ON COMPOSITE SORBENT 'SILICA GEL-SODIUM ACETATE' SYNTHESIZED BY SOL-GEL METHOD

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A composite sorbents 'silica gel-sodium acetate' synthesized by sol-gel method is investigated in this work. The composite structure is investigated by wide-angle X-ray scattering. The change of the adsorption properties of CH_3COONa crystals is revealed to be resulted from their dispersion up to nanoscale. The sorption capacity of the composite 'silica gel-sodium acetate' is stated to be 0.42 g/g that exceeds noticeably the linear superposition of the sorption capacity of the silica gel and sodium acetate mechanical mixture. The difference between water sorption kinetics for massive sodium acetate and for 'silica gel-sodium acetate' composite is shown. The solar adsorptive chiller design is developed. Its operating characteristics are determined when 'silica gel-sodium acetate' composite is used.

Keywords: composite sorbent, adsorption, coefficient of energy performance, solar refrigeration, adsorptive chilling, energy efficiency.

Introduction

Modern chillers and air-conditioners are electric energy intensive devices especially in a hot season. Irreplaceable resources are used up for electricity generation, a power-supply system overload and serious accidents being resulted from electric power consuming in a summer season. Using of solar adsorptive chillers is one of the methods for electric demand decreasing and low-potential heat energy utilization. Their base elements of construction are solar collector, adsorber, condenser and evaporator placed in cold box [1]. Generally, adsorptive chiller operating is realized into two stages. The first is adsorption and refrigerant (cooling agent) evaporation decreasing of temperature in cold box. The second is adsorbent regeneration, i. e. desorption and cooling agent condensation. At this stage adsorbent is heated up to regeneration temperature by external heat sources. Water [2], ammonia [3] and methanol [4] are used as cooling agents. Sorbents are activated carbon [4], silica gel [5], zeolite [6], salts $MnCl_2$, NH_4Cl [7], NaBr, $BaCl_2$ [8] and composite materials $LiNO_3$/vermiculite [9], LiCl/silica gel [10]. Operating process is complicated with low sorption capacity of typical sorbents and environmental danger and corrosiveness of massive salts. Furthermore, during massive salts hydratation in steady-state condition water access anhydrous salts is generally disabled by hydrate films formation, deceleration of sorption and cooling being resulted from these processes. Massive salt cycling is impossible without mechanical dispersion during operating process. This problem can be surmounted by generation of nano-disperse sorbents 'salt inside porous matrix' [11]. However, the central failure of all considered materials is regeneration temperature which is higher then 90⁰C. This chacteristic abridges essentially utilization of alternative energy sources and low-potential heat emissions of industrial units. Furthermore, high regeneration temperature results in coefficient of performance energy: 0.136−0.159 for activated carbon [4], 0.28−0.35 for zeolite [6], 0.3−0.32 for massive salts [7], for composite materials 0.33−0.59 [9,10]. The main tendencies of chilling machine cycle efficiency rising consist in specific-purpose synthesis of sorbents which are corresponded to adsorptive heat reformer cycle requirements, specifically, to highest possible coincidence between 'strong' and 'weak' isosteres [12]. However, purposeful development of composite sorbent 'salt inside porous matrix' demands of intensive study of extensive evidence on both sorption equilibrium and sorption kinetics.

The aim of present work is development of solar adsorptive chiller based on composite sorbent 'silica gel-sodium acetate' synthesized by sol-gel method and investigation its operating properties.

Experimental

The composite sorbents 'silica gel - sodium acetate' synthesized from tetraethoxysilane, CH_3COONa, ethanol (as a solvent) and hydrochloric acid (as a catalyst) were chosen as the objects of investigation. Polyionenes served as organic modifiers. The structural formulas of polyionenes are given below:
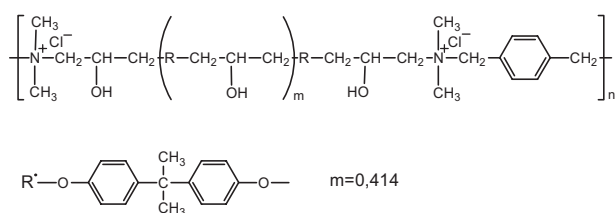
Synthesis technique is described in detail in [11]. Diameters of the composite sorbents granules are 0.5−1.5 mm.

Wide-angle X-ray scattering (WAXS) profiles of the samples were collected in the transmission mode using a DRON-2,0 diffractometer. Nickel-filtered $Cu−K_\alpha$ radiation (radiation wavelength λ=0.154 nm) was produced by an IRIS-M7 generator at an operating voltage of 30 kV and a current of 30 mA. The scattering intensities were measured using a scintillation detector scanning in 0.2^0 steps over the range of angles of 2 to 40^0 [13]. IR-spectrum were registered on UR-20. The DTA analysis was conducted on a Paylic-Paylic-Erday derivatograph Q-1500, using a heating rate of 10^0C/min, from room temperature to 900^0C. The samples mass is 200 mg. Water sorption kinetic investigation were carried out with the setup described in [14].

The basic elementary units of adsorptive chiller (Fig. 1) are adsorber (1), evaporator (4) placed in cold box (5) and condenser (7). A coil pipe (2) is installed in adsorber for reducing the influence of weather during regeneration. Chilling box volume is 0.175 м^3. It is made of aluminium sheets thickness 0.5 mm. Mineral wool is used as thermal insulation. Transparent cellular polycarbonate plastic CAH (thickness is 8 mm, integral transmittance coefficient is 0.88) is fixed on adsorber front side. A sorbent is placed on adsorber back side. Composite material 'silica gel-sodium acetate' is used as a sorbent. In the case of suggested polymer solar collector are installed the double decker absorber with tubes for sorbate circulation, solar energy absorber, transparent insulation placed on absorber, tubes for input and output of sorbate and heat insulation. At that, solar radiation absorber fixed on the lower deck is made of mixture of activated carbon (5%) and sorbent (95%), the upper deck being filled with air. Tubes for circulating sorbate are placed inside the solar radiation absorber. The absorber upper deck filled with air is used as transparent insulation in this construction (Fig. 2). The solar collector surface area is 1.7 m^2.

Adsorptive chiller works according to thermo-dynamic cycle shown on Fig. 3. Cold generation due to adsorption and evaporation of water is described by line 3−4−1. Sorbent regeneration resulted from desorption and condensation of water is represented as line 1−2−3.
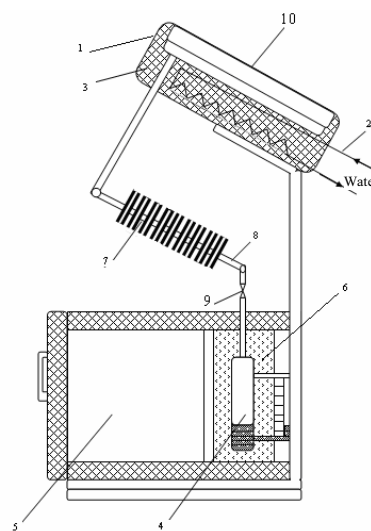

Fig. 1. Scheme of solar sorption chiller: 1 − adsorber; 2 − coil pipe; 3 − sorption material; 4 − evaporator; 5 − cold box; 6 − water accumulator of chill; 7 − condenser; 8 − pipe; 9 − tap; 10 − transparent cellular polycarbonate plastic
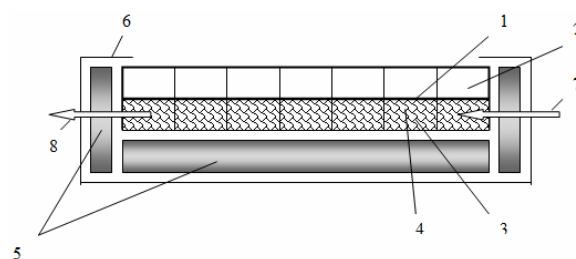

Fig. 2. Polymer solar collector cross-section: 1 − absorber; 2 − absorber upper deck being transparent insulation; 3 − absorber lower deck; 4 − absorber of solar radiation; 5 − heat insulation; 6 − case; 7, 8 − tubes for input and output of sorbate
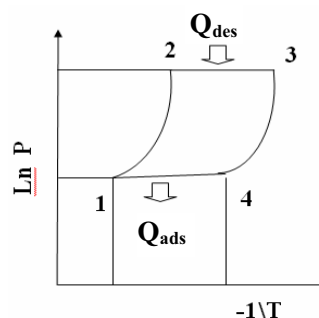

Fig. 3. Adsorption chiller operating cycle

Temperatures of environment, absorber and air layer between absorber and transparent insulation are controlled by group of thermal converters of industrial version Pt1000. Their operation range is from $−50^0$C to 500^0C, the class of error limit is A, limit deviation from nominal static characteristic is

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±(0.15+0.002t). Radiant flux density is measured by pyranometer Kipp&Zonen CM21 disposed to prevent shadowing on surface of heat exchanging. Total solar radiation density is registered in the range of wave-length from 300 to 3000 nm. Setup tolerances are 1%, operative temperature range is from −40 to +80⁰C.

Regeneration temperature of adsorbent is stated up-to-date 60⁰C. Environment temperature is maintained from 15 to 35⁰C.

The coefficient of performance (COP) of an adsorptive chilling system is calculated as the ratio between its capacity to remove heat from a cold source and the energy supplied for its operation:

$$COP_c = \frac{Q_e}{Q_{reg}} , \qquad (1)$$

where Q_e corresponds to the cooling effect produced at the evaporator, Q_{reg} is only the energy that is effectively transferred to the adsorber to promote its regeneration.

The value of Q_e is determined as the difference between the refrigerant latent heat of vaporization and the sensible heat required to cool down the refrigerant from the condensing temperature to the evaporation temperature:

$$Q_e = M \square [h_e - C_p \square (T_c - T_e)], \qquad (2)$$

where M is the mass of refrigerant desorbed in one operation cycle, h_e − refrigerant latent heat of vaporization, C_p is the specific heat of the liquid refrigerant, T_c and T_e correspond to the temperatures of condensation and evaporation, respectively.

The value of Q_{reg} is determined as the sum of the heat Q_{d1} applied to the adsorbent when both temperature and pressure increased along the line 1−2 (Fig. 3) and the heat Q_{d2} caused the adsorbent to release the refrigerant vapor (Fig. 3, line 2−3):

$$Q_{reg} = Q_{d1} + Q_{d2}. \qquad (3)$$

Coefficient of performance (COP) of a solar chilling system is defined as a ratio of the heat removed from the cold source (Q_e) and all the solar energy received by the solar collector surface (Q_s) [1]:

$$COP_s = \frac{Q_e}{Q_s} . \qquad (4)$$

The solar energy provided to the system is defined as a product of the solar collector surface area A_c by the integration of the solar radiation flux on the collector surface (G) during one day of operation [1]:

$$Q_s = A_c \cdot \int_0^{24 \cdot 3600} Gdt . \qquad (5)$$

On the other hand, effective or net solar COP is determined assuming that the cooling effect is used to cool down the water [1] or other substances contained in the cold box:

$$COP_{s(n)} = \frac{m \cdot c_p \cdot \Delta T_l}{A_c \cdot \int_0^{24 \cdot 3600} Gdt} , \qquad (6)$$

where m is the mass of water or other substances placed in the cold box, c_p is their specific heat, DT_l is their temperature variation.

When quantity of heat received by composite taken into account, Eq. (6) transform to:

$$COP_{s(n)\eta} = \frac{m \cdot c_p \cdot \Delta T_l}{\eta \cdot A_c \cdot \int_0^{24 \cdot 3600} Gdt} , \qquad (7)$$

where η is the solar collector coefficient of efficiency.

Furthermore, net solar COP is calculated by experimental average monthly temperature dependences of environment and within the cold box in the course of 24 hours:

$$COP = \frac{k \left(\int_0^{24 \cdot 3600} T_{env} d\tau - \int_0^{24 \cdot 3600} T_{ch} d\tau \right)}{A_c \cdot \int_0^{24 \cdot 3600} Gdt} , \qquad (8)$$

where k corresponds to heat capacity of the cold box material and air in it, kJ/K:

$$k = C_M \cdot M_M + C_A \cdot M_A + C_S \cdot M_S , \qquad (9)$$

where C_M, C_A and C_S are specific heats of cold box material, air in it and substances placed into cold box, M_M, M_A, M_S are masses of cold box material, air in it and substances placed into cold box, $\int_0^{24 \cdot 3600} T_{env} d\tau$ and $\int_0^{24 \cdot 3600} T_{ch} d\tau$ are areas under temperature curves of environment and inside cold box.

Solar collector efficiency having been taken into consideration, effective solar COP is determined as

$$COP_\eta = \frac{k \left(\int_0^{24 \cdot 3600} T_{env} d\tau - \int_0^{24 \cdot 3600} T_{ch} d\tau \right)}{\eta \cdot A_c \cdot \int_0^{24 \cdot 3600} Gdt} . \qquad (10)$$

Different methods were used when COP calculated. As efficiency of solar adsorptive chillers analyzed, a controlling factor is determination of

Coefficients of performance of adsorptive chiller based on composite sorbent 'silica gel-sodium acetate'

Month	Total monthly average solar radiation value, kWh/m^2	COP_c	$COP_{s(n)}$	$COP_{s(n)_\eta}$	COP	COP_η
May	5,99	1.021	0.175	0.291	0.100	0.167
June	5,86	1.021	0.179	0.298	0.102	0.171
July	6,06	1.021	0.173	0.288	0.099	0.165
August	5,66	1.021	0.185	0.308	0.106	0.176
September	4,41	1.021	0.238	0.396	0.136	0.227

supplied heat: quantity of heat consumed by sorbent regeneration, absorbed by collector for 24 hours or received by composite. Various methods of COP calculation are given in [1].

According to results given in the Table, coefficients of performance of solar adsorptive chiller differ substantially. To ascertain efficiency COP of investigated adsorptive chiller is compared to COP of chilling devices using water as refrigerant. Values of net COP of developed chiller estimated by theoretical calculations and determined with experimental results are highly competitive with solar adsorption water coolers and refrigerators used working pairs 'silica gel-water' and 'zeolite-water' [1]. Moreover, enhanced sorption properties of composite sorbent 'silica gel-sodium acetate' allow reducing solar collector surface.

Results and discussion

Investigation of synthesized composite sorbent is focused mainly on its sorption properties and on possibility of reversible sorption and desorption of water vapor because of their defining thermochemical performance and low-potential heat storage. These characteristics are affected by distribution and dispersivity of salt in silicon oxygen skeleton.

Wide-angle X-ray scattering (WAXS) is carried out to estimate these effects. WAXS profiles of the anhydrous silica gel KSM (1), composite sorbent 'silica gel-sodium acetate' hydrated (2) and dehydrated (3) after 10 cycles of sorption and desorption are on Fig. 4. Amorphous order of structural organization is typical for silica gel. Crystalline reflexes according to sodium acetate are observed on WAXS profile of composite sorbent. Crystalline peaks disappearance is noticed after composite hydration, i. e. composite is passed into amorphous state. It is significant that after multiple cycling (sorption-desorption) intensity and shape of peaks are not changed, it being evidence of composite structure invariability and absence of migration processes of sodium acetate in composite.

It is confirmed by DTA-analyses results. The composite 'silica gel-sodium acetate' mass loss after desorption during heating to 140−1000^{0}C is 2%. The composite mass loss after adsorption is observed at 32−100^{0}C to be 42%.
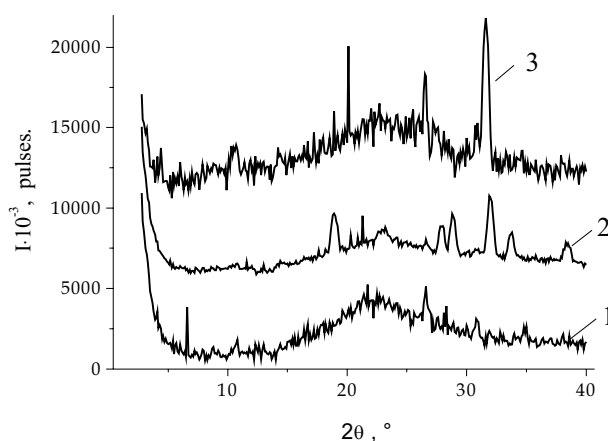

Fig. 4. WAXS profiles of the anhydrous samples: 1 − silica gel KSM; 2 − composite 'silica gel-sodium acetate' hydrated; 3 − composite 'silica gel-sodium acetate' dehydrated

DTA analyses of trihydrate sodium acetate, anhydrous sodium acetate and silica gel KSM dried to constant mass at 100^{0}C confirms difference of investigated composite sorbents and massive crystalline hydrate thermal physical properties silicon oxygen matrix investigated composites. Anhydrous sodium acetate during heating to 1000^{0}C losses 5% by mass. Mass losses of $CH_3COONa \square 3H_2O$ are 39% at 75−140^{0}C. Mass loss silica gel KSM due to adsorbed water removal at heating to 1000°C is 15%.

Thus, sorption properties of composite 'silica gel-sodium acetate' are differed rather from massive salt and silica gel due to peculiarities of this disperse system. Sorption capacity of the «pure» silica gel is 0.18 g/g, massive sodium acetate is 0.65 g/g, the composite 'silica gel-sodium acetate' is 0.42 g/g. From common rule of additivity sorption capacity of linear superposition of mechanical mixture of silica gel and sodium acetate is calculated by formula:

$$X_\Sigma = a \cdot X_{H_2O}^{salt} + (1-a) \cdot X_{H_2O}^{SiO_2}, \qquad (11)$$

where a is sodium acetate mass concentration in composite a=0.30; $X_{H_2O}^{salt}$, $X_{H_2O}^{SiO_2}$ are sorption capacities of massive salt CH_3COONa and silica gel, i. e. quantities of water (g) adsorbed by 1 g sorbent,

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$X_{H_2O}^{salt}$ =0.65 g/g, $X_{H_2O}^{SiO_2}$ =0.18 g/g.

Linear superposition of sorption capacity of mechanical mixture of silica gel and sodium acetate is 0.32 g/g. Thus, sorption properties of synthesized composite sorbent 'silica gel-sodium acetate' are not linear combination of silica gel and massive salt properties. Therefore, synthesized composite sorbents are possessed of unique structure increasing rate of reaction of the crystalline hydrate and water vapor in developed pores of silicon oxygen matrix.

This assumption is confirmed by results of kinetic measurements.

Kinetic curves of water sorption with composite investigated and massive salt are presented on Fig. 5. Considerable differences of water vapor sorption mechanism for composite sorbent 'silica gel-sodium acetate' and massive salts are revealed. Kinetic regime of massive salts hydration is collaborated with S-shaped forms of the kinetic curves in coordinate . On the contrary, initial regions of composite sorbents kinetic curves are nearly linear, being evidence of process diffusion nature, i.e. chemical reaction of water and disperse salts occurs rapidly, and water transport on silica gel pore system is rate determinant factor. Equilibrium for massive salts attains well after as compared with composites.

Thus, a rate of salt-water vapors reaction occurring in developed pores of silicon-oxygen matrix is enhanced by unique structure of synthesized composite sorbents.

Then testing of synthesized composite sorbent 'silica gel-sodium acetate' was carried out in the solar adsorptive chiller. The tests were realized from May to September 2014. Typical daily dependences of the temperatures of environment, absorber, transparent insulation, sorbent and temperature in the cold box for July are presented at the Fig. 6. Investigated solar adsorptive chiller can be divided conditionally into two modules. The first module of cold box is chilled due to water evaporation. The second one is a solar collector module where a sorbent regeneration occurs.

According to Fig. 6, temperature in the solar collector increases up to 50−60⁰C during daylight. A water removal is shown by investigation of the composite sorbent thermal properties to occur at the temperatures exceeded 60⁰C.

A sorbent regeneration is ascertained to be almost completed over a period of time from 10.00 to 14.00 irrespective of weather conditions. It is evidence of an adequacy of theoretical calculations and a good choice of the reserve coefficient of the solar collector.

Temperature in the cold box depends on adsorptive chiller operating cycle. However, it should
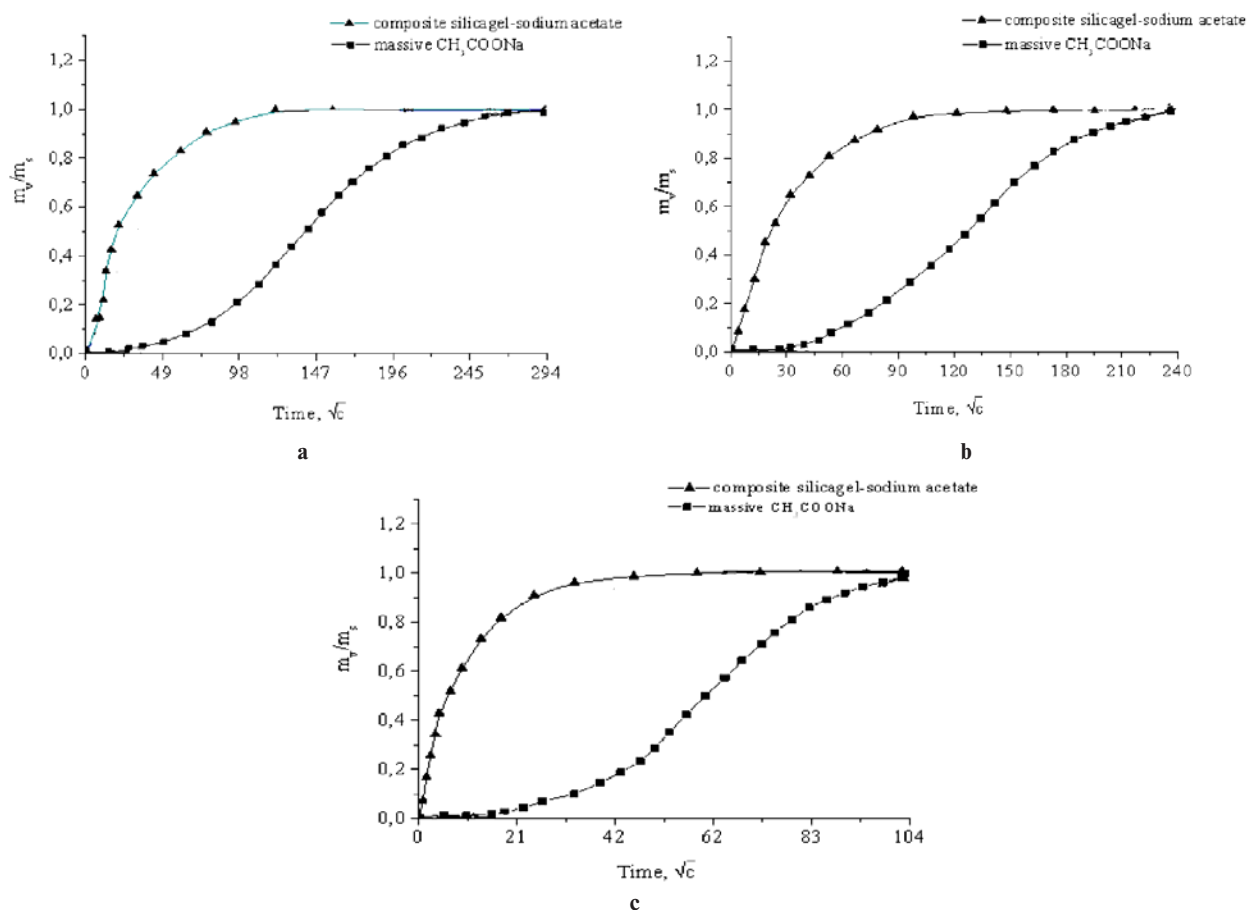

Fig. 5. Kinetic curves of water sorption (P_{H_2O} =1010,8 mbar), t ⁰C: a − 20; b − 40; c − 60

be noted that temperature in the cold box does not exceed 10⁰C at steady-state conditions.

Solar radiation intensity data are given in the Table 1. A collector optimal disposition is stated previously to be south facing at the slope of 30⁰ with horizon [15]. At that, radiation intensity absorbed by collector increases into 1.1−1.5 times depend on month.
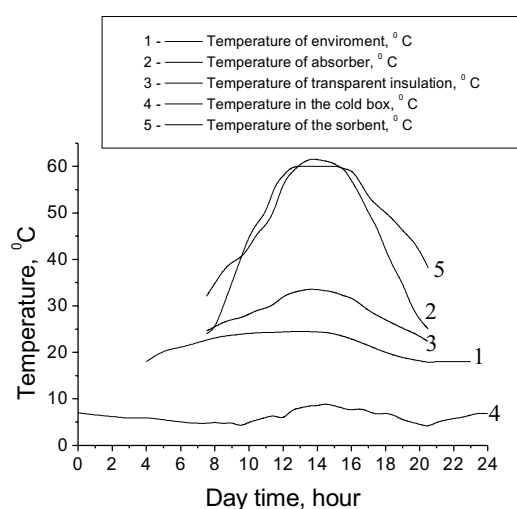

Fig. 6. Daily dependences of the temperatures of environment, absorber, transparent insulation, in the cold box and sorbent

Conclusions

Properties of composite sorbents 'silica gel-sodium acetate' synthesized by sol-gel method are investigated. Substantial difference of their sorption properties and silica gel and sodium acetate mechanical mixture is revealed. A qualitative difference between rate-determinative steps of processes of water vapor adsorption with 'silica gel-sodium acetate' and massive salt is stated. Changing of sorption properties of sodium acetate is shown to be caused by its dispersion up to nano-scale.

The solar adsorptive chiller used sorbents studied is developed. A solar collector is applied as heat source for sorbent regeneration. Temperature in the cold box is maintained to be 6−10⁰C in summer period.

Different methods of estimation of solar adsorptive chilling system efficiency were analyzed.

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