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Heat Transfer Enhancement of the Adsorber of an Adsorption Heat Pump

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Abstract. Three methods for improving the heat transfer of the adsorber have been developed in this paper. First, an electrically conductive polyaniline was applied for enhancing the thermal conductivity of adsorbent bed. A thermally conductive composite of polyaniline and adsorbent was prepared by chemical oxidative in situ polymerization of aniline onto the surface of adsorbent particles. A thin thermal conducting net on the surface of the adsorbent particles was grown. The experimental results indicated that the thermal conductivity of this composite could be increased to approximately 4 times that of the raw adsorbent. Second, the adsorbent bed was shaped by a compressing process. This process can reduce the thermal resistance among the adsorbent particles and the contact thermal resistance between the adsorbent bed and the heat exchanger. The thermal conductivity of the shaped adsorbent bed itself from the tests can be increased 30% when the density of the solid adsorbent bed is 1.5 times that of its original density. Furthermore, the adsorption capacity of the above treated adsorbent did not decrease obviously. Third, a proper design of adsorber has been introduced and analyzed. Further tests of this design will be conducted soon.

Keywords: adsorber, polyaniline, adsorbent bed-shaping, heat transfer enhancement

1. Introduction

The adsorption heat pump is a new energy-saving and environmentally friendly refrigeration and heating system. It is mainly composed of an adsorber, a condenser and an evaporator, where the adsorber containing the adsorbent replaces the compressor of the conventional vapor-compression heat pump. By selecting an appropriate adsorbent/adsorbate couple and using the low-grade thermal sources such as solar energy, natural gas and waste heat from industries, this closed system can perform the refrigeration or heating cycle (Meunier, 1979; Tchernev, 1979; Tan et al., 1992). Each cycle can be divided into two periods: heating the adsorber to release the adsorbate (equal to the vapor-exhausting process of conventional heat pump) and cooling the adsorbent to trap the adsorbate (equal to the vapor-compressing process of conventional heat pump) (Critoph, 1988, 1989; Critoph, 1992).

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The adsorber consisting of a heat exchanger and a suitable amount of microporous adsorbent is the main component of this kind of heat pump. In most of previous studies, the adsorber was a fixed bed structure formed by distributing the raw adsorbent particles into a heat exchanger. The heat transfer rate of the adsorber from inside to outside is low. It results a long cycle time and thus limits the commercialization of the adsorption heat pump (Zhu et al., 1993, 1996). The heat transfer resistance of the adsorber mainly comes from the solid adsorbent particles themselves, the contact resistance among particles and between the adsorbent fixed bed and heat exchanger. Some enhancement techniques have been introduced by many researchers (Guilleminot et al., 1990, 1992; Strauss, 1992; Cacciola et al., 1992; Groll, 1992). A comprehensive view on the state-of-the-art in these different enhancement techniques can be gathered from *the Proceedings of the Symposium on Solid Sorption Refrigeration* in Paris. In this paper, three methods for improving the heat transfer rate of adsorber have been developed. First, an electrically conductive polyaniline was applied for enhancing the thermal conductivity of adsorbent bed. Second, the adsorbent bed was shaped by a compressing process to reduce both the thermal resistance among the adsorbent particles and the contact thermal resistance between adsorbent bed and the heat exchanger. Third, a proposed new design of adsorber was introduced and analyzed.

2. Heat Transfer Enhancement of Adsorbent Bed by Using Polyaniline

2.1. Preparation of Polyaniline/Zeolite Composite

Polyaniline is an intrinsic high electrically conductive polymer. It has the advantages including simple preparation, nice stability, high electric conductivity, and so on. Some researchers have coated the polyaniline to the surface of some inorganic materials to obtain the electrically conductive additives (Wang, 1995). Based on these studies analogously, the polyaniline was first used to enhance the heat transfer of the adsorbent zeolite bed by us. A thermally conductive composite of polyaniline and adsorbent zeolite was prepared by chemical oxidative in situ polymerization of aniline onto the surface of adsorbent particles. A thin thermal conducting net on the surface of adsorbent particles was grown. The process of preparing the composite involves four materials which are aniline, zeolite 13X, HCl and $(NH_4)_2S_2O_8$. The best conditions of preparation could be found through the experiments reported in the article (Wang, 1997).

2.2. Characterization of the Thermal Conductivity

A special device based on the steady state flat principle was designed and constructed to measure the effective thermal conductivity of each sample. The schematic diagram of the experimental apparatus is shown in Fig. 1.

The adsorbent box was made of two flat copper plates and a circle-shaped frame made of insulation materials, which was placed between the plates to form a dam for the tested adsorbent. Two plates served as the heater and the cooler respectively. An infrared lamp served as the thermal source for the heater and the cooler was exposed to the air. Thus the temperature gradient in the axial direction can be obtained. The electric power provided to the infrared lamp could be adjusted by a transformer. The heat flow through the bed was measured with a heat flow ratio-measuring instrument directly. The temperature gradient in the bed was measured with the copper-constantan thermocouples and the values could be collected by a computer data-collecting system. The test rig was calibrated against a standard sample and less than 7% error was found.

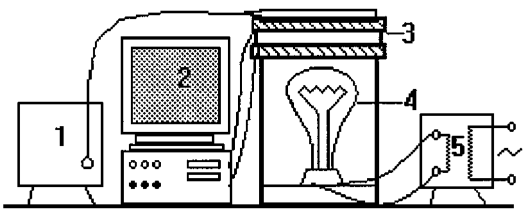

Figure 1. Schematic diagram of thermal conductivity-measuring system.

The method above is normally employed to measure the thermal conductivity of the insulation materials. Generally, if the length factor between the radial and the axial is no less than 7 and the periphery in the radial direction is insulated, the heat transfer in the tested sample is regarded as one dimension in the axial direction. Then the temperature gradient in the axial direction ΔT and the heat flow ratio q are recorded. The effective coefficient of the heat transfer could be calculated by the Eq. (1) based on the Fourier's law of thermal conductivity.

$$\lambda_e = \sigma \times q/\Delta T \tag{1}$$

The experiments were carried out under air and the adsorbents were in their natural density. Though the thermal conductivity of adsorbent varies with its water content, its value here was measured under air from the convenient viewpoint and enough to evaluate the enhancement results. The effective thermal conductivity λ_e was plotted as the function of the average temperature of the tested adsorbent bed T as shown in Fig. 2. It can be seen that the thermal conductivity of the treated adsorbents have been significantly increased compared to the raw zeolite adsorbent.

For the sake of quantitative considerations regarding of the thermal conductivity of each sample, the average effective thermal conductivity λ_a under the conditions between the adsorption temperature of 40°C and the desorption temperature of 150°C was defined. The thermal conductivity and the physical characteristics of each sample were given in Table 1.

The thickness of the polyaniline net on the surface of the zeolite was determined by the mass factor of the polyaniline in the composition pellet, the density of the pure polyaniline and the parameters of the raw

Table 1. The average thermal conductivity λ_a of polyaniline/zeolite composites.

Sample number	Raw zeolite	1#	2#	3#	4#	5#	6#	7#	8#	9#
Particle porosity	0.32	0.30	0.29	0.29	0.30	0.30	0.30	0.30	0.30	0.29
Diameter (mm)	1.80	1.90	1.90	1.90	1.90	1.90	1.90	1.90	1.90	1.90
Polyaniline/zeolite (wt%)	0	12.84	14.32	15.68	13.11	16.02	13.06	14.86	14.27	13.02
λ_a (W/m · °C)	0.056	0.162	0.204	0.235	0.179	0.246	0.177	0.217	0.192	0.178

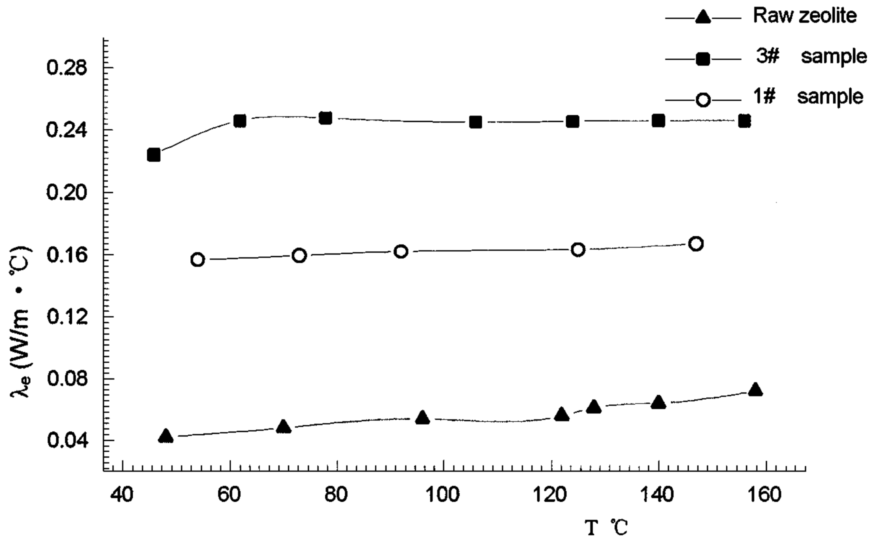

Figure 2. The thermal conductivity values λ_e as a function of temperature T.

zeolite pellet. Though the mass factor between zeolite and polyaniline of each sample was different, the thickness of the net was very small and the value of all composition samples in our study was regarded as about 0.05 mm. Table 1 showed that the average effective thermal conductivity of the composites could be increased 2–4 times that of the raw zeolite adsorbent.

2.3. *Characterization of the Adsorption Capacity*

The adsorbents are expected to have not only a good thermal conductivity but also a good adsorptive character with the corresponding adsorbate. The adsorption isotherms for water in composites and raw zeolite were measured gravimetrically at various temperatures. A rig as shown in Fig. 3 has been built.

A small glass container filled with the measured sample was hung by a quartz spring which has a sensitivity of 0.05 mm/mg. The length of the quartz spring which can be measured by a high-measuring instrument represents the relative weight of the container. A constant

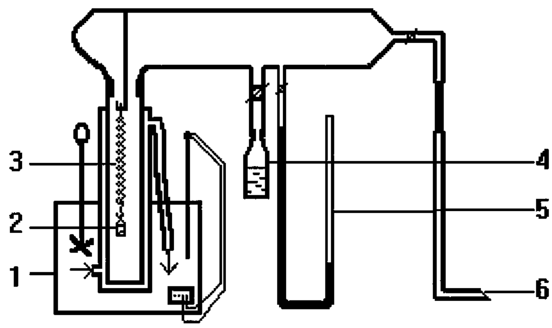

Figure 3. Schematic diagram of adsorption capacity-measuring system.

sample weight indicated the adsorption equilibrium. A heated bath controlled the adsorption temperature. A vacuum system was used to obtained a vacuum <1 Pa for this system. The pressure of the system was shown by a mercury manometer. When the desired adsorption temperature was controlled, the relation of the amount adsorbed and the vapor pressure in the adsorption

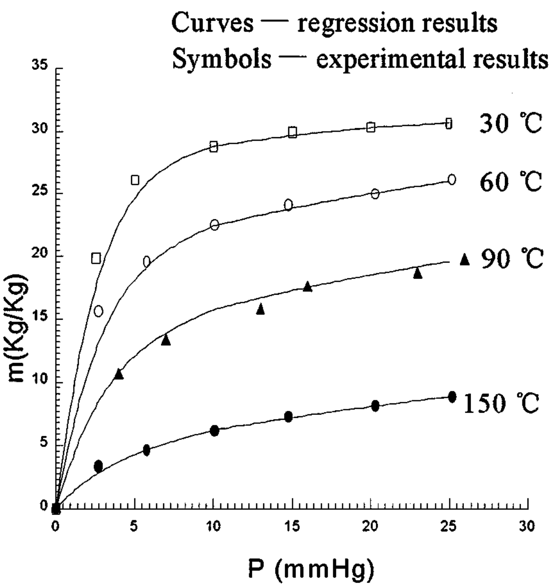

Figure 4. The curves of the adsorption isotherms for the 13X zeolite pellet (the diameter of 1.80 mm).

equilibrium state could be measured. The measurement was made at four different adsorption temperatures.

The adsorption isotherms for water in the 13X zeolite pellets with the diameter of 1.80 mm and the nine types of sample pellets with the diameter of about 1.90 mm were measured. A typical group of the symbol curves for one sample (the 13X zeolite pellets) was plotted in Fig. 4. The equilibrium data were correlated by the model of the micropore volume filling theory, which was adopted by other researchers (Critoph, 1988, 1989; Critoph, 1992). The equation of this theory was described as Eq. (2).

$$\ln m = \ln \rho V_0 - K(T \ln P_0/P)^n \qquad (2)$$

The resulting solid curves calculated from the Eq. (2) were also shown in Fig. 4. The model seems to give agreement with the measurements. To a given sample, the V_0, K and n are constants and can be calculated from regression Eq. (2) by using the least-square method and the experimental data. To assess the adsorption capacity of each sample quantitatively, the difference of the amount adsorbed ($\Delta m = m_{\text{ads}} - m_{\text{des}}$) between the adsorption state and the desorption state in a practical cycle of the adsorption heat pump was defined and calculated from the adsorption Eq. (2). Four levels of temperature adopted in the calculation were the adsorption temperature of 40°C, the evaporation temperature of 10°C (corresponding to the adsorption pressure), the desorption temperature of 150°C, the condensation temperature of 40°C (corresponding to the desorption pressure). The calculated results were shown in Table 2.

Table 2 showed that the values of $V_0, K, \Delta m$ for composites were slightly smaller than those of the raw zeolite. However, the value of n was increased somewhat. It can also be seen that the differences of all values are within 10%. The reason for the decrease of V_0 may be that a little amount of cavities of composites were blocked up by the polyaniline. But the polar molecule of polyaniline can also change the characteristics of the surface of the adsorbent. This led to the increase of n corresponding to the number of molecule adsorbed on sites and decrease of K related with the characteristic adsorption energy E $[K = (R/E)^n]$. For these reasons, conclusion could be drawn that the Δm of the composites decreased unobviously.

2.4. *Comparisons of Several Methods for Enhancing Heat Transfer*

In order to improve the heat transfer of the adsorbent beds, many methods have been attempted. Critoph uses

Table 2. The results of regression.

	Raw zeolite (13X)	Sample (1#)	Sample (2#)	Sample (3#)	Sample (4#)	Sample (5#)	Sample (6#)	Sample (7#)	Sample (8#)	Sample (9#)
V_0	0.310	0.307	0.295	0.291	0.303	0.296	0.305	0.298	0.302	0.293
n	1.58	1.63	1.73	1.80	1.61	1.74	1.66	1.72	1.69	1.79
$k \times 10^{-6}$	6.578	4.378	1.926	1.041	5.187	1.739	3.408	2.113	2.695	1.132
E	15.83	16.12	16.73	17.52	15.94	16.98	16.35	16.57	16.42	17.45
Δm	14.837	14.687	13.987	13.382	14.544	13.882	14.533	14.217	14.431	13.507
e (%)	0	−1.01	−5.73	−9.81	−1.97	−6.44	−2.05	−4.18	−2.74	−8.96

forced convection between the adsorbent granules and refrigerant itself to achieve high heat transfer rates within the bed. The additives such as graphite and metallic powder have also been adopted to intensify the heat transfer of adsorbent bed by many researchers. However, by adding a fixed amount of heat conduction power into the pure zeolite beds simply, the results may be not satisfying. Most of researchers' interests have been focused on introducing a heat conduction matrixes such as Cu foam and gas flow channels into the zeolite beds and then consolidating the beds. A good heat transfer adsorbent bed has been obtained. By these means, the mass factor between the metal and adsorbent must be increased significantly, which is not beneficial to increase the *COP* of the adsorption heat pump. In the meantime, it is difficult to get highly porous (≥90%) metal foams though they are designed by some researchers. Guilleminot dispersed a mount of the copper powder into the NaX zeolite adsorbent bed. The thermal conductivity of the treated bed is 0.17 W/m · °C while the value of the pure zeolite bed is 0.09 W/m · °C. Furthermore, he reported that the heat conductivity of a consolidated material made of copper metallic foam and zeolite was enhanced by a factor closed to 100 (8.3 W/m · °C instead of 0.09 W/m · °C). Based on our experiments, the best mass factor of the composition between the polyaniline and the dry zeolite is 16.11 : 83.89. Consequently, this mass factor was adopted for preparing the samples of aluminum powder/zeolite, graphite powder/zeolite and copper powder/zeolite. These samples were also tested with our testing rigs and the experimental results were given in Table 3.

Our testing results indicated that the thermal conductivity of the raw zeolite, copper powder/zeolite, polyaniline/zeolite is 0.056 W/m · °C, 0.124 W/m · °C, 0.258 W/m · °C respectively. The enhancement factor of the polyaniline/zeolite is 4.6 while the value of the copper powder/zeolite is 2.2. Table 3 also indicated that all of the four methods could cause the decrease of adsorption capacity and the increase of thermal conductivity. However, a small amount of additives dispersed into the adsorption medium by a mechanically mixing process could not form a continuous thermally conductive phase. But a small amount of polyaniline coating to the surface of adsorbent directly by a chemically adsorbent bed (In our study, the particle bed is not consolidated. Further study will be focused on the shaping process of the particle bed and the design of the heat exchanger which was briefly introduced in this paper. This treating process can reduce the contact resistance significantly.). So the same amount of 16.11% polyaniline by weight could obtain a nice result of enhancing the adsorbent bed.

The physical model of the thermal conductivity of the polyaniline/zeolite composite can be presented in Fig. 5 simply. Electrical circuits offer a helpful analogy in analyzing it.

The zeolite particle was simplified as a square and its thermal resistance was numbered 2. The polyaniline net over the surface of the zeolite particle was divided into four parts represented by four rectangles with the width equal to the thickness of the polyaniline net and the length equal to the side width of the zeolite particle square. Their thermal resistance was numbered 1, 3, 4, 5 respectively. Supposed that the side width of the

Table 3. Comparisons of several methods.

Methods	Raw zeolite pellet (ϕ1.80)	Aluminum powder (zeolite)	Graphite powder (zeolite)	Copper powder (zeolite)	Polyaniline net (zeolite)
Additive/adsorbent (wt%)	0	16.11	16.11	16.11	16.11
Δm (kg/kg)	14.837	13.531	13.864	13.836	13.917
λ_a (W/m · °C)	0.056	0.092	0.102	0.124	0.258

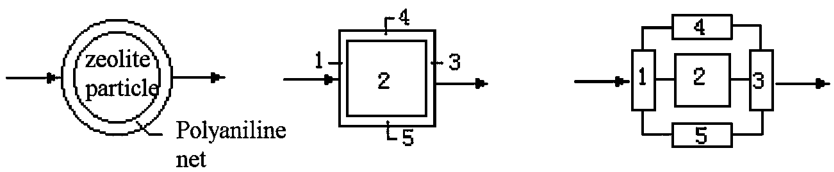

Figure 5. Schematic drawing of thermal conductivity of the polyaniline/zeolite composite.

zeolite particle square 2 be equal to 1 and the thickness of the polyaniline net be equal to h, the following Eq. (3) can be obtained

$$R_2 = \frac{1}{\lambda_s}, \quad R_1 = R_3 = \frac{h}{\lambda_j}, \quad R_4 = R_5 = \frac{1}{\lambda_j h} \qquad (3)$$

Then the total conductivity of the composite can be expressed as the Eq. (4).

$$\lambda_e = \frac{(4h^2+1)\lambda_j\lambda_s + 2h\lambda_j^2}{2h\lambda_s + \lambda_j} \qquad (4)$$

Because of $\lambda_s \ll \lambda_j$, the Eq. (4) can be simplified as the Eq. (5).

$$\lambda_e = \lambda_s + 2h\lambda_j \qquad (5)$$

In case of $\lambda_s \ll \lambda_j$ in the Eq. (5), then a small thickness h of the polyaniline net can also increase the thermal conductivity of the zeolite particle efficiently. This is the main reason that we employed the polyaniline as thermal conductive material to coat the surface of the zeolite particle for enhancing its thermal conductivity.

3. Heat Transfer Enhancement by Shaping Adsorbent Bed

3.1. Preparation of the Shaping Adsorbent Bed

By compressing the adsorbent bed to shape it, the density of the adsorbent bed can be increased and the thermal resistance among the adsorbent particles and the contact thermal resistance between the adsorbent bed and the heat exchanger can be reduced. In addition, the weight of filled adsorbent per volume of the adsorber can be increased and the life of adsorbent can also be prolonged. These seem even more important to an adsorption heat pump.

The shaped adsorbent bed can be prepared by consolidating or sintering processes (Groll, 1992). In this study, a suitable proportion of adsorbent, water, clay was mixed up and compressed with a self-made apparatus. Then it was put into a vacuum oven and desiccated at the temperature of 110°C for 24 h. The shaped bed with a definite geometry is tubular, as shown in Fig. 6. The density of adsorbent bed can be adjusted by changing the weight of the adsorbent fed into the module.

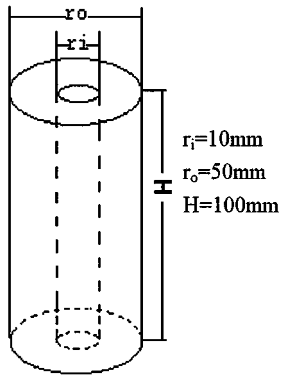

Figure 6. The geometry of the shaped bed.

3.2. Thermal Conductivity Measurements of the Shaped Adsorbent Bed

Small-scale equipment simulating a practical adsorption heat pump was built for studying the effective thermal conductivity of the shaped adsorbent bed, as shown in Fig. 7.

The thermal source was offered by an inserted electric heating rod. A dynamometer measured the power supplied for the rod in the bed. The methods for measuring the temperature and the pressure were the same as Fig. 1. The effective thermal conductivity of the bed can be calculated by the following Eq. (6).

$$\lambda_e = Q \times \ln(r_o/r_i)/(\Delta T \times 2\pi \times H) \qquad (6)$$

The effective thermal conductivity (λ_e) obtained from the experiments at different density of the bed were plotted in Fig. 8. It can be seen that the effective thermal conductivity of the shaped bed raised with the increase of the density of the adsorbent bed linearly. However, the adsorption capacity of the shaped bed must be considered in the same time, the gains to be

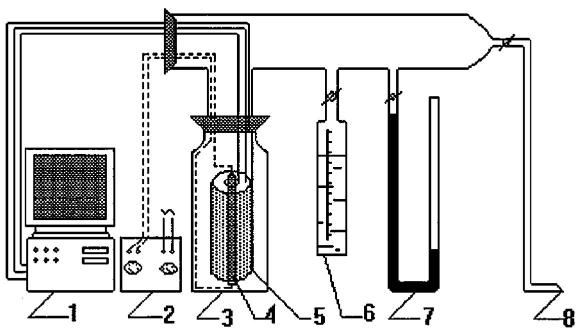

Figure 7. Schematic drawing of the adsorption heat pump system.

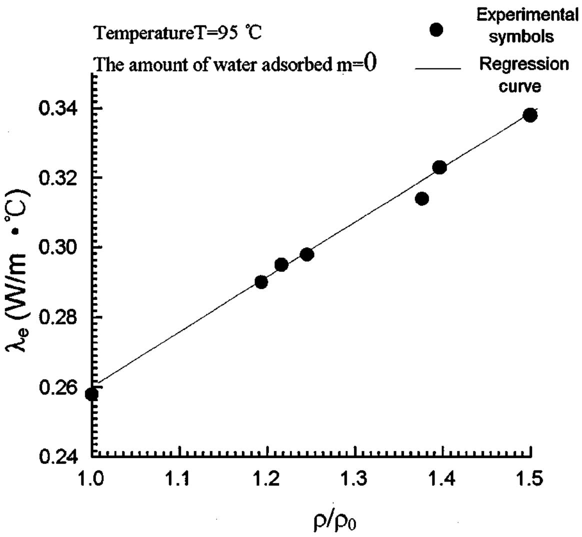

Figure 8. The effective thermal conductivity λ_e as a function of the relative density of the bed ρ/ρ_0.

made in trying to obtain very high values of the density are comparatively limited. The effective thermal conductivity of the shaped adsorbent bed itself from this experimental tests could be increased 30% when the density of the solid adsorbent bed was 1.5 times of its original density. Furthermore, the adsorption capacity of a small part of it was measured with the rig of Fig. 3. The results indicated that the adsorption capacity didn't decrease obviously. The reason may be that the characteristics of microposity in the adsorbent corresponding to its adsorption capacity has not changed during the shaping period. If the macroporosity between the particles which doesn't contribute to adsorption capacity is given enough space for the adsorbate molecular to move in the bed, it could be decreased to an optical value by a compressing process. The optimal conditions of the shaping process are investigated now.

4. Heat Transfer Enhancement by Optimizing Design of the Adsorber

4.1. A Design of the Adsorber

The design of the heat exchanger depends on the shape of the adsorbent bed. A design of the adsorber corresponding to the tubular-shaped bed was proposed by the authors, as shown in Fig. 9. It is a shell and tubular structure. The tubes in the adsorber are in a coupling configuration because the shaping adsorption bed is cased to the outside of a metallic tube. The heating and cold fluid alternatively flow in the tube side, while the adsorbate vapor flow through the shell side.

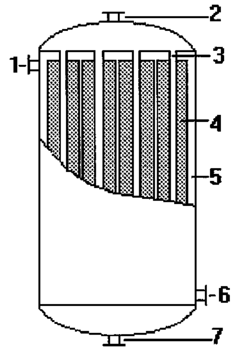

Figure 9. Structure of a new design adsorber.

4.2. Analysis of the Adsorber

For the purpose of improving heat and mass transfer to and from the adsorber, a major development area must lie not only for the heat transfer enhancement of adsorbent bed but also in the design of adsorber itself. The heat exchanger must have large heat transfer area as possible as it could be. Old adsorption heat pumps all use fins to improve heat transfer, but this is at the expense of increasing the thermal capacity of the adsorber significantly. The heat contacting surface of this adsorber is three dimensional. So the heat transfer areas per weight of the heat exchanger is relatively high and quite effective for heat transfer. In addition, the problem of swelling and efflorescence on filled adsorbent bed can be adminished to a small degree. The structure of the adsorber is simple, compact, easily produced and maintained.

For a practical equipment design, the adsorption properties, environmental conditions, equipment characteristics and the parameters of adsorber must taken into account and optimized. In this sense, the design proposed here is not a perfect one and is to be considered just as an idea.

5. Conclusions

1. An electrically conductive polyaniline was applied for enhancing the thermal conductivity of adsorbent bed by a chemical polymerization process. The testing rigs were designed and built to measure the thermal conductivity and adsorption capacity of the adsorbents. The experimental results indicated the thermal conductivity of this composite could be increased to approximately 4 times that of raw adsorbent and the adsorption capacity decrease did not obviously. The results of this technique were compared with other measures for enhancing thermal

conductivity of adsorbent and found that this technique is a nice one.

2. The adsorbent bed was shaped by a compressing process. This process can enhance the heat transfer of the adsorbent bed by increasing the weight of the filled adsorbent per volume of adsorber. The thermal conductivity of the shaped adsorbent bed can be increased 30% when the density of the solid adsorbent bed is increased 150%.
3. A new design idea of adsorber has been introduced and analyzed. Further work should be conducted to optimize it both by considering heat and mass transfer enhancement and minimizing the cost.

Nomenclature

λ_e	conductivity of zeolite/polyaniline composite	$W/m \cdot {}^\circ C$
λ_a	the effective thermal conductivity	$W/m \cdot {}^\circ C$
σ	the length of heat flow	m
q	the heat flow ratio	$J/m^2 \cdot s$
ΔT	the difference of temperature	$^\circ C$
m	the amount adsorbed in equilibrium	kg/kg
T	adsorption temperature	$^\circ C$
P	vapor pressure of system	Pa
V_0	the limiting volumes of adsorption space	m^3
K	the adsorption parameter	—
n	the distribution parameter	—
P_s	the saturation vapor pressure	Pa
E	the characteristic energy	J
λ_j	the thermal conductivity of polyaniline	$W/m \cdot {}^\circ C$
h	thickness of polyaniline net	m
R	thermal resistance	$m^2 \cdot K/W$
Q	heat flow through the bed	J/s
r_i	the insider diameter of shaped bed	m
r_o	the outer diameter of shaped bed	m
H	the height of shaped bed	m

Acknowledgments

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