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# REVIEW OF ADSORPTION AND THERMAL CHARACTERISTICS OF ACTIVATED CARBON AND ITS APPLICATION IN ANG STORAGE AND ACS SYSTEMS

**Abstract.** Porous materials act like a sponge for different types of gases. This property of porous materials is widely used in adsorbed gas storage (ANG) and cooling systems (ACS). Adsorption based gas storage has a lot of advantages, it allows to store almost the same amount of gas at more than 6 times lower pressure compared to compression storage and there is no need on expensive compression equipment for preliminary multi-stage compression. Adsorption principle is also used in cooling systems. It performs the function of compressor to convert the refrigerant from gas state back to a liquid state. Adsorption cooling system does not require electricity for operation, this makes possible to use ACS in remote underdeveloped regions. But ACS systems are bulky and have low specific cooling power, which limits its wide use and propagation. In this paper a comprehensive review about working principles of cooling systems and on methods of gas storage using adsorption principle was done. Terminology, temperature effects, adsorption characteristics of an activated carbon which is considered as the most popular adsorbent were investigated and given below.

**Keywords:** adsorption, activated carbon, adsorption cooling systems (ACS), adsorbed natural gas (ANG), storage.

**1. APPLICATION IN COOLING SYSTEMS.** According to International Institute of Refrigeration (IIR), approximately 15% of all electricity produced worldwide is used for refrigeration and air conditioning [5, 14]. There are two types of cooling systems: a) sorption cooling systems and b) vapor compression cooling systems. In the Table 1 the main pros and cons of each of them are illustrated.

Working principle of both adsorption and compression cooling systems lies in the use of a refrigerant with a very low boiling point (less than -18 $^0$C). Refrigerants after taking the heat of the surrounding start boiling and evaporating. Evaporated particles of the refrigerant take some heat away with them, thereby provide cooling effect. The main difference between these two systems is the way how the refrigerant is changed from a gas state back to a liquid state so that the cycle could repeat.

In adsorption cooling systems gaseous refrigerant is *absorbed by another material*, and the temperature of refrigerant-saturated material increases, which leads to the refrigerants to evaporate out. Hot gaseous evaporated refrigerants pass through a heat exchanger, where they give their thermal energy outside the system and condense. After all, condensed refrigerant go to the initial compartment, where it starts its next cycle. Whereas in compression cooling systems gaseous refrigerant passes through compressor which increases its temperature above surrounding temperature, to assure the refrigerant to give away its thermal energy to the environment.

Adsorption refrigerators need only heat so they can function (utilizes solar or low grade waste heat which is in excess in power plants and automobile engines), and have no moving parts except refrigerant. It is a fully thermally activated refrigeration system. Whereas, a compressor refrigerator requires electrical or mechanical energy for operation (it uses an electrically powered compressor).

**Working pairs for ACS.** Working pair (adsorbent and adsorbate) is the most important element of any ACS. The performance of ACS depends on selection of working pair and on their thermal and adsorption properties. Other properties such as latent heat, freezing point and saturation vapor pressure,

Table 1 – Comparison of sorption and compression cooling systems

| | Advantages | Disadvantages |
|---|---|---|
| Adsorption cooling systems (ACS) | a) utilize natural and benign refrigerants such as water, methanol, ethanol, ammonia, CO2, R1234ze, etc.<br>b) zero global warming potential<br>c) can be driven with solar energy or waste heat which is abundant in summer when the cooling power is the most needed<br>d) can operate off-grid, autonomously<br>e) low operating cost<br>f) simplicity of construction, lack of moving parts<br>g) simple control<br>h) quiet operation, no vibration | a) bulkiness<br>b) higher costs<br>c) low performance |
| Vapor compression cooling systems | a) compact | a) employs high global warming refrigerants such as R134a (GWP = 1430) for mobile air conditioning and R410a (GWP = 1725) for residential air-conditioning (chlorofluorocarbons (CFCs) or Hydrofluorocarbons (HFCs))<br>b) stresses electric grids in summer<br>c) vibration problems, noise pollution |

Table 2 – Working pairs and their characteristics [5, 14]

| Adsorbent | Adsorbate (refrigerant) | Heat of adsorption (kJ/kg) | Evaporation temperature ($^{o}$C) | COP |
|---|---|---|---|---|
| Silica gel | Water | 2800 | 10 $^{o}$C | 0.4 |
| | Methyl alcohol | 1000-1500 | | |
| Zeolite (various grades) | Water | 3300-4200 | 5 $^{o}$C | 0.9 |
| | Carbon dioxide | 800-1000 | | |
| | Methanol | 2300-2600 | | |
| | Ammonia | 4000-6000 | | |
| Activated alumina | Water | 3000 | | |
| | Ethane | 1000-2000 | | |
| | Ethanol | 1200-1400 | | |
| Activated carbon | Methanol | 1800-2000 | | 0.12 [18] |
| | Water | 2300-2600 | | |
| | Ammonia | 2000-2700 | 3 $^{o}$C | 0.67 |
| | Carbon dioxide ($CO_2$) [12] | | | |
| Calcium Chloride | Methanol | | | |

Table 3 – Ways of overcoming the drawbacks of adsorption cooling systems

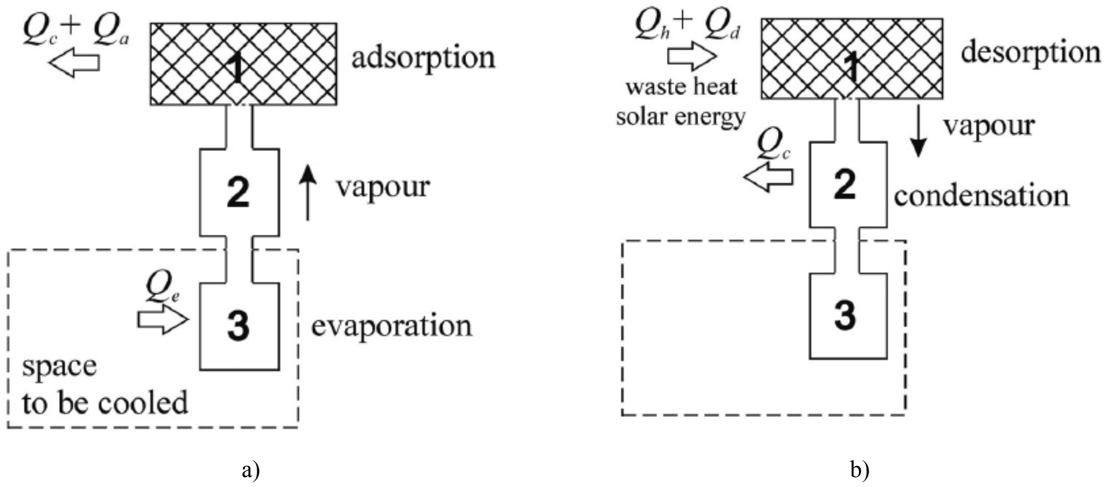
| № | Methods | |
|---|---|---|
| 1 | Presenting new adsorbents | a) Specific heat capacity [1,4]<br>b) Density [1,4]<br>c) Thermal conductivity [1,4]<br>d) With higher sorption rate [5] |
| 2 | Presenting new refrigerants | |
| 3 | Employing new heat sources | |
| 4 | Improving heat transfer coefficients | a) Increasing the number of fins in finned tube heat exchangers [1,4]<br>b) Consolidating the adsorbent [1,4]<br>c) Increasing heat transfer area of adsorber bed i.e., design of new adsorber bed [5] |
| 5 | Decreasing the driving temperature | |
| 6 | Using heat recovery | |

toxicity, flammability, corrosion, etc., also have same importance while selecting the working pair. The common working pairs and their characteristics are illustrated in the Table 2 below. Depending on the freezing and boiling points of refrigerants, working pairs can be applied for different purposes: refrigeration, ice making, air conditioning, engine chilling etc.

Adsorption cooling systems are good for application in remote locations, where electrical resources are limited, because they can operate off-grid, autonomously. But this system suffers with bulkiness (of mass and volume) and low specific cooling power. In Table 3 the ways of making sorption cooling systems more compact and productive are illustrated [1-5].

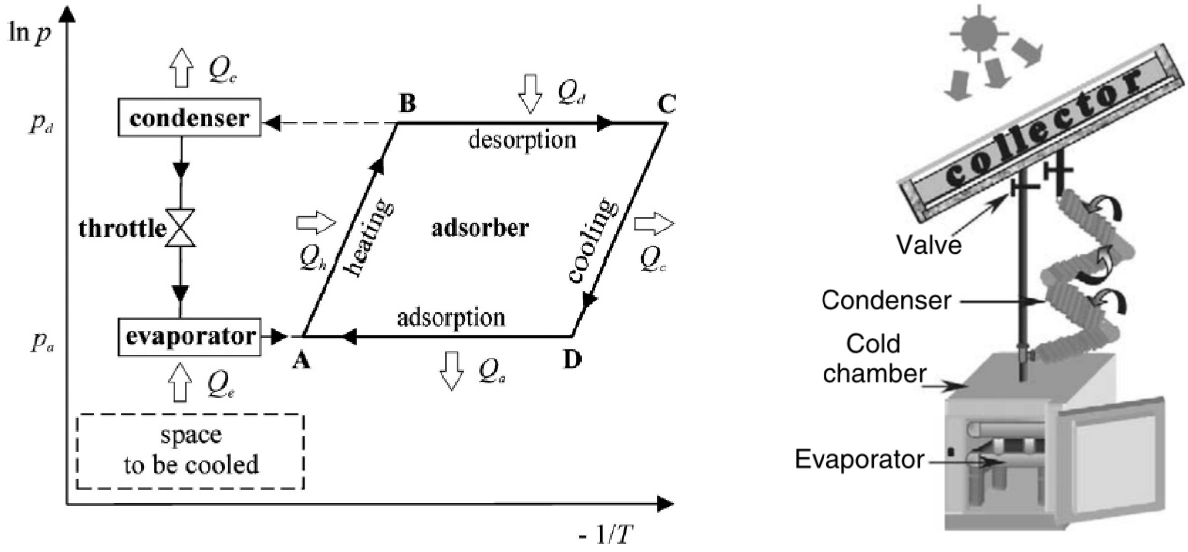
**Operating principle.** There are two main types of adsorbents: *stationary (solid)* and *dynamic (liquid)*. Adsorption systems for each of them are built differently. In ACS with stationary absorbent only the adsorbate circulates. When in ACS with dynamic adsorbent both the adsorbate and adsorbent circulate. For example, in ammonia/water ACS the water performs the function of an adsorbent, whereas in water/activated carbon pair it is an adsorbate. In this work only operation principle of ACS with solid adsorbent is considered. Activated carbon, silica gel and zeolite are related to solid adsorbents. In case of usage of solid adsorbent, the system operates intermittently. To make it work continuously two or more adsorbent beds are required. In each of these adsorbent beds adsorption and desorption occur alternately [14, 15, 17]. Fig.1 and Fig.2b illustrate the schematics of one-bed adsorption chiller, where 1 – collector/generator/adsorber, 2 – condenser and 3 – evaporator. Adsorbate, which is initially in the evaporation unit, takes the heat of the environment being cooled, $Q_{evap}$, and evaporates out. In Fig.1a it is shown the adsorption step which includes 1) *pre-cooling* of the bed with cooling water leading to the decrease in temperature and pressure in the bed (see Fig.2, section $C \rightarrow D$) and 2) *adsorption* of evaporated gas at constant adsorption pressure, $p_a$ (section $D \rightarrow A$). During this step the heat of adsorption $Q_a$ and cooling $Q_c$ are emitted to the environment. And at point A, at the end of this step, the adsorption bed is saturated with adsorbate. Fig.1b illustrates the desorption step that includes 3) *pre-heating* of the bed with waste heat to increase the temperature and pressure up to condensation pressure (section $A \rightarrow B$) and 4) *desorption* during which adsorbent is removed from the bed and flows into the condensers, where it gives some energy $Q_{cond}$ to the surrounding, condenses and flows into the reservoir (section $B \rightarrow C$). During this step the heat of desorption $Q_d$ and waste heat $Q_h$ are taken by the adsorption bed. At point $C$ the adsorbent bed is fully regenerated and cycle repeats.

In solar solid ACS an activated carbon is heated during the day and cooled at night [16]. As we see, one-bed adsorption chiller does not chill continuously, but this disadvantage of ACS can be solved by adding few more adsorption beds. Examples of continuous systems can be found in [19, 20]. Through multi-bed operation the COP might be improved.




a) b)

Figure 1 – Working principle of one-bed solid ACS: a) adsorption step, b) desorption step [14]




a) b)

Figure 2 – Thermodynamic analysis of ACS:
a) the ideal cycle in the clapeyron diagram [14]; b) main compartments of simple ACS [16]

**P-T-W (pressure-temperature-concentration) diagram.** For thermodynamic analysis of working pairs the P-T-W diagrams are used. It shows the relation among pressure, adsorption temperature and adsorption uptake [16]. For example the PTW diagram of a carbon based composite adsorbent/$CO_2$ pair investigated in [12] is shown in Fig.3. The diagonal lines correspond to the adsorption uptake. In this case the refrigerant concentration in the adsorption bed varies between 0.6 and 0.93.

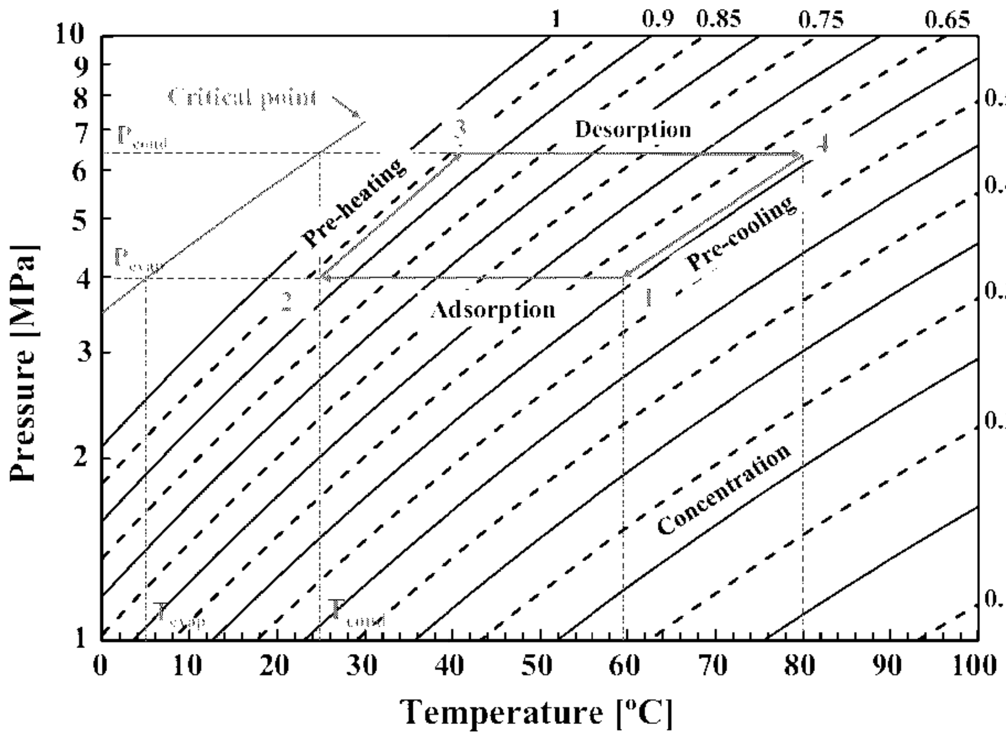



Figure 3 – Example of PTW diagram: composite adsorbent/$CO_2$ [12]

**Coefficient of performance (COP).** COP is the ratio of the heat extracted by the refrigerant evaporation $Q_e$ (cooling energy) to the waste heat amount $Q_h$ received by the adsorption bed. In solar ACS, $Q_h$ is the solar irradiation received by the area of the collector. For example, in [16] the cooling performance of solar powered adsorption refrigerator (with an activated carbon/methanol working pair) was tested during 14 days. During this period the daily mean ambient temperature varied between 14-18 $^0$C. Depending on the weather the irradiation received by the collector varied from 12 000 to 27 000 kJ/m$^2$. The temperature achieved in the cooling box by the evaporation varied between 8.1 $^0$C and -5.6 $^0$C. Thereby the COP of the refrigeration unit is found to be 5-8%.

**2. APPLICATION IN GAS STORAGE.** Activated carbons are used in natural gas and hydrogen storage. Porous materials act like a sponge for different types of gases. The gas may then be desorbed/extracted when subjected to higher temperatures. Gas storage in activated carbons is a promising method because the gas can be stored in a low pressure, low mass, low volume environment that would be much more feasible than storage in bulky compression tanks (see Table 4) [6]. Here the storage capacity is expressed in terms of volume per volume (v/v), the volume of stored gas at standard temperature and pressure per volume of storage space.

Table 4 – Comparison of Compression and Adsorption natural gas storage [9]

| | CNG Tanks (Compressed Natural Gas) | ANG Tanks (Adsorbed Natural Gas) |
|---|---|---|
| Material | Heavy wall cylindrical steel tank | Extruded aluminum tank filled with carbon monolith |
| Storage at pressures | $> 200\ atmos\ (3000\ psi\ or\ 21\ MPa)$ | 34.54 atmos (500 psi or 3.5 MPa) |
| Cost and energy consumption during charging | Expensive 4 stage compression needed using ~15% energy of the gas [9].<br><br>1.65 MJ/kg energy is consumed during filling the CNG tank from 1bar to 200 bar (197.4 atmos) [13] | Single-stage compression [11]<br><br>0.86 MJ/kg [13] |
| Storage capabilities | Store/deliver ~220 − 240 v/v based on internal volume. No consideration of wall thickness or envelop box.<br><br>Internal volume is ~70% of envelope, so storage is really about 160 v/v. | Store 185 v/v<br>Deliver ~150 v/v<br><br>(Storage capacity is always greater than the delivered capacity, by around 15%, sometimes may reach 30% [10]) |
| Disadvantages | | 1. Impurities in natural gas can block the micropores and over many charging/discharging cycles may result in decrease in storage capacity.<br>2. Needs thermal regulation |
| | ANG at 1/6 the pressure store 85%, deliver 70% that of CNG | |

Figure 4 –
ANG versus CNG on $CH_4$ delivery [9]

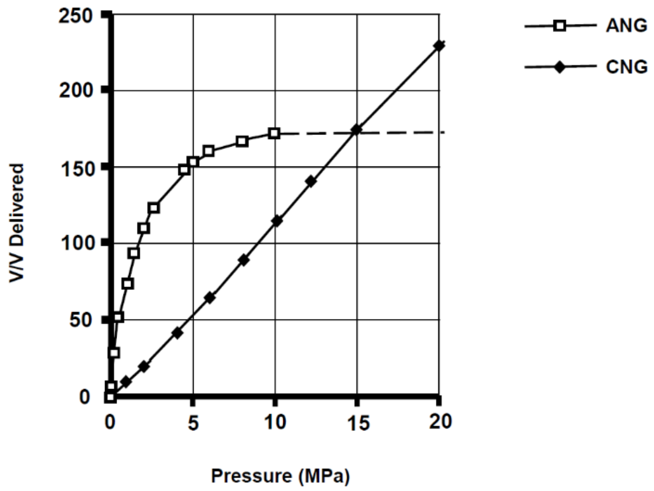


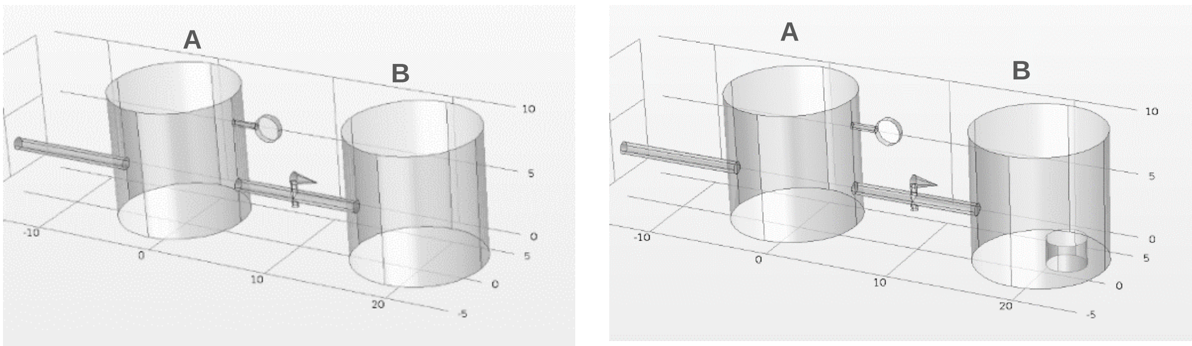
Fig.4 illustrates the storage capacities of ANG and CNG at different pressures. ANG delivers 3 times the volume of CNG at 5 MPa. At ~10MPa ANG reaches plato. CNG at 20MPa delivers ~30% more gas than ANG at that pressure.

**3. CONCEPTS RELATED TO ADSORBENTS.** There are two types of adsorption: physical and chemical. During chemical adsorption the adsorbate and adsorbent form a new type of molecules and these molecules are decomposed during desorption process. Whereas during physical adsorption there is no any new molecule synthesis, and it happens due to weak van der Walls forces between adsorbent and adsorbate molecules [5]. For adsorption cooling most refrigerant molecules are nonpolar molecular gases like ammonia, methanol, ethanol and other hydrocarbons that can be *absorbed physically* by activated carbon, zeolite and silica gel.

**Adsorbed gas amount.** Gas Law states that the products of volume and pressure stay constant for given amount of mass at fixed temperature. If to consider two chambers connected with a valved pathway (Fig.5a), the product of pressure and volume of a gas in chamber A, when the valve is closed, is equal to the product of a decreased pressure and an expanded volume after opening the valve.

$$\text{Total moles of gas} \propto PV$$

$$P_A V_A = P_{AB} (V_A + V_B)$$




a) b)

Figure 5 – Schematics of experimental set-up for the adsorbed gas amount measurement

Now if to put some porous material into the chamber B (Fig.5b), for example an activated carbon, the products of pressure and volume before and after opening the valve will be no longer equal, because molecules adsorb onto surface of material. The amount of adsorbed gas is found by subtracting initial and resulting products of volume and pressure. This principle is used in finding adsorption characteristics of materials [9].

$$P_A V_A > P_{AB} (V_A + V_B)$$

$$\text{Adsorbed gas amount} \propto P_A V_A - P_{AB} (V_A + V_B)$$

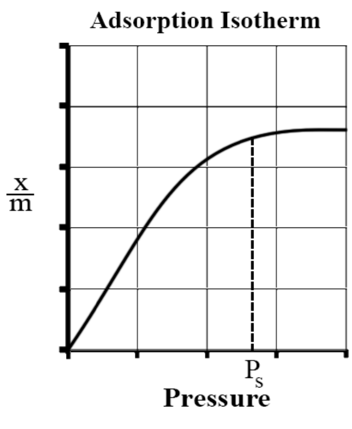



Figure 6 – Basic Adsorption Isotherm [8]

**Adsorption isotherm.** Adsorption process is usually studied through adsorption isotherm. It is the graph of dependency of adsorption uptake on pressure at constant temperature. In Fig.6, *x* is the amount of adsorbed adsorbate and *m* is the amount of adsorbent. So the adsorption uptake is the ratio of grams adsorbate to gram adsorbent. Saturation pressure $P_S$ is the value of pressure after which adsorption does not occur anymore.

**Apparent and skeletal densities.** Porous materials have two kind of density, apparent (or bulk density) and true (or skeletal density). *True (or skeletal) density* can be measured by filling the sample with helium since it will easily penetrate all the pores (up to 2 Angstrom). At the same time, He will not be adsorbed in the material. The equipment is named pycnometer. The simplest type of gas pycnometer (see Fig.7) consists of two chambers, one to hold

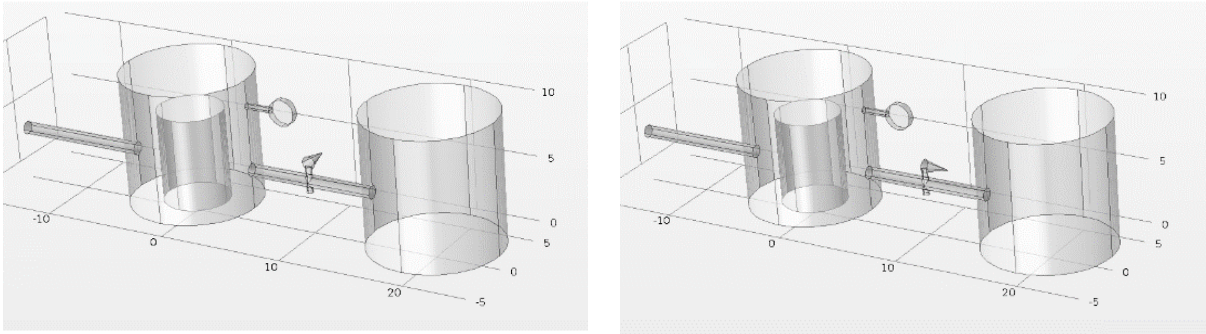


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Figure 7 – Working principle of gas pycnometer,
(a) the sample chamber pressurized only, (b) lower pressure after volume expansion

the sample and the reference chamber with known internal volume. The device also includes a valve between two chambers and pressure measuring device connected to the first chamber [7]. The working equation for a gas pycnometer is:

$$V_s = V_c + \frac{V_r}{1 - \frac{P_1}{P_2}}$$

Where $V_s$ is the sample volume, $V_c$ is the volume of the empty sample chamber, $V_r$ is the volume of the reference volume, $P_1$ is the first pressure (i.e. in the sample chamber only) and $P_2$ is the second (lower) pressure after expansion of the gas into the combined volumes of sample chamber and reference chamber.

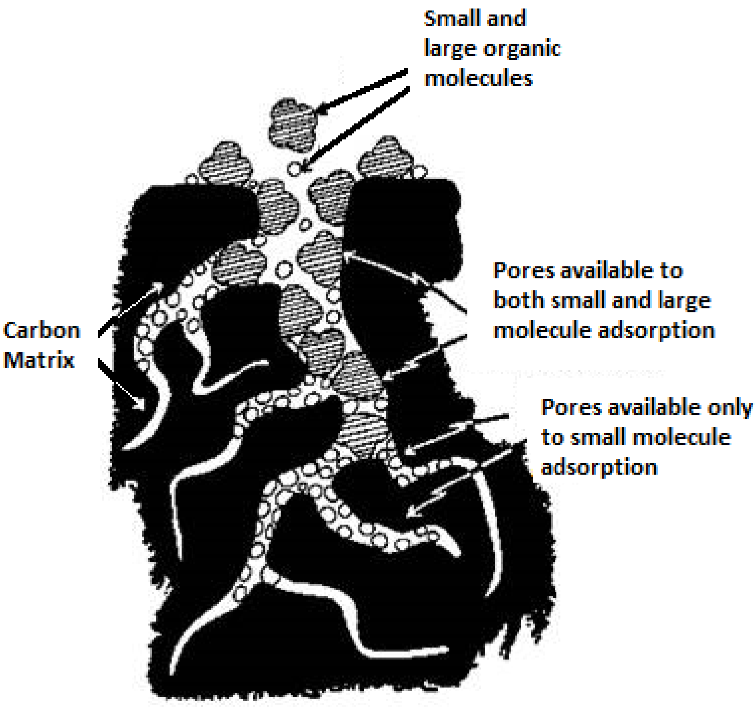


Figure 8 – Carbon pores illustration

*Skeletal density* of activated carbon investigated in [1] is about $\rho_s = 2200\ kg/m^3$. *Apparent (or real) density* is just the ratio of the weight to volume of the sample. For example an apparent density of activated carbon might be about $\rho_a = 275\ kg/m^3$.

**Micropore volume and adsorbent porosity.** A typical carbon is a mixture of micropores, mesopores, macropores and void space [10]. The pore is said to be micropore if its diameter varies between $2 - 20\ \dot{A}$, mesopore if $20 - 50\ \dot{A}$ and macropore $> 20\ \dot{A}$ [9]. In the voids and larger pores, the gas is stored at the gas phase. The *adsorption porosity* for a given adsorbate is found by exclusion the skeletal volume fraction and micropore volume fraction from the total volume fraction (equation below). It is convenient to express micropore volume in $cm^3g^{-1}$. For given specific sort of activated carbon the micropore volume is 1.7 $cm^3g^{-1}$. Therefore adsorption porosity $\gamma = 0.4075$ [1].

$$\gamma = 1 - \frac{\rho_a}{\rho_s} - \nu_\mu \rho_a$$

**Adsorbent particle density.** Porous carbon exhibits great range of densities, pore volumes and pore size distribution. Particle density is convenient to use for carbon characterization. Adsorption particle density is the ratio of adsorbent mass by the volume occupied by particles including micropores volume (therefore $\rho_p = 464.1\ kg \cdot m^{-3}$):

$$\rho_p = \frac{\rho_a}{1-\gamma} = \frac{\rho_a}{\frac{\rho_a}{\rho_s} + \nu_\mu \rho_a}$$

Highest adsorbed methane density is found in pores of effective pore width 7.4 $\dot{A}$. "Ideal" carbon would have only pores of 7.4 $\dot{A}$, density of porous carbon 0.75 g/mL, maximum methane capacity at 298 K is 152 g/L, ~230 v/v.

**Temperature effects.** The ANG storage is accompanied by temperature effects. During filling process, the temperature in the vessel rises, causing capacity loss. So the adsorption process is exothermic. Whereas during discharging process the temperature gradually decreases, which means that process is endothermic. In [11] the thermal changes during adsorption and desorption at different filling and extraction rates are investigated. Adsorbent tested in that study was a granular activated carbon and adsorbate was commercial methane. The 0.5 liter vessel used to store gas. The methane pumped into the vessel up to 3.5 MPa (or ~35 atm) then pumped out straightaway till atmospheric pressure. Fig.9 illustrates how the temperature, gas uptake and pressure change over time (*t*) during adsorption and desorption processes. The boundary that splits timeline into two parts is the moment when charging ends and discharging begins.

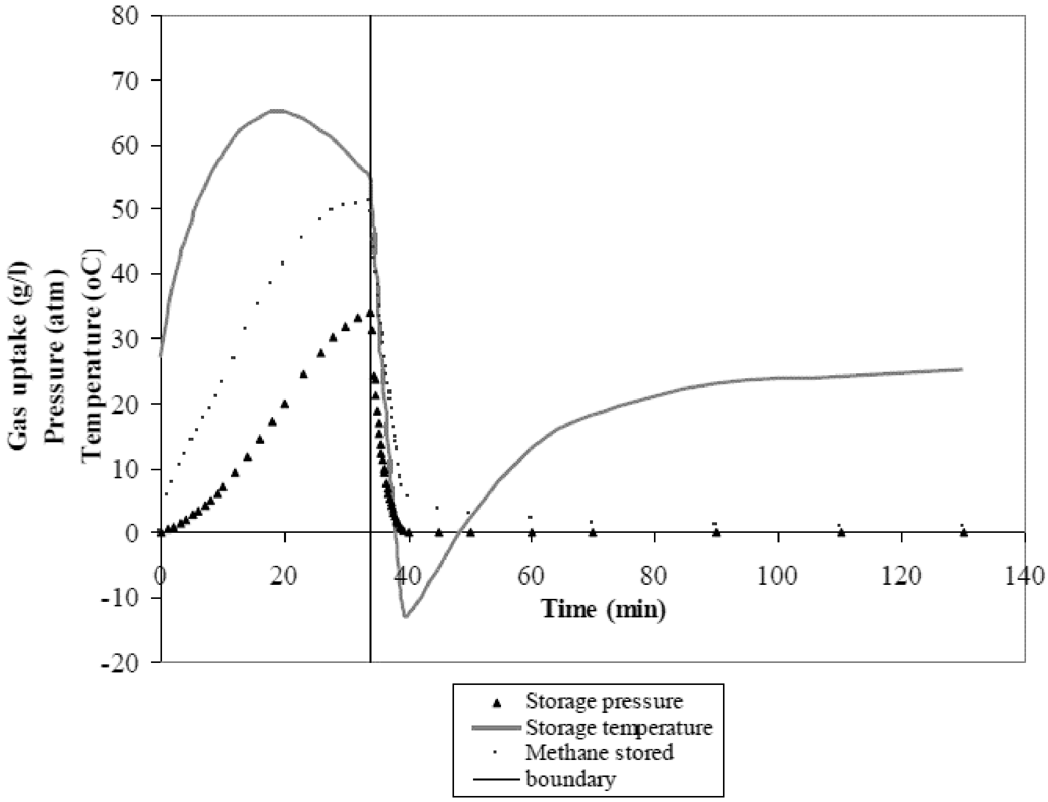


Figure 9 – Temperature, gas uptake and pressure change over time during filling and discharging ANG tank

Table 5 – Storage capacity, the highest temperature over filling process,
delivery capacity and the lowest temperature over discharging at different flow rates [11]

| Flow rate (*l/min*) | Storage capacity (*l/l*) | The highest temperature | Delivery capacity (*l/l*) | The lowest temperature |
|---|---|---|---|---|
| 1.0 | 85.70 | 43 °C | 70.61 | - 14 °C |
| 6.0 | 76.77 | 65 °C | 68.60 | -36 °C |
| 10.0 | 64.14 | 75 °C | 63.00 | -47 °C |

The results of experiments on measuring storage and delivery capacities of ANG vessel at different flow rates 1.0, 6.0 and 10.0 *l/min* can be found in Table 5. Results of experiment show the storage and delivery capacities are higher at slower flow rates. As higher the filling and discharging rate as sharper the temperature drop.

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**БЕЛСЕНДІРІЛГЕН КӨМІРДІҢ АДСОРБЦИЯЛЫҚ ЖӘНЕ ЖЫЛУЛЫҚ СИПАТТАМАЛАРЫ МЕН ОНЫҢ ТАБИҒИ ГАЗДЫ САҚТАУ ЖӘНЕ АДСОРБЦИЯЛЫҚ САЛҚЫНДАТУ ЖҮЙЕЛЕРІНДЕ ПАЙДАЛАНЫЛУЫНА ШОЛУ**

**Аннотация.** Кеуекті материалдар әртүрлі газдарды губка тәрізді сорып алады. Кеуекті орталардың бұл қасиеті адсорбциялық сақтау және салқындату жүйелерінде кеңінен қолданылады. Газдарды адсорбцияланған түрде сақтау қоймаларының көптеген артықшылықтары бар. Мысалы бұл технология компрессионды газ сақтау технологиясымен салыстырғанда 6 есе кем қысымда шамамен бірдей газ мөлшерін сақтау мүмкіндігін береді және алдын ала көпсатылы сығуға арналған қымбат компрессионды жабдықтарды талап етпейді. Адсорбциялау принципі салқындату жүйелерінде де кеңінен қолданылады. Мұндай жүйелерде айтылған принцип хладагентті газ күйінен қайтадан сұйық күйге айналдыруға арналған компрессор міндетін атқарады. Адсорбциялық салқындату жүйелері толық жұмысын атқару үшін электр қуатын қажет етпейді, сондықтан бұл технология электр қуаты жоқ нашар дамыған аймақтарда қолданылуы мүмкін. Бірақ, адсорбциялық салқындату жүйелері көп орынды талап етеді және меншікті салқындату қуаты төмен, соған сәйкес бұл технологияның қолданылуында және таралуында шектеулер бар. Берілген жұмыста адсорбция принципін қолданатын салқындату және газдарды сақтау жүйелерінің жұмыс істеу принциптері бойынша жан-жақты шолу жүргізілген. Ең жақсы адсорбент болып саналатын, белсендірілген көмірдің температуралық эффекттері, адсорбциялық қасиеттері және терминологиясы зерттеліп, нәтижелері төменде көрсетілген.

**Түйін сөздер:** адсорбция, белсендірілген көмір, адсорбциялық салқындату жүйелері (АСЖ), табиғи газды адсорбциялық сақтау (ТГАС), сақтау.

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**ОБЗОР АДСОРБЦИОННЫХ И ТЕПЛОВЫХ ХАРАКТЕРИСТИК АКТИВИРОВАННОГО УГЛЯ И ЕГО ПРИМЕНЕНИЕ В СИСТЕМАХ АДСОРБЦИОННОГО ХРАНЕНИЯ ГАЗА И ОХЛАЖДЕНИЯ**

**Аннотация.** Пористые материалы имеют свойство поглощать газ как губка. Это свойство пористых материалов широко используются для адсорбционного хранения газа и систем охлаждения. Хранение газа в адсорбированном состоянии имеет множество достоинств, данная технология позволяет хранить приблизительно такой же объем газа, как в компр ессионных газо-баллонах при 6 раз меньшем давлении и не требует дорогих компрессионных оборудований для предварительного многоступенчатого сжатия газа. Принцип адсорбции также используется в системах охлаждения. В которых, данный принцип выполняет функцию компрессора для преобразования хладагента из газового состояния обратно в жидкое состояние. Система адсорбционного охлаждения не требует электричества для полноценной работы, это позволяет использование данной технологии в отдаленных слаборазвитых регионах. Однако системы адсорбционного охлаждения громоздки и имеют низкую удельную мощность охлаждения, что ограничивает их использование и распространение. В работе проведен всесторонний обзор принципов работы систем охлаждения и методов хранения газа в адсорбированном состоянии. Терминология, температурные эффекты, адсорбционные характеристики активированного угля, который считается наиболее популярным адсорбентом были исследованы и результаты приведены ниже.

**Ключевые слова:** адсорбция, активированный уголь, адсорбционные системы охлаждения (АСО), адсорбционное хранение природного газа (АХПГ), хранение.

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