A REVIEW ON ADSORPTION WORKING PAIRS – EQUILIBRIUM EQUATIONS

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Abstract. *In face of greenhouse effect and growing energy demand, severe restrictions were imposed on the traditional vapour compression cycle, which promoted research for alternative refrigeration cycles. Adsorption cooling is a thermally driven system, which uses natural refrigerants (low or zero GWP and ODP) and requires little maintenance because of the low number of moving parts. Nevertheless, the COP and SCP of the cycle are too low to be economically viable. Improvements in various aspects are being made to make adsorption a competitive alternative. This work presents a review of the following solid adsorption working pairs: activated carbon-methanol, activated carbon-ammonia, zeolite-water and silica gel-water. It contains the operation temperature, applications, drawbacks and different types for each working pair. Equilibrium equations were presented with the coefficients of affinity for several types of working pairs.*

Keywords*: adsorption cooling, equilibrium equations, working pairs*

1. INTRODUCTION

The cold production refer to the general process of altering air properties such as temperature, humidity and velocity to achieve desired conditions of heat transfer from a low to a hot temperature environment. It has applications in uncountable fields of human life: food conservation, air conditioning, vaccine conservation, etc. For many decades, the primary cold production system used was the vapor compression with synthetic refrigerants (CFC, HCFC and HFC), which presents a high coefficient of performance. However, this type of technology causes serious environmental problems, it is estimated that its machines emissions, resulting from anomalies during operation of after useful life, accounts for 33% of the greenhouse effect (Edmunds et al., 1987).

Despite the expected 71% rise in energy consumption between 2003 and 2030 focusing only on the economy growth, the concern with sustainability and development of more rational ways to use energy has risen since the Montreal Protocol (1987) and Kyoto Protocol (1997) were established, aiming to severely reduce the emissions of the greenhouse gases. In the last recent years it has reached another dimension after the historical agreement of the USA and China to the 2012 Climate Conference (COP-17) to promote a legal commitment of reducing greenhouse gases emissions, including financing, technology exchange and development of new energy systems, revealing the decline trend of the use of vapor compression cycles (Fernandes et al., 2014).

The alternative that has emerged as a sustainable substitute to those pollution processes is the development of adsorption systems, aiming to counter the aforementioned effects, without reducing the desired level of comfort (Florides et al., 2002). They use heat as primary energy source, sustainable refrigerants such as methanol and water and have no movable parts, which makes the machines noiseless and with minimal maintenance needs. Adsorption is also attractive since it does not have crystallization problems and can operate at temperature as low as 50°C, while absorption systems require at least 70°C (Askalany et al., 2012).

Actually, the major problem involving this new technology is that while vapor compression and absorption are intrinsically continuous processes, adsorption is an intermittent one, and at least two adsorbent beds out of phase must be used to work in a quasi-continuous way, what corresponds to the hybrid systems. Because of this, the efficiency of the system is greatly reduced, providing the worst COP among the three systems.

Even though efficiency is a major problem for adsorption, the possibility of using solar energy at low temperatures shines a light on cost reduction for the process, revealing the possibility of promoting great savings in energy consumption (Cabrera et al., 2013). However, this energy source is intermittent (day and night) and unpredictable (rainy and cloudy days), what shows the need of research on materials able to improve the efficiency of the system and make its use viable. The northern part of Brazil as some parts of Africa have regions with pretty high insolation, and insufficient access to electricity, this way, in those cases, it is imperative that alternative means of cold production are provided (N'Tsoukpoe et al., 2014).

From this motivation, and due to the importance of this point, this paper features the use of solar adsorption technology, an overview of the adsorption working pair characteristics and capacities at different conditions of environmental temperatures and types of materials and its applicability depending on the use. It also introduces a review on adsorption equations of state.

2. EQUILIBRIUM EQUATIONS

Adsorption equilibrium of a certain pair represents the upper limit and maximum capability for the solid to adsorb the gas. In order to reach this limit, the gas is kept in contact with the solid for enough period of time (Hassan et al., 2015). Different equilibrium theories have been proposed for the adsorption isotherms. Henry's Law, valid for weak concentrations; Langmuir's theory, which takes into account adsorption in monomolecular layers; Gibbs's theory, which is based on the perfect gas equation and treats the adsorbate as microscopic and bi-dimensional; Adsorption Potential theory, which is based purely on thermodynamics and is suitable for microporus materials. The most common used equilibrium equations in adsorption are Dubinin, Langmuir, Freundlich, Toth and Cacciola.

The adsorbate–adsorbent equations of state are crucial to theoretical study and simulation work of adsorption-based cooling systems because they provide the required knowledge about the adsorption pair characteristics and capacities at different conditions of working temperatures according to the materials variability.

2.1. Dubinin Equations

The Dubinin equations are the most important model to describe adsorption equilibrium. In this model, the fraction of micropore volume, which is occupied by the adsorbate phase, is chosen to have a functional form of the Weibull distribution. Equation (1) shows the volume of adsorbate per unit of mass. W (m^3/kg) is the adsorption capacity, W_0 (m^3/kg) is the maximum adsorption capacity (volume of the pores), R is the adsorbate phase gas constant, E is the characteristic energy of adsorption, $P_s(T)$ is the adsorbate saturation pressure, n is parameter to describe the surface heterogeneity of the adsorbent. Equation (1) with n = 2 is regarded as Dubinin-Radushkevich equation (D-R).

$$W = W_0(T) exp\{-[RT/E \ln(P_s(T)/P)]^n\} \tag{1}$$

A more generalized form of the previous D-R model, being able to describe adsorption of gases in microporus adsorbents particularly appropriate for activated carbon with large pores heterogeneousness. This model is known as Dubinin-Astakhov (D-A) equations, and what makes it superior than the other ones is due to its thermodynamic character (Hassan, 2013). Equation (2) shows the equation for adsorption capacity, while Eq. (3) shows the equation for concentration. X (kg/kg) is the adsorbate concentration ratio, X_0 (kg/kg) is the maximum possible adsorbate concentration ratio and D is the coefficient of affinity. It is important to notice that D is dependent on the working pair and the brand and type of the adsorbent.

$$W = W_0(T) exp\{-D[T \ln(P_s(T)/P)]^n\} \tag{2}$$

$$X = X_0(T) exp\{-D[T \ln(P_s(T)/P)]^n\} \tag{3}$$

Critoph (1996) proposed a variation in the D-A equations, that works when the refrigerant works as a perfect gas. Equations (4) and (5) shows the modified equations. T_s is the adsorbate phase saturation temperature while K is an experimental constant.

$$W = W_0(T) exp\{-K[T/T_s(P) - 1]^n\} \tag{4}$$

$$X = X_0(T) exp\{-K[T/T_s(P) - 1]^n\} \tag{5}$$

2.2. Langmuir Equation

Langmuir theory was the first developed adsorption isotherm. It was originally developed to monolayer adsorption, however it was redesigned for multilayer (Hassan et al., 2015). This theory is based on thermodynamics and statistics. Equations (6) shows the Langmuir equations.

$$X = \sum_{i=1}^{i=n} \frac{X_{oi}\alpha_i P}{1+\alpha_i P} \tag{6}$$

In this equation, n is the number of surface sites, X_{oi} and α_i are temperature dependent parameters. Literature recommends n = 3; for this case, X_{oi} and α_i are given below. $\alpha_{j,i}$, $\beta_{0,i}$ and E_i are experimental parameters.

$$X_{oi} = \sum_{j=0}^{j=3} \frac{\alpha_{j,i}}{T^j}; \quad \alpha_i = b_{0,i} exp\left(\frac{E_i}{T}\right) \tag{7}$$

2.3. Freundlich Equations

Freundlich equation is an empirical equation used to describe equilibrium data.

$$X = X_0 \left(\frac{P}{P_s(T)}\right)^{(1/n)} \tag{8}$$

Saha et al. (1995) modified Freundlich equation to be more fitting with variations of temperature. Equations (9) and (10) shows this modified form. This modified equation is called S-B-K equation.

$$X = A(T) \left(\frac{P}{P_s(T)}\right)^{B(T)} \tag{9}$$

$$A(T) = \sum_{i=0}^{i=3} A_i T^i \; ; \quad B(T) = \sum_{j=0}^{j=3} A_j T^j \tag{10}$$

2.4. Toth Equations

Freundlich equations is not valid in low or high pressures. Toth equation is an empirical equation that accounts for this problem. It is suitable for systems with sub-monolayer coverage and works well with extreme pressures.

$$X = X_0 \frac{\beta P}{[1+(\beta P)^{\tau}]^{1/\tau}} \tag{11}$$

β and τ are depend on the adsorption pair. B is temperature dependent and takes the form $\beta = \beta_0 exp[Q_{sh}/(RT)]$, where Q_{sh} is the heat of adsorption and R is the gas constant for the gas phase.

$$X = X_0 \frac{\beta_0 exp[Q_{sh}/(RT)]P}{[1+\{\beta_0 exp[Q_{sh}/(RT)]P\}^{\tau}]^{1/\tau}} \tag{12}$$

2.5. Cacciola Equation

Cacciola et al. (1993) proposed the following equilibrium equation:

$$\ln(P) = a(X) + \frac{b(X)}{T} \tag{13}$$

The pressure is given in mB and the temperature in K. The coefficients a(X) and b(X) are evaluated experimentally as shown in Eq. (14).

$$a(X) = \sum_{i=0}^{i=3} a_i X^i \; ; \quad b(X) = \sum_{j=0}^{j=3} b_j X^j \tag{14}$$

3. ADSORPTION WORKING PAIRS

There are a wide number of adsorption working pairs and its choice must be carefully made by considering the affinity, physical and thermodynamic properties, the desired use of the refrigerating system, availability and cost (Fernandes et al., 2014). The adsorbent should provide large adsorption capacity and change of adsorption capacity with temperature variation, more flat desorption isotherm and good compatibility with the adsorbate. The adsorbate should have high latent heat per volume, defined freezing point and saturation vapor pressure, no toxicity, no flammability, no corruption, good thermal stability, etc. However, no working pair can meet all the requirements (Wang, D.C. et al., 2010).

The most common working pairs used in solid adsorption are: activated carbon-methanol, activated carbon-ammonia, zeolite-water and silica gel-water. Research in working pairs points to the development of composite materials (Choudhury, 2013).

The performance of the adsorption cooling system depends mainly on the working pair used. Since each pair differs from the other in terms of physical and thermodynamic properties, the choice of the working pair has a great effect on the system performance. A well system should have the characteristics of large adsorption capacity and a strong affinity of the absorbent for the refrigerant under consideration (Hassan et al., 2015).

Cloudhury et al. (2013) compiled a table with the different ranges of working pairs, heat source temperature and kind of solar collector, it is reproduced in Tab. 1.

Table 1. Regeneration temperature and collector types for some common working pair

Adsorbent – adsorbate pair	***Heat source temperature***	***Type of solar collector***
Silica gel – water	60-85°C	Flat plate (FPC) /evacuated tube collector (ETC)
Activated carbon – methanol	80-100°C	Heat pipe based evacuated tube collector
Calcium Chloride – ammonia	120-150°C	Low concentrator augmented collector
Activated carbon – ammonia	>150°C	Parabolic trough collector (PTC)
Zeolite – water	~170°C	PTC, concentrated collectors

3.1. Activated carbon – methanol

Activated carbon-methanol is one of the most commonly used working pairs in adsorption systems because of their large cyclic adsorption capacity, low desorption temperature being suitable for solar operation, low adsorption heat and high evaporating latent heat of methanol. This working pair however cannot operate at high temperatures higher, because the methanol decomposes at 120°C. High vacuum is needed and methanol has high toxicity (Wang, D.C. et al., 2010).

Zhao et al. (2008) tested Calgon activated carbon WS-480 with methanol and found the following parameters for X_0, D, and n: 0.269 kg/kg, $9.08x10^{-6}$, and 1.781. Alghoul et al. (2009) designed a dual purpose adsorption cycle, for refrigeration and water heating. Malaysian type granular activated carbon (AC-5060) and methanol was used as working pair. The values used for W_0, D, and n are $0.363x10^{-3}$ m^3/kg, 0.00002067, and 1.599.

Critoph (1996) measured seven different carbon based working pair in different temperature conditions. In this work, R32 was used, which is a refrigerant based on methanol, ammonia, and butane. The author found the following parameters for X_0, K and n for 208C type carbon: 0.476 kg/kg, 2,4634, and 1.3880, and for monolithic carbon he found: 0. 461 kg/kg, 2.6729, and 1.3326.

Wang, L.W. et al. (2006) did an experimental work with activated carbon-methanol (YKAC,14-20 mesh, produced from coconut shell) and found for X_0, K and n: 0.45 kg/kg, 13.38, and 1.5. Wang, R.Z. et al. (1997) studied a new adsorption refrigeration pair that could potentially be used in household applications. In the work, the Shangai YK (coconut shell type activated carbon) paired with methanol was tested. The authors found for X_0, K, and n: 0.284 kg/kg, 10.21, and 1.39.

Tierney (2008) simulated a driving convection thermal wave chiller using activated carbon-methanol as working pair, and adopted the following W_0, K, and n for the simulation: $0.356x10^{-3}$ m^3/kg, 32.65, and 2.0. The same author studied with activated carbon cloth and used for W_0, K, and n: $0.602x10^{-3}$ m^3/kg, 1.272, and 8.8135. Isosteres from the experimental work of Leite et al. (2005) were used do find those values.

3.2. Activated carbon – ammonia

Activated carbon-ammonia needs desorption pressure can reach 1.6 MPa. This high pressure helps the heat capacity ratio and the mass transfer. However, ammonia is toxic, has a displeasant smell, corrosive to cooper and may leak. Recent advancements improved the leakage problem and attracted many researchers (Wang, D.C. et al., 2010).

Wang, L.W. et al. (2006) experimental results for Eq. (4) with (YKAC,14-20 mesh, produced from coconut shell) activated carbon-ammonia were X_0, K, and n: 0.29 kg/kg, 3.57, and 1.38. Wang, L.W. et al. (2012) worked on a new consolidated composite activated carbon adsorbent to work with ammonia. The author used expanded natural graphite (ENG) soaked in sulfuric acid. The X_0, K, and n found for the granular activated carbon and composite adsorbent of activated carbon were respectively: 0.4655 kg/kg, 4.282, and 0.81; and 0.4703 kg/kg, 5.551, and 0.89.

Critoph (1996) measured three different types of carbon with three different refrigerants. The X_0, K and n parameters found when ammonia was used were: 0.290 kg/kg, 3.1853, and 1.0957 (208C carbon), 0.270 kg/kg, 4.3772, and 1.1965 (monolithic carbon), and 0.282 kg/kg, 4.6342, and 1.8065 (PVDC-Based carbon).

Tamainot-Telto and Critoph (2001) investigated two types of monolithic carbons for high performance adsorptions heat pumps. Ammonia was used as refrigerant in both cases. They X_0, K, and n parameters for the samples were: 0.3629 kg/kg, 3.6571, and 0.94 for the first simple (LML127 carbon) and 0.3333 kg/kg, 3.6962, and 0.99 for the second sample (LML128 carbon).

3.3. Zeolite – water

Zeolite-water has a much larger latent heat than other traditional working pairs. It should be pointed out that this working pair is suitable for air conditioning, since it cannot be used at temperatures below 0°C due to water freezing. This system was designed to use where the variation temperature of the adsorber is slow, and therefore, because of the high sensible heat, the SCP is lowered (Wang, D.C. et al., 2010). While zeolite is a natural mineral, it can be made artificially. There are over 40 types natural zeolites, and about 150 types synthetics ones. The most common zeolites used in adsorption are 4A, 5A, 10X, 13X (Askalany et al., 2012). Zeolite-water is the working pair normally used when high generation adsorption temperature is available, because of that, it is common to find studies of thermal wave adsorption using zeolite-water.

Douss and Meunier (1988) did a numerical and experimental work, and adopted the following W_0, D, and n for their simulations: 0.269x10^{-3} m³/kg, 1.8x10^{-7}, and 2.

Lu et al. (2003) proposed a zeolite 13X-water air conditioning system for the driver's cab of an internal combustion locomotive. Numerical and experimental studies were carried. For the simulation, the D-A was used, and the parameters X_0, K, and n were: 0.261 kg/kg, 5.36, and 1.73.

Solmuş et al. (2010) experimentally determined the parameters for Eq. (4) for natural zeolite (88-95% klinoptilolit – 0.5 mm grain size), using different zeolite temperatures and water vapor pressures. The authors used the D-A equation and found for X_0, K, and n: 0.1219 kg/kg, 5.052 and 1.4 (adsorption process); 0.1249 kg/kg, 3.620, and 1.2 (desorption process); 0.1233 kg/kg, 4.268 and 1.3 (average).

Zhang and Wang, R.Z. (2002) designed a zeolite 13X-water hybrid adsorption-ejector refrigerator driven by solar energy. The author used for the following values for the parameters X_0, K, and n: 0.261 kg/kg, 5.36, and 1.73, however the source was not specified.

Douss et al. (1988) performed an experimental investigation with adsorption heat pump using zeolite NaX-water working pair. Cacciola and Restuccia (1995) did a numerical study with a reversible heat pump. Zeolites 4A and 13X were compared. Cacciola equation was used in both studies. Table 2 shows the coefficients a(X) and b(x) for the different zeolites.

Table 2. Cacciola equation coefficients for 3 types of zeolite-water pairs

Type	***a_o***	***a_1***	***a_2***	***a_3***	***b_0***	***b_1***	***b_2***	***b_3***
4A	14.8979	0.95408	-63.666x10^{-3}	1.8488x10^{-3}	-7698.85	214.981	-18.4589	0.152605
13X	13.4244	1.10854	-73.176x10^{-3}	-1.6448x10^{-3}	-7373.78	67.2292	0.562447	-3.4867 x10^{-3}
NaX	8.9	-9.54 x10^{-4}	4.52 x10^{-7}	-4.48 x10^{-11}	3400	0.55344	-1.69 x10^{-4}	1.66 x10^{-8}

3.4. Silica gel – water

Silica gel-water is an interesting pair because it can be driven by low temperature heat source, with is great for solar energy purposes; however, it has low heat and mass transfer. Since the adsorbate is water, it cannot operate under 0°C and it also needs high vacuum. It is mainly used for air conditioning purposes.

Poor heat and mass transfer properties are the primary reason of low adsorption systems efficiency. Various approaches exists to enhance the system, like heat and mass transfer, and the development of adsorbents with improved properties. The new silica gel HKUST-1 (copper benzene-1,3,5-tricarboxylate) has an increase of water uptake of 93.2% (Wang et al., 2014) and a composite silica gel bound to PVP increased thermal conductivity by 78.6% (Sharafian et al., 2014).

Recent researches show that composite adsorbent with thermal conductivity enhancement or consolidated adsorbents can greatly enhance the system performance, a KSK zeolite-$CaCl_2$ + water working pair had an increase of COP and cooling capacity of 25% and 20% respectively (Choudhury et al., 2013).

Thu et al. (2013) studied the thermo-physical characteristics of silica gel using. Commercial available (Type-RD 2560, Type-A5BW and Type-A++) silica gel were tested with water as adsorbate. The W_0, E, and n parameters values found were respectively: 0.327x10^{-3} m³/kg, 4.384 kJ/mol, and 1.35 (Type-RD 2560), 0.455x10^{-3} m³/kg, 3.585 kJ/mol, and 1.25 (Type-A5BW), and 0.489x10^{-3} m³/kg, 3.804 kJ/mol, and 1.35.

Di et al. (2007) did a numeric and experimental study with silica gel-water pair. The authors adopted for X_0, K, and n: 0.346 kg/kg, 5.6, and 1.6. Silica type was not informed; however, COP had a maximum deviation of 7.5% when compared with experimental results.

Chihara and Suzuki (1983) did air drying experiments by adsorption swings using the silica gel (Fuji A type)-water pair. The authors fitted the experimental data using the Freundlich equation and found for X_0 and n: 0.346 kg/kg, and 1.6. Cho and Kim (1992) studied numerically and experimentally a silica gel (Fuji RD type)-water pair. The author fitted the data using the Freundlich model to find for X_0 and n: 0.552 kg/kg, and 1.6.

Miyazaki et al. (2009) proposed a new cycle time allocation in silica gel-water based adsorption chillers. In their study, the authors compared Silica gel RD and Silica Gel SWS-1L (a new material, impregnated with $CaCl_2$). The RD Silica Gel was described by the S-B-K, coefficients are present in Tab. 3. SWS-1L Silica Gel was described by the modified Toth equation, with the following X_0, β_0, Q_{sh}, and τ parameters: 0.8 kg/kg, 2x10^{-12} Pa^{-1}, 2760 kJ/kg, and 1.1.

Table 3. Coefficients A e B for RD silica gel-water pair

n	***0***	***1***	***2***	***3***
A(n)	-6.5314 kg	0.072452 kg/K	-0.23951x10^{-3} kg/K²	0.25493x10^{-3} kg/K³
B(n)	-15.587	0.15915 K^{-1}	-0.50612x10^{-3} K^{-2}	0.5329x10^{-6} K^{-3}

4. COMMENTS

The adsorption equations of state are crucial to the theoretical study and simulation work of adsorption-based cooling systems. Dubinin–Astakhov equations proved to be a robust method, that works for all working pair (especially activated carbon working pairs), even though there is a tendency of using fitting data methods for zeolite and silica gel.

Activated carbon-methanol is one of the most commonly used working pairs in adsorption systems, proving to deserve its status because of their large cyclic adsorption capacity, low desorption temperature that makes it suitable for solar operation, low adsorption heat and high evaporating latent heat of methanol, despite its requirements to operate at temperatures lower than 120°C.

Silica gel-water showed to be an interesting pair because it can be driven by low temperature heat source, what is great for solar energy purposes, and it was found that, despite its low mass and heat transfer capacity, it was able to achieve the best cooling COP of quasi continuous operation that aims to raise the system efficiency. However, further research is required to enable silica gel/water use. Since the adsorbate is water, it cannot operate under 0°C and it also needs high vacuum. The solution seems to be the development of new material to reduce the problems with the silica gel. The one named HKUST – 1(copper benzene – 1,3,5 – tricarboxylate) has an increase of water uptake of 93.2% and a composite silica gel bound to PVP increased thermal conductivity by 78.6%. There are numerous applications of silica gel-water pair as air conditioner.

5. CONCLUSION

In this article, a revision of common adsorption working pairs was presented. Firstly the equilibrium equations of state were presented, which is fundamental in any numerical work. Secondly, a brief description of most common working pairs (activated carbon-methanol, activated carbon-ammonia, zeolite-water and silica gel-water) was given, along with numerous numeric values for the parameters of the equilibrium equations, compiled from the literature.

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