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TECHNICAL NOTE

Effect of pressure on the moisture adsorption of silica gel and zeolite 13X adsorbents

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Abstract The effect of pressure on the adsorption characteristics of spherical adsorbents of zeolite 13X and RD silica gel is numerically analyzed in this study. The numerical model considers simultaneous heat and mass transfer in a spherical desiccant particle, which accounts for diffusion of moisture into the particles by both Knudsen and surface diffusion. The calculations indicate a dramatic difference of the adsorption behaviors between silica gel and zeolite 13X at a higher pressure of 7.5 atm due to the capability of adsorption and the diffusive ability of adsorbate within the adsorbent. For a lower system pressure of 1 atm, the variation amid silica gel and zeolite 13X is opposite to that at $P = 7.5$ atm. This is because the amount of the adsorbate for silica gel at $P = 1$ atm is significantly reduced. At a higher system pressure of 7.5 atm, the initial water content casts a very small influence on the adsorption behaviors for silica gel. However, for a normal pressure of 1 atm, a detectable difference is encountered subject to initial water contents. On the other hand, the initial water content casts appreciable influence on the adsorption characteristics for zeolite 13X.

List of symbols

C_b	Specific heat of adsorbent (J kg^{-1} K^{-1})
$C_{p,e}$	Specific heat of humid air (J kg^{-1} K^{-1})
D_{app}	Apparent diffusivity of zeolite 13X (m^2 s^{-1})
D_t	Total diffusivity of silica gel (m^2 s^{-1})
G_a	Mass velocity (kg s^{-1} m^{-2})
g'	The derivative of equilibrium isotherm ($=\rho(\partial m_1/\partial W)_T$)
H_{adsavg}	Heat of adsorption (J kg^{-1})
h_c	Sensible heat transfer coefficient
K_G	Gas side mass transfer coefficient (kg m^{-2} s^{-1})
m_1	Water vapor mass fraction, kg water/kg humid air
P	Pressure (atm)
r	Radial coordinate (m)
r*	Dimensionless radial coordinate, r/R (dimensionless)
R	Particle radius (m)
RD	Regular density (microporous)
RH	Relative humidity (%)
S_g	Specific pore surface area (m^2 kg^{-1})
t	Time (s)
T	Temperature (°C)
T_e	Ambient temperature (°C)
T_s	The average temperature of the adsorbent (°C)
W	Water content (kg water/kg dry adsorbent)

Greek symbols

ε	Porosity (dimensionless)
Λ	Thermal conductivity (W m^{-1} K^{-1})
ρ	Density (kg m^{-3})
τ	Tortuosity factor for intraparticle gas diffusion, dimensionless

Subscripts

0	Initial value
1	Water vapor
2	Dry air
avg	Average value

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b	Bulk
e	Surrounding humid air
eff	Effective value
K	Knudsen diffusion
s	Surface diffusion
p	Particle
total	Total value

1 Introduction

Compressed air had been implemented in many industrial and commercial processes such as cleaning, drying, manufacturing, and the like. Note that the water vapor content increases as the pressure rises. Therefore, it is imperative to remove the water vapor out of the ambient for applications like precision pneumatic tools. One of the methods to reduce the water content from compressed air is via condensing water vapor from compressed air and the method is primarily based on using the refrigeration system. However, the refrigeration method can only achieve the temperature that is generally lower than the pressure dew point 4.5 degrees [1]. For acquiring a very low pressure dew point temperature like −40 °C or even −70 °C, adsorption is proved to be more effective than that of refrigeration condensing method [1]. The key element of the adsorption system is adsorbent. Adsorbents like silica gel (SiO_2) and zeolite had been commonly used in a variety of dehumidification applications such as buildings and industrial humidifier. They have a very high moisture adsorption capacity due to zeolite's microporous structure of internal interlocking cavities which gives a high internal surface area (up to 800 m^2 g^{-1}, or 108–109 m^2 m^{-3}) [2]. Water vapor can diffuse through a porous medium by ordinary, Knudsen and surface diffusion. For silica gel at atmospheric pressure, the contribution by ordinary (Fick's Law) diffusion is normally negligible when the pore diameter is less than 200 Å [2], hence only the latter two mechanisms of diffusion need to be considered for the average pore diameter of regular density (RD) silica gel is 11 Å and it is around 68 Å for an intermediate density (ID) silica gel.

There had been numerous researches concerning the adsorption in normal pressures. However, the only published study in association with the effect of elevated pressure on the adsorption characteristics was carried out by Golubovic and Worek [3] who numerically investigated the effect of pressure on the adsorption/desorption wheel. They found that adsorption and desorption capacities of desiccant materials change as they are exposed to higher pressures. Also, as the operating pressure increases, the adsorption and desorption characteristics are dramatically affected and the optimum separation factor of desiccant material increases with operating pressure. The study provides valuable information about the influence of pressure on the desiccant wheel but their model considered only the exterior driving force outside the adsorbent, the internal diffusion resistance of the adsorbent is not taken into account. Until now, there is virtually no study concerning the effect of pressure on the fundamental heat and mass transfer of the adsorbent. In this regard, it is the objective of this study to bridge the gap. The external driving force outside the adsorbent and the internal diffusion resistance are considered in the investigation and efforts are stressed primarily on the adsorbents of silica gel and zeolite 13X (Faujasite).

2 Theoretical model

The basic model is similar to that of the Pesaran and Mills [2], the adsorbent is regarded as a spherical particle having a radius of 1.94 mm subject to ambient condition having relative humidity of 54.2 and 9.6 %. The adsorbents examined in this study include silica gel and zeolite 13X whose basic physical properties are taken from Pesaran and Mills [2], Kim and Ryn [4], and Ryn [5] and are tabulated in Table 1. Some details about the auxiliary relations like total diffusivity, heat of adsorption, mass transfer coefficient, convective heat transfer coefficient, and isotherms for silica gel and zeolite 13X can be found from previous studies by Pesaran and Mills [2], Ruivo et al. [6], Ni and San [7], San and Dai [8], Sladek et al. [9] and Gacciola and Restuccia [10]. However, at this stage, the isotherm of Zeolite 13X-Water vapor in air is not available. Hence the isotherm of Zeolite 13X-pure water vapor suggested by

Table 1 Physical properties of RD silica gel and zeolite 13X

Property	RDsilica gel (Pesaran and Mills [2])	Zeolite 13X (Kim et al. [4], Ryu et al. [5])
Porosity (volume void fraction), ε	0.485	0.25 (Kim et al. [4])
Average pore radius (1×10^{-10}m), Å	11	230 (Kim et al. [4])
Tortuosity factor, τ	2.8	
Specific pore surface area (10^5 m^2 kg^{-1}), S_g	7.8	7.26 (Kim et al. [4])
Bulk density, (kg m^{-3}), ρ_b	721.1	689 (Kim et al. [4], Ryu et al. [5])
Particle density, (kg m^{-3}), ρ_p	1,129	
Thermal conductivity, (W m^{-1} K^{-1}), λ	0.144	
specific heat, (J kg^{-1} K^{-1}), Cp	921	924 (Ryu et al. [5])

Cacciola and Restuccia [10] is used. It is recommended that further research for determination isotherm of Zeolite 13X-water vapor in air should be done in future.

The physical model is depicted in Fig. 1, considering a spherical adsorbent with an initial constant water content of W_0 and a uniform temperature T_0. Note that no temperature gradient in in the adsorbent particle is assumed and the lumped analysis of heat transfer is performed. The present lumped analysis provides a quick understanding of the response of adsorbent. For further examination of the influence of temperature variation, an exploration of the temperature non-uniformity is recommended in the future work.

The adsorbent is then suddenly placed in a ambient condition with the specific humidity varying with time, $m_{1,e}(t)$. Based on the water vapor diffusion within the adsorbent along with the considerations of (1) low mass transfer rate of the adsorption process; (2) Knudsen diffusion and surface diffusion are the major transport processes; and (3) the adsorption–desorption rate of water molecules is comparatively higher than the diffusion rate, Pesaran and Mills [2] developed the following heat and mass equations (further elaborations of the assumptions and considerations can be found from [2]):

The mass balance of the adsorbate in the solid

$$\frac{\partial W}{\partial t} = \frac{1}{r^2}\frac{\partial}{\partial r}\left[r^2 D_t \frac{\partial W}{\partial r}\right] \tag{1}$$

where D is the diffusivity, and for silica gel it is evaluated using the total diffusivity relation from [2], i.e.

$$D_t = D_{s,eff} + D_{K,eff}\frac{g'(W)}{\rho_p} \tag{2}$$

where D_t is a function of both gel water content W and particle temperature. $g\prime(W)$ is the derivative of equilibrium isotherm, $g'(W) = \rho(\partial m_1/\partial W)_T$. For zeolite 13X, the diffusivity based on the measured apparent diffusivity, D_{app}, from San and Dai [8]. Note that there is no experimental data reported for high pressure conditions. Hence, the mass transfer diffusivity by San and Dai [8] for low pressures was used. Yet their values and correlations are used when for the present adsorption modeling in high pressure. Despite its questionable applicability of in high pressure; however, since there is no relevant data reported in high pressures, the authors have to use their measurements and presume the applicability in high pressure. The equilibrium isotherm of the silica gel [2] and zeolite 13X [10] are given in the following:

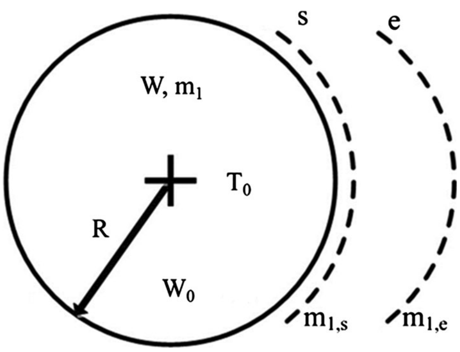


Fig. 1 Schematic of the diffusion of a spherical adsorbent with an initial water content of $W_0 = f(r)$ having a uniform temperature T_0

For RD silica gel:

$$\mathrm{RH} = 0.0078 - 0.05759W + 24.15554W^2 - 124.78W^3 + 204.226W^4 \tag{3}$$

For zeolite 13X:

$$\ln(P) = a(W) + (b(W))/T \tag{4}$$

where

$$a(W) = 13.422 + 1.10854W - 73.176\times10^{-3}W^2 + 1.6448\times10^{-3}W^3 \tag{5}$$

$$b(W) = -7373.78 + 67.2292W + 0.562447W^2 - 3.4867\times10^{-3}W^3 \tag{6}$$

where P is the vapor pressure (mbar), T is the adsorbent temperature (K). The associated initial and boundary conditions are:

$$\text{I.Cs.}\quad W(r,t=0) = W_0; T(r,t=0) = 24^\circ\mathrm{C} \tag{7}$$

$$\text{B.C.1.}\quad \frac{\partial W}{\partial r}|_{r=0} = 0 \tag{8}$$

$$\text{B.C.2.}\quad -\rho_p D\frac{\partial W}{\partial r}|_{r=R} = K_G(m_{1,s} - m_{1,e}) \tag{9}$$

At the interface, $r = R$, coupling (or equilibrium) condition is:

$$m_{1,s}(t) = f[W(r=R,t), T_s, P_{total}] \tag{10}$$

The B.C.2 is the continuity relation of water at the interface. The energy conservation equation for the adsorbent is given as

$$\left(\frac{4}{3}\pi R^3\right)\rho_b C_b\frac{\partial T_s}{\partial t} = \left(4\pi R^2\right)\left(h_c(T_e - T_s) - H_{adsavg}K_G\left(m_{1,s} - m_{1,e}\right)\right) \tag{11}$$

where the first term on the left hand side of Eq. (11) is the stored energy, C_b is the specific heat of adsorbent, H_{adsavg} is the heat of adsorption, h_c is the sensible heat transfer coefficient, T_s is the average temperature of the adsorbent and T_e is the ambient temperature. The associated relationship is given by Pesaran and Mills [2] where the mass transfer coefficient K_G and heat transfer coefficient h_c are evaluated using the following relationship:

$$K_G = 1.7 G_a \mathrm{Re}^{-0.42} \tag{12}$$

$$h_c = 1.6 \mathrm{G}_a \mathrm{Re}^{-0.42} \tag{13}$$

The heat of adsorbent, H_{adsavg} from [2, 4, 5] is correlated as:

For RD silica gel:

$$H_{adsavg} = 2501.6 - 2.3862T + 1 \times 10^{-3}T^2 - 2 \times 10^{-5}T^3 - 375.867 - 550\log(W) + 420W \tag{14}$$

For zeolite 13X:

$$H_{adsavg} = (2501 - 2.381T) \times \left(1.4712 - 3.6061W_{avg} + 17.5738W_{avg}^2 - 30.4424W_{avg}^3\right) \tag{15}$$

The average water contents of the adsorbent, W_{avg} is obtained by integrating Eq. (1) as follows:

$$W_{avg}(t) = \frac{\int 4\pi r^2 W(r,t)\rho_p dr}{(4/3)\pi R^3 \rho_p} \tag{16}$$

The foregoing equations with the initial condition, boundary conditions, and the auxiliary relations are solved using a commercial package COSMOL. In the simulation, the total number of nodes is 101. The initial time step is 0.001 s and the maximum time step is 0.1 s. The convergence criterion is 0.001.

3 Results and discussion

To elaborate the adsorption characteristics, the following parameters are defined and plotted in the figures:

Driving force of external moisture adsorption,

$$\Delta \mathrm{m}_{1,\mathrm{s}} = \mathrm{m}_{1,\mathrm{e}} - \mathrm{m}_{1,\mathrm{s}}(\mathrm{t}) \tag{17}$$

Ratio of driving force for external moisture adsorption,

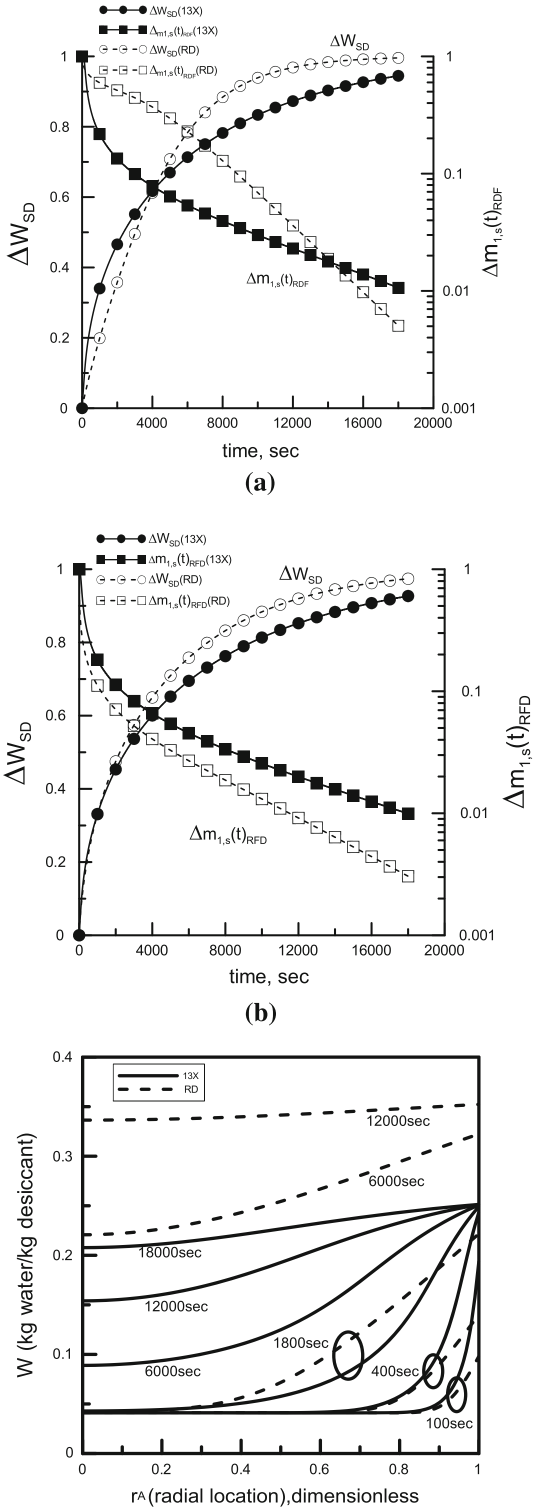
$$\Delta \mathrm{m}_{1,\mathrm{s}}(\mathrm{t})_{\mathrm{RDF}} = \left(\mathrm{m}_{1,\mathrm{e}} - \mathrm{m}_{1,\mathrm{s}}(\mathrm{t})\right) / \left(\mathrm{m}_{1,\mathrm{e}} - \mathrm{m}_{1,\mathrm{s}}(\mathrm{t}=0)\right) \tag{18}$$

Saturation degree of moisture transport,

$$\Delta \mathrm{W}_{\mathrm{SD}} = \left(\mathrm{W}_{\mathrm{avg}}(\mathrm{t}) - \mathrm{W}_0\right) / (\mathrm{W}_{\mathrm{final}} - \mathrm{W}_0) \tag{19}$$

Moisture adsorption rate is given as $\Delta \mathrm{W}_{\mathrm{avg}}/\Delta \mathrm{t} = d\mathrm{W}_{\mathrm{avg}}/d\mathrm{t}$. For a further examination of the adsorption characteristics, the 10 % driving force of moisture adsorption (i.e., $\Delta \mathrm{m}_{1,\mathrm{s}}(\mathrm{t})_{\mathrm{RDF}} = 0.1$) is labeled in the figure to highlight the turning point separating the low/high region of adsorption rate.

The effect of pressure on the adsorption characteristics versus time for silica gel and zeolite 13X is depicted in Fig. 2. As the pressure is elevated to 7.5 atm, the corresponding relative humidity is also increased from 9.6 to 72 %. As expected, the $\Delta \mathrm{m}_{1,\mathrm{s}}(\mathrm{t})_{\mathrm{RDF}}$ decreases considerably with time. However, in Fig. 2a at a given initial W_0 of 0.041 kg kg^{-1} dry adsorbent, one can see a dramatic difference between silica gel and zeolite 13X. For instance, an appreciable difference of the ratio of driving force, $\Delta \mathrm{m}_{1,\mathrm{s}}(\mathrm{t})_{\mathrm{RDF}}$, between these two adsorbents is seen especially when the time is less than 8,000 s. It appears that the ratio of driving force for the external moisture adsorption of silica gel is considerably higher than that of zeolite 13X. The difference may be explained from two aspects, the first one is associated with the capability of adsorption while the second one is due to the diffusive ability of adsorbate within the adsorbent. Figure 3a shows the equilibrium isotherm for silica gel and zeolite 13X and the data are taken from [2, 4]. It clearly shows that at a higher pressure of 7.5 atm the amount of the adsorbate for silica gel at equilibrium state is 0.357 kg water/kg dry desiccant which is almost five times higher than that at 1 atm (0.0765 kg water/kg dry desiccant). On the other hand, the amount of adsorbate at the equilibrium state for zeolite 13X varies slightly from 0.22 kg water/kg dry zeolite 13X to 0.25 kg water/kg dry zeolite 13X when the pressure is raised from 1 to 7.5 atm. The results can also be made clear from the saturation degree of moisture transport, $\Delta \mathrm{W}_{\mathrm{SD}}$, which also clearly depicts that the zeolite 13X adsorbent is more saturated than that of silica gel when the time is less than 4,000 s. Basically, the results are also related to the total diffusivity amid these two adsorbents. As can be seen from Fig. 3b which is taken from San and Dai [8], the apparent diffusivity, $\mathrm{D}_{\mathrm{app}}$, for zeolite 13X first decreases with the water content W to a minimum value near W = 0.17 kg water/kg dry adsorbent, followed by a moderate rise when W is further increased. However, the total diffusivity for silica get shows a monotonic increase in association with W. It is interesting to know that $\mathrm{D}_{\mathrm{t,silica\_gel}}$ is slightly lower than that of $\mathrm{D}_{\mathrm{t,zerolite\_13X}}$ when W is less than 0.061 kg/kg dry adsorbent. In addition, for a specified $\mathrm{W}_0 = 0.041$ kg water/kg dry, the corresponding relative humidity for zeolite 13 X (RH = 0.003 %) is much lower than that of silica gel (RH = 3.8 %). Thus, the initial driving force of the external moisture for zeolite 13X exceeds slightly that of silica gel. Therefore, one can see more water content is adsorbed to the adsorbent ($\mathrm{W}_{\mathrm{avg,13X}} > \mathrm{W}_{\mathrm{avg,RD}}$) at the initial stage of the transient process. However, as shown in Fig. 3(b) the total diffusivity for zeolite 13X is decreased with the rise of water contents when the water content is less than 0.17 kg/kg dry adsorbent. The results can be further made clear about the variation of water contents (W) with elapsed time within the spherical adsorbent for an initial water content 0.041 kg/kg dry adsorbent as shown in Fig. 2c. As expected, the water content increases and approaches the

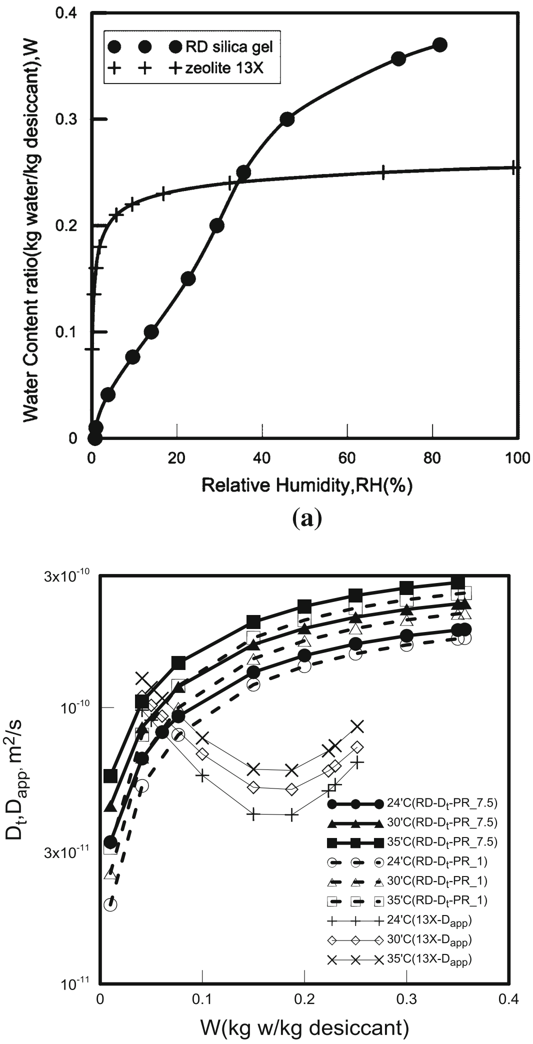


(a)

(b)

(c)

Fig. 2 Effect of the pressure on the adsorption characteristics of zeolite 13X and silica gel and the variation of water content within the adsorbent. **a** P = 7.5 atm, **b** P = 1 atm, **c** Variation of water content alongside the adsorbent for P = 7.5 atm and $W_0 = 0.041$ kg/kg dry adsorbent



(a)

(b)

Fig. 3 Equilibrium isotherms (**a**), total diffusivity and apparent diffusivity (**b**) for RD silica gel and zeolite 13X

saturated water contents with elapsed time and water content rises comparatively rapidly near the adsorbent surface. However, as shown in Fig. 2c, the water content within the adsorbent for zeolite approaches its saturated concentration more quickly as compared to silica gel, thereby suggesting a larger internal diffusion resistance. The results shown in Fig. 2c also imply that the internal diffusion plays an essential role in the water vapor transport process. A close comparison shows that the driving force of external moisture adsorption for zeolite 13X is inferior to that of silica gel when time is roughly higher than 200 s while the total diffusivity of silica gel exceeds that of zeolite 13X when the elapsed time is about 300 s. The combined effect accentuates the adsorption behavior of silica gel, leading to a better adsorption characteristic of

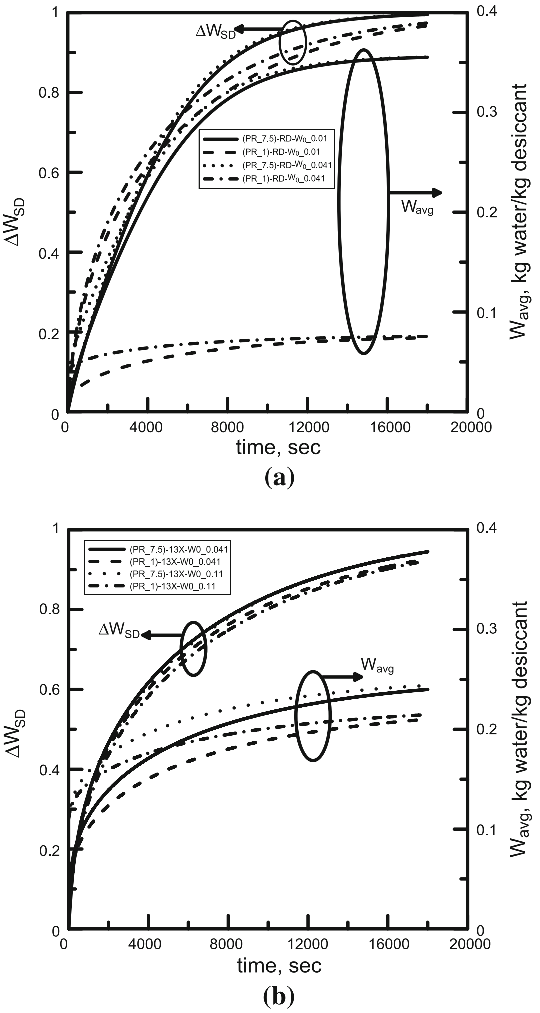


Fig. 4 Effect of the initial water content of the adsorbent on the adsorption characteristics of zeolite 13X and silica gel subject to system pressure. **a** P = 7.5 atm, **b** P = 1 atm

silica gel. The results can be further seen in Fig. 2a that silica gel approaches the 90 % degree of saturation in about 8,810 s while it is about 13,772 s for zeolite 13X at an initial water content of 0.041 kg/kg dry adsorbent.

The foregoing discussion is applicable for high pressure operation (P = 7.5 atm), results for 1 atm is shown in Fig. 2b. Notice that the variation of the $\Delta m_{1,s}(t)_{RDF}$ amid silica gel and zeolite 13X is opposite to that at P = 7.5 atm. The ratio of driving potential for zeolite 13X is always superior to silica gel throughout the calculation time. This is not surprising and can be also explained from the isotherm in Fig. 3(a) where the amount of the adsorbate for silica gel at P = 1 atm is significantly dropped to 0.0765 kg water/kg dry desiccant while no significant variation is seen for zeolite 13X. As a result, the saturation degree, ΔW_{SD}, between zeolite 13X and silica gel is rather small when the elapsed time is less than 3,000 s.

The effect of initial water content on the adsorption behavior for silica gel and zeolite 13X subject to the system pressure is shown in Fig. 4 where W_0 is 0.041 or 0.01 kg water/kg dry adsorbent. It is interesting to know that the initial water content casts a very small influence on the adsorption behaviors for silica gel at a high pressure of 7.5 atm. As seen in Fig. 4a, the time required to reach 90 % saturation water content RD silica gel is about 9,200 s when $W_0 = 0.01$ kg water/kg dry adsorbent while it is about 8,800 s for $W_0 = 0.041$ kg water/kg dry adsorbent. The difference in time is less than 5 %. On the other hand, for a normal pressure of 1 atm, the time difference for reaching the 90 % saturation water content exceeds over 40 %. This is because the presence of high pressure has increased the ambient relative humidity from 9.6 to 72 %, and the saturated water content is 0.357 kg water/kg dry silica gel when P = 7.5 atm as compared to 0.0765 kg/kg silica gel at P = 1 atm. However, this is not the case for zerolite 13X as shown in Fig. 4b. The time required to reach the 90 % saturation water content amid $W_0 = 0.041$ and 0.01 kg/kg dry adsorbent is considerably increased since the saturated water contents is insensitive to change of the elevated pressure. It is also interesting to know that the degree of saturation is also insensitive to change of initial water contents, and this is applicable for both silica gel and zeolite 13X.

4 Conclusions

This study numerically examines the influence of elevated pressure on the adsorption characteristics of spherical adsorbents. The adsorbents include zeolite 13X and RD silica gel and the pressures are 1 and 7.5 atm, respectively. The numerical model considers simultaneous heat and mass transfer of adsorbent particle having spherical configuration. The modeling takes into account the Knudsen and surface diffusion. Some major conclusions are given in the following:

1. For a higher pressure of 7.5 atm, the calculations indicate a dramatic difference of the adsorption behaviors between silica gel and zeolite 13X, and this phenomenon is especially pronounced when time is less than 8,000 s. This is associated with the capability of adsorption and the diffusive ability of adsorbate within the adsorbent. The diffusive ability within the adsorbent plays an essential role in the overall adsorption behavior.
2. For a lower system pressure of 1 atm, the variation amid the silica gel and zeolite 13X is opposite to that at P = 7.5 atm. The ratio of driving potential for zeolite

13X is always superior to silica gel throughout the calculation time. This is because the amount of the adsorbate for silica gel at $P = 1$ atm is significantly dropped to 0.0765 kg water/kg dry desiccant which is only about one fifth of that at $P = 7.5$ atm. On the other hand, the effect of pressure on the saturation water content for zeolite 13X is rather small.

3. The effect of initial water content on the adsorption behavior for silica gel and zeolite 13X subject to the system pressure is also different. At a higher system pressure of 7.5 atm, the initial water content casts a very small influence on the adsorption behaviors for silica gel. However, for a normal pressure of 1 atm, a detectable difference is encountered subject to the initial water contents. On the other hand, this is not the case for zeolite 13X. Appreciable difference is found for the required time to reach the 90 % of saturation water content amid $W_0 = 0.041$ and 0.01 kg/kg dry adsorbent. This is because the saturated water content for zeolite 13X is insensitive to change of the elevated pressure.

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