

ON THE DYNAMICS OF HUMIDITY ADSORPTION

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Abstract. *This work presents a numerical model for the dynamics of the simultaneous heat and mass transfer problem in the adsorption/desorption process of water vapor by silica gel. Employing the finite volume method, the model solves the coupled equations for a two dimensional transient case. Simulation results have showed good agreement with experimental data available in the literature. It is also presented a hierarchy of desirable thermophysical properties that candidates to be employed as humidity sensors have to attain.*

Keywords: *adsorption, water vapor, silica gel, humidity sensor.*

1. Introduction

Air humidity measurement and control have many important industrial applications and also in air conditioning (Debat, 1996). Traversa (1995) reported the state-of-the-art of humidity measurement, highlighting ceramic materials as those more promising for humidity sensor construction. However, humidity sensors built using hygroscopic materials possess high time constants. Thus, they are usually employed as a covering on an inert substrate. Estimates with good precision of time constants of several candidate materials will compose a valuable contribution to guide the selection and application of those as humidity sensors. Among ceramic materials, silica gel was selected because its great affinity to water, what makes it a large usage desiccant material.

Air humidity sensors built through deposition of a thin silica gel layer on another material has been object of some studies (Randin and Zullig, 1987, Taosun *et al.*, 1993, Innocenzi *et al.*, 2001, Bearzotti *et al.*, 2003). These works concentrate on silica film preparation and in experimental determination of sensor performance parameters.

The dynamics of air moisture uptake by a silica gel film has other important applications. Biwas *et al.* (1984) proposed the construction of desiccant beds through deposition of a thin silica layer on parallel metallic plates among which air flows, with advantage of resulting in smaller friction energy loss in comparison with the traditional package bed filled with silica spherical particles. Furthermore, rotatory honeycomb wheels have increasing application in air desiccation, and it consists of silica covering on a metal mainframe.

When a humid air stream passes on a silica gel film, water vapor mass transfer is a purely diffusive phenomenon. Some authors considered the transient convective mass transfer of air moisture to a silica gel bed (Pesaran and Mills, 1987a, 1987b, San and Jiang, 1994, San *et al.*, 2002). Others studied transient water vapor mass diffusion within a single silica gel spherical pellet (Kruckels, 1973, Pesaran and Mills, 1987a, 1987b, Ni and San, 2002). Bostain *et al.* (2002) has experimentally studied water vapor uptake by a silica aerogel block using neutron radiography. Their focus was experimental only, so the presented analytical solutions were as simple as those encountered in diffusive mass transfer general textbooks (Crank, 1975, Cussler, 1984).

The analysis here presented treats the problem in the plan, i.e., in a two-dimensional way. Two opposite faces are insulated and the remaining others are exposed to a humid air stream, whose upstream velocity, temperature and water vapor concentration are considered known and uniforms, as well as initial temperature and water adsorbed mass distributions inside the solid.

2. Physical-mathematical model

2.1. Equilibrium relationship

Silica gel is an incompletely dehydrated polymeric structure of colloidal silicic acid with the formula $\text{SiO}_2 \cdot n\text{H}_2\text{O}$. This amorphous material consists of spherical particles 2-20 nm in size, sticking together to form the adsorbent silica gel (Unger, 1979).

For a given gaseous adsorbate-solid adsorbent system, the adsorption equilibrium data can be related as $W^* = W^*(p, T)$, where W is the absorbed mass (kg adsorbate / kg adsorbent), p is the adsorbate partial pressure, T is the temperature and superscript * refers to equilibrium condition. If equilibrium data are all obtained at the same temperature, a plane plot, known as isotherm, becomes possible.

For water vapor adsorption in purely micro-porous silica, Langmuir's isotherm is obtained (Unger, 1979). Within Langmuir-isotherm low adsorbate concentration range, Henry's law is approached (Ruthven, 1984):

$$W^* = K p^* \quad (1)$$

where K is the Henry's constant, expressed in Pa^{-1} .

The validity of Eq. (1) on silica gel-water system modeling was experimentally confirmed by Ng *et al.* (2001). The temperature dependence of Henry's law constant obeys the Van't Hoff equation (Ng *et al.*, 2001):

$$K = K_0 \exp[Q_{st} / R_w T] \quad (2)$$

where Q_{st} is the isosteric heat of adsorption in J kg^{-1} , R_w is the water specific gas constant and the subscript 0 designates a reference condition.

Replacing Eq. (2) into Eq. (1):

$$W = K_0 \exp[Q_{st} / R_w T] \cdot p \quad (3)$$

Empirical parameters of Eq. (3) can be determined at saturation condition, always indicated here by the subscript s , when maximum silica gel water content is in equilibrium with 100%-relative-humidity neighboring air. It is still needed to verify if calculated parameters remain valid throughout the considered temperature range.

Applying ideal gas model to free water vapor, the equilibrium relationship correlating free water vapor concentration (C) in kmol m^{-3} and adsorbed water mass (W) in $\text{kg water / kg silica}$ can be expressed as:

$$W = K_0 \exp[Q_{st} / R_w T] \cdot (p_s / C_s) \cdot C \quad (4)$$

2.2. Mass transfer equation

Crank (1995) presents the physical-mathematical modeling of the absorption of one substance by another through which it can diffuse and with which it can also react chemically. If the reaction by which the immobilized reactant is formed proceeds very rapidly compared with the diffusion process, local equilibrium can be assumed to exist between the free and immobilized components of the substance in diffusion. In the simplest case, a linear equilibrium relationship, as expressed by Eq. (1), must hold.

In the problem approached here, water vapor adsorption by silica gel behaves like an instantaneous and reversible chemical reaction.

The local rate of adsorption is assumed instantaneous because it approximates the order of collision frequency of the gas on the solid surface, which is much greater than the transport processes (Yang, 1997).

Pesaran and Mills (1987a, b) noted that the mechanism that limits the mass transfer rate within a silica gel bed is the transport of water vapor inside silica gel micropores. They proposed the solid side predominant resistance model (SSR), which assumes that internal pellet diffusivity is representative of the overall diffusivity. The applicability of SSR model to silica gel-water vapor system was confirmed again by San *et al.* (2002). Pesaran and Mills (1987a) concluded that for silica type Regular Density (RD) only surface diffusion needed to be considered, and for silica type Intermediate Density (ID), both surface and Knudsen diffusion must be considered. They proposed Eqs. (5) and (6) for calculating silica RD and ID effective diffusivities (D), respectively.

$$D = D_s = (1 / \tau_s) 1.6 \times 10^{-6} \exp(-0.947 \times 10^{-3} H_{ADS} / T) \quad (5)$$

$$D = D_s + (1 / \tau_K) \varepsilon_p 22.86 a \sqrt{T} (g'(W) / \rho_p) \quad (6)$$

In the above equations, the subscripts s and k refer to surface and Knudsen diffusion mechanisms respectively, τ is the adimensional tortuosity factor that must be equal to unity as recommended by San *et al.* (2002), ε is the porosity, $g'(W)$ is the isotherm derivative $(\partial C / \partial W)_T$, the subscript p refers to silica particle, a is the silica pore radius in m^{-10} , H_{ADS} is the heat of adsorption in J / kg water and ρ is the density.

Assuming negligible free water vapor accumulation within silica gel pellets, and constant and uniform effective diffusivity, besides local equilibrium, using t to designate the time, the mass transfer equation has become:

$$\varepsilon \partial C / \partial t = D (\nabla^2 C) - (1 - \varepsilon) K \partial C / \partial t \quad (7)$$

2.3. Heat transfer equation

It was assumed here that the solid and gas phases inside the silica gel block possess the same temperature, i.e., local thermal equilibrium holds. Kim and Jang (2002) verified that the assumption of local thermal equilibrium is always valid in all porous media when the conduction heat transfer mode is dominant.

Considering constant and uniform all effective thermo-physical properties, as well as heat generation by adsorption process, the heat transfer equation can be expressed as:

$$\rho c \partial T / \partial t = k(\nabla^2 T) + (1 - \varepsilon) H_{ADS} \rho \partial W / \partial t \quad (8)$$

In Eq. (8), k is the silica thermal conductivity. Silica specific (c) and adsorption heats were treated taking into account their variations with adsorbed water mass. As suggested by Pesaran and Mills (1987a), at each time level new spatial mean values obtained from corresponding Eq. (9) and Eq. (10) for silica RD or Eq. (11) for silica ID were applied.

$$c = 4186 W + 921 \quad (9)$$

$$H_{ADS} / 1000 = -12400W + 3500 \quad W \leq 0.05 \quad H_{ADS} / 1000 = -1400W + 2950 \quad W > 0.05 \quad (10)$$

$$H_{ADS} / 1000 = -300W + 2095 \quad W \leq 0.15 \quad H_{ADS} / 1000 = 2050 \quad W > 0.15 \quad (11)$$

Including the above mentioned treatments and replacing the equilibrium relationship into heat source term, the heat transfer equation has become:

$$\rho \partial (cT) / \partial t = k(\nabla^2 T) + (1 - \varepsilon) \rho \partial (H_{ADS} KC) / \partial t \quad (12)$$

2.4. Other model assumptions

It was also assumed that the reestablishment velocity of thermal and mass boundary layers after any thermal or mass disturbance is high in comparison with velocities of diffusive phenomena that occur in the silica.

3. Numerical model

It was developed a numerical model, using finite volume method (Patankar, 1980). Boundary conditions were included in algebraic equation system through the application of balance equations to boundary volumes, which have the same dimensions as internal volumes, as recommended by Maliska (1995). Details of mentioned numerical method can be seen in Costa Filho (2004). It was used a full implicit numeric scheme, in order to allow the evaluation of boundary conditions at the same time marching level. Convective heat and mass transfer coefficients were obtained through Reynolds-Colburn Analogy (RCA) and Chilton-Colburn Analogy (CCA), respectively. Although these analogies were initially derived to a flat plate with surface temperature and concentration constants, both had been proved to be for generalized usage (Welty, 1984). Since the humid air flow is steady state, calculated coefficients are constants. This seems to be inconvenient since convective coefficients certainly vary throughout time evolution.

4. Conjugated problem

The objective of this conjugated analysis is to coupling solve diffusive problem in silica and convective problem in humid air stream.

In the steady state incompressible laminar flow of a constant properties fluid, the momentum and thermal boundary layer equations can be solved separately (Arpaci and Larsen, 1984). Energy becomes a linear equation, since in its solution, velocity vector components are previously known from solution of flow equations. Then, it is possible to obtain new solutions from already available ones for thermal boundary layer equation through application of the Duhamel's theorem. Thus, the solution of equation of a thermal boundary layer on a flat plate that has an arbitrary variation of its surface temperature can be composed from the overlap of solutions in which the plate temperature experiments only a step change at a pre-fixed length (x_0) from the plate edge, represented by Eq. (13) (Eckert and Drake, 1974). It had better to highlight that here it is considered a transient thermal boundary layer within a steady state hydrodynamic boundary layer. Temperature difference is assumed to be small such that fluid physical properties can be assumed as constants and then the velocity field, as described by Blasius' solution, stays unaffected.

$$Nu_x = hx / k = 0,332\sqrt[3]{Pr}\sqrt{Re_x}1/\left(\sqrt[3]{1-\left(\frac{x_0}{x}\right)^{3/4}}\right) \quad (13)$$

Equation (13) can be used for gases with acceptable errors (Eckert and Drake, 1974). In it, Nusselt, Prandtl and Reynolds numbers are respectively represented by Nu , Pr and Re , x designates a distance from plate edge and h indicates the convective heat transfer coefficient at this position.

To extend the solution presented here to transient regime, it is necessary to suppose that at each time level a steady state regime condition is held, and time marching is done by a picture to picture evolution. This approach is referred as *quasi steady* analysis. This assumption is based on the fact that thermal diffusivity of air is much larger than silica's one. So, it was able to compare how much a given thermal disturbance happened in the surface is diffused in the air and in the silica in the same period. Disturbance propagation within thermal boundary layer was treated as purely diffusive, that is the most unfavorable case, since close to the surface the diffusive term prevails on the convective terms in thermal equation boundary layer.

Conjugated problem numerical solving was added to main program as following: At each new time level, the main program releases temperature and concentration fields within the silica, using third type boundary conditions with convective heat and mass transfer coefficients already available from last iteration. Through application of heat and mass flow continuities at interface, its temperature and concentration becomes updated. Interfacial temperatures are needed in order to apply the above described analytical solution, which leads to new values to convective heat transfer coefficients, from which using CCA, new values to mass transfer coefficients are also obtained. Anterior and updated convective coefficients are individually compared, and if all errors are lesser than a pre-fixed tolerance, the computing skips to the next time level; otherwise, new iterations at the same time level are carried out until tolerance criteria are attained.

5. Results

5.1. Tested materials

Properties from silica RD and ID from Pesaran and Mills (1987a), used as testing materials are presented in Tab. 1.

Table 1. Material properties from two type tested silica.

Material Properties	Silica RD	Silica ID
Pore Mean Radius (Å)	11	68
Particle Density (kg m ⁻³)	1129	620
Bulk Density (kg m ⁻³)	721.1	400.6
Maximum Moisture Content (kg kg ⁻¹)	0.4	0.21
Equilibrium Relation	Eq. (4)	Eq. (4)
Effective Mass Diffusivity (m ² s ⁻¹)	Eq. (5)	Eq. (6)
Specific Heat (J kg ⁻¹)	Eq. (9)	Eq. (9)
Adsorption Heat (J kg ⁻¹)	Eq. (10)	Eq. (11)
Thermal Conductivity (W m ⁻¹ K ⁻¹)	0.198	0.198

5.2. Numerical model validation

Figure 1 shows a comparison among experimental data from Bostain *et al.* (2002) and data generated by the developed computational simulation model for water vapor uptake by a silica aerogel parallelepiped at some pre-selected distances from the adsorption face. The remaining five faces were all insulated against mass transfer.

Aerogel is an unusual porous material, produced by a sol-gel process that results in a solid material with a unique microstructure composed of nanometer-size particles and pores. Silica aerogel is one of the most common forms of aerogel and consists of mostly air with the remainder being a wispy matrix of silica (Bostain *et al.*, 2002).

The best fit to experimental data was obtained with the value of $2,2 \times 10^{-11} \text{ m}^2 \text{ s}^{-1}$ for the water vapor effective mass diffusivity in silica gel, which is within its range determined experimentally by Lu *et al.* (1991) and Ni and San (2002). Only to the smallest depth, 2,72 mm, the numeric result was far from the experimental one. Bostain *et al.* (2002) experimental data are referred to their third run. Runs 1, 2 and 3 reused the same aerogel specimen after a drying step, so one can suppose that aerogel water adsorbed initial content were not totally uniform. Assuming that this region was a little dryer at the beginning of the run, since it is closer to the exposed face, a better fit was achieved.

Within simulations, the maximum observed increase in aerogel mean temperature was 0,2 °C, what has validated Bostain *et al.* (2002) not to consider energy balance equation in their analysis. The high aerogel bulk porosity reduces its water mass adsorption capacity and consequently, it associated heat generation.

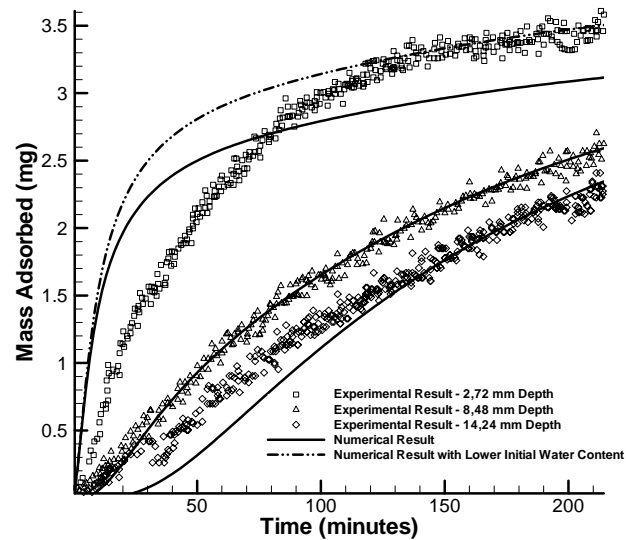


Figure 1. Comparison among numerical results with experimental ones from Bostain *et al.* (2002) on water vapor uptake by an aerogel parallelepiped at some pre-selected distances from the adsorption face

3.3. Simulation results

Figure 2 presents a comparison between simulation results generated by conjugated analysis, and those obtained using convective heat and mass transfer coefficients derived respectively from RCA and CCA analogies. Simulation conditions are summarized in Tab. 2. One can note that conjugated analysis has led to different results only to temperature. So, mass transfer results are not influenced by the fact if problem approach is conjugated or not.

Table 2. Conditions assumed at simulation cases illustrated in Figs. 2, 3 and 4.

Simulation case parameters		Simulation case parameters	
Length (mm)	10	Silica initial temperature (°C)	25
Thickness (mm)	10	Air flow relative humidity (%)	50 or 75
Humid air uniform velocity (m s^{-1})	0.5	Relative humidity (%) within voids in silica	50 or 75
Humid air flow temperature (°C)	25		

Figures 3 and 4 present respectively temperature and free water vapor concentration profile evolutions during adsorption at the same conditions of Fig. 2. It is important to note that significantly slower mass transfer rate determinates transient phenomena duration.

Pesaran and Mills (1987a and b) analysis are also valid here. Immediately following the step change on humid air stream conditions, the dry silica adsorbs water vapor and liberates heat at a high rate; consequently the silica temperature increases rapidly, as well as its water adsorbed mass. The silica gradually loses its adsorptive capacity due to the increase in silica water content and temperature. The maximum in temperature is reached when the cooling effect of the air flow balances the heat of adsorption being released, and thereafter the reduced rate of adsorption causes temperature to decrease. The reverse of the above explanation is valid for the desorption cases.

Figure 5 shows sensor time constant variation with silica film thickness and with humid air uniform flow velocity, for silica types RD and ID. Transient phenomena in silica ID is considerably shorter because its lower water adsorption capacity and higher water vapor mass diffusivity, allowing a thicker silica film to be used as humidity sensor. Simulation conditions for Fig. 5 are summarized in Tab. 3.

It can be observed that sensor time constant increases with humid air uniform velocity, when the sensor is drier than air stream, and when sensor initial water content is higher, it happens exactly the opposite. This occurs because air uniform flow velocity exerts significant influence just on sensor temperature, and mass diffusion is faster when temperature is higher.

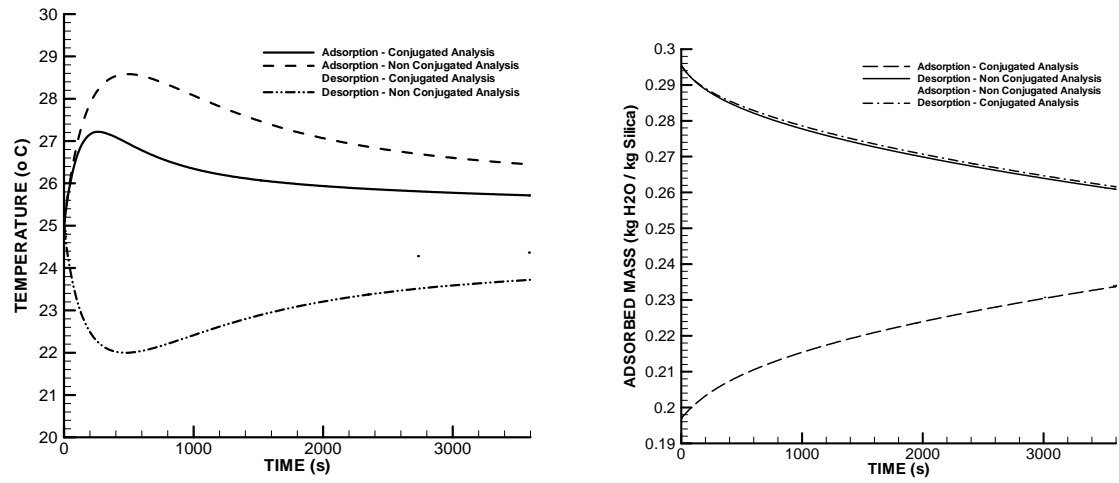


Figure 2. Comparison among conjugated and non conjugated approach results in silica mean temperature and mean adsorbed water mass

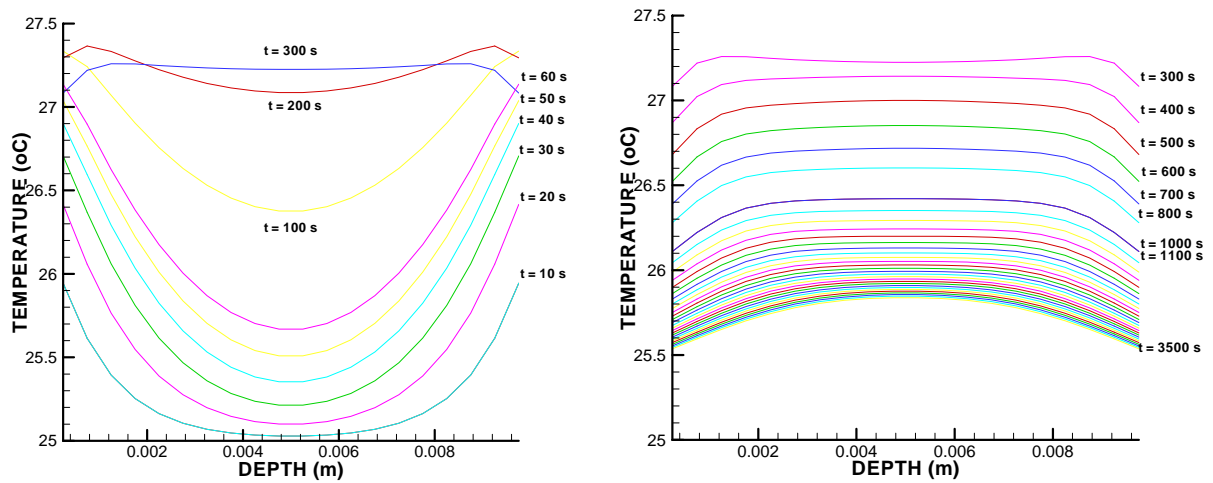


Figure 3. Temperature profile evolution in silica block during adsorption

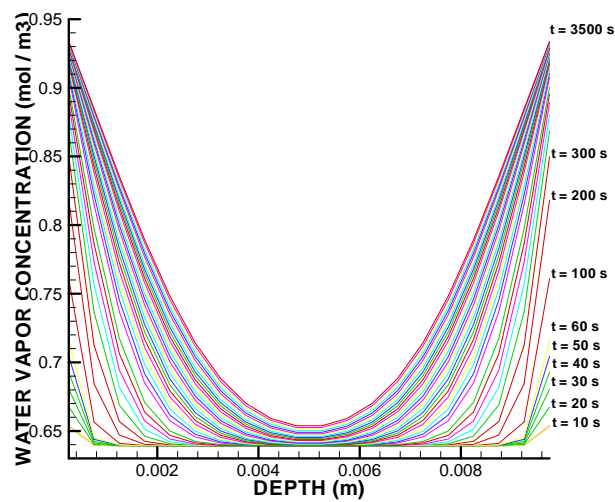


Figure 4. Free water vapor concentration profile evolution in silica block during adsorption

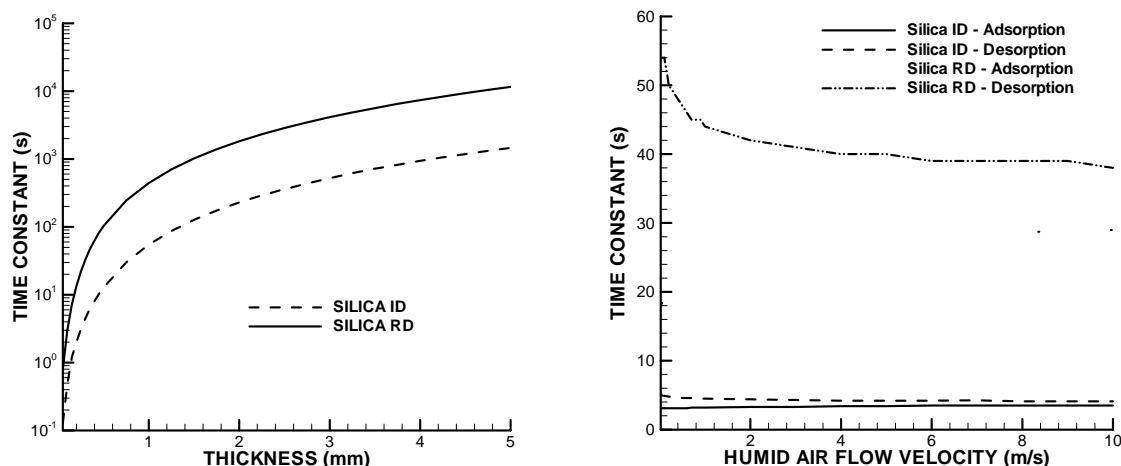


Figure 5. Time constant variation with silica film thickness and humid air flow velocity

Table 3. Conditions assumed at simulation cases on Fig. 5.

Simulation case parameters	Time constant – film thickness plot	Time constant - air uniform velocity plot
Length (mm)	10	10
Thickness (mm)	*	0.25
Humid air uniform velocity (m s^{-1})	0.5	*
Humid air flow temperature ($^{\circ}\text{C}$)	25	25
Silica initial temperature ($^{\circ}\text{C}$)	25	25
Air flow relative humidity (%)	75	50 or 75
Relative humidity (%) within voids in silica	50	50 or 75
Also insulated against heat and mass transfer on opposite face to exposed one		

(*) : parameter under investigation

4. Conclusions

As already pointed, mass transfer rates are practically insensible to problem approach (conjugated or not). Since mass transfer rates dictates transient duration, conjugated analysis is unnecessary in the present investigation on selecting silica gels for air humidity sensor construction.

The candidate material should possess high mass and thermal diffusivities, in order to faster reaching new equilibrium condition after any humid air change.

Sensor response time decreases with material porosity. On the other hand, lower porosity means less adsorption sites and worse sensibility, since variation of material electric properties is directly associated to interfacial electrochemical adsorbate-adsorbent interactions.

Focusing on transport phenomena, sensor material must possess low adsorption heat. Silica gel is a glassy and porous material, what makes it a powerful thermal insulator. Heat generation during adsorption accelerates mass diffusion. At desorption, heat absorption promotes silica temperature to decrease, which reduces water vapor mass diffusivity and displaces equilibrium relation towards adsorbed phase. So, sensor design must focus on desorption step. Otherwise, from the sensor functioning point of view, adsorption heat must be high. This parameter is linked to adsorbate-adsorbent affinity, which determinates sensor sensibility. So, adsorption heat target value can be found through a balance between undesired resultant cooling effects at desorption and aimed sensibility.

Specific heat is inversely proportional to thermal diffusivity. On the other hand, higher specific heat better limits undesirable temperature decrease at desorption critical step. So, specific heat optimal value passes through a maximum.

Considering the presented test results and analysis, in order to simultaneously achieve good sensibility and short response time, sensor must be built through a very thin silica RD coverage.

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