Handbook of Tables of Thermodynamic Parameters and Mass Transfer Coefficients of Wet Materials

by

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Contents

Annotation

The forms of moisture binding to materials are analyzed from the standpoint of binding energy; proceeding from fundamental concepts of the thermodynamics of irreversible processes, expressions for the chemical potential of mass transfer for various forms of binding are obtained in explicit form; extensive systematized experimental data for sorption and desorption isotherms (over 1800 isotherms) are presented, on whose basis new data on thermodynamic parameters and mass transfer coefficients for about 900 materials are obtained and their analysis is given.

The book is intended for a broad segment of scientific workers and engineering staff of chemical processing, heat engineering, food industry, and allied specialities.

Introduction

Modern development of the chemical industry and allied branches, in which new technological processes are designed alongside the intensification of those existent, imposes new requirements as to further development of the theory of heat and mass transfer and the accumulation of reference data without which engineering calculations are unfeasible.

Only a correct solution of the problems of heat and mass transfer of the substance in wet materials that are subjected to technological treatment will provide their high quality.

As a result of the investigations conducted by A. V. Luikov, it is established that mass transfer of the absorbed substance is determined by the form of its binding to the frame of a solid body. A different intensity of the energy of moisture binding to the material, alongside its structure that specifies the moisture and vapor motion inside the material, determined the kinetics of drying and moistening of materials.

The works of P. A. Rebinder, M. M. Dubinin, S. M. Lipatov, A. V. Dunamskii, Yu. L. Kavkazov, A. A. Rode, et al., have established basic principles of the binding of the absorbed substance to capillary-porous colloidal bodies and revealed properties of the bound moisture, i.e., have developed a modern theory of the forms of moisture binding to wet materials.

The moisture removal from the body involves a breaking of moisture binding to the body, on which a certain energy is spent. Therefore the forms of moisture binding should be classified by the principle of studying the intensity of binding energy.

By such a principle, P. A. Rebinder's scheme has been constructed that is taken as a basis in the current study.

Relying on P. A. Rebinder's theory, the study considers the forms of moisture binding to materials from the standpoint of the binding energy and uses the well-known assumptions that the chemical potential of mass transfer of the substance in wet materials is a factor of rate of the mass transfer processes.

Proceeding from fundamental concepts of the thermodynamics of irreversible processes and classical thermodynamics, expressions for the chemical potential of mass transfer for various kinds of binding are obtained in an explicit form, which makes it possible to determine the direction and limit of a spontaneous progress of the processes of mass transfer of the substance from one part of the system to another, since a spontaneous progress of the processes of interaction between different parts of the system is possible only in the direction of equalization of the factor of rate for all parts of the system. The attainment of the same values of this factor is the limit of a spontaneous progress of the process under these conditions and, therefore, is the equilibrium condition.

Extensive experimenal data for sorption and desorption isotherms were systematized and processed for calculating thermodynamic parameters and mass transfer coefficients in colloidal capillary-porous bodies.

It is known that thermodynamic parameters and mass transfer coefficients characterize the transfer rate and are the basis for calculating and controlling hydrothermal processes. They determine the ability of the material to absorb and release moisture in a hydrothermal process and account for the structural and mechanical changes occurred.

It is possible to present the following (incomplete) list of problem that cannot be solved without using thermodynamic parameters and mass transfer coefficients in colloidal capillary-porous bodies [141]: automatization of technological processes involving the hydrothermal treatment of nonmetallic materials, processing of experimental data on mass transfer in a dimensionless form, application of analytical solutions of heat and mass transfer to practical calculations, calculation of the quantity of mass that has passed from one body to another on their direct contact, design of thermal devices and apparatus associated with the kinetics of heat and mass transfer, analysis of the forms of moisture binding to the material, etc.

The foregoing demonstrates how important it is to have data on thermodynamic parameters and mass transfer coefficients for colloidal capillary-porous bodies. The accumulation of such data and their study is among the most important tasks of the science of heat and mass transfer.

The creation of an artificial climate is of special importance for the conducting of technological processes in such industries, where hygroscopic materials serve as a raw material to be treated. In these industries, the relative humidity and temperature of air in production areas determine the grade of products and therefore, the conducting of the technological process.

Thus, for example, air conditionings is needed in manufacturing fibers (especially man-made) at textile factories, in color printing in polygraphy, in storing leather at shoe factories, for providing a normal tableting process at pharmaceutical plants, in manufacturing and processing film in the film industry, at tobacco and tea factories, in

producing cocoa powder and caramel at confectionary factories, at bread-baking plants, etc.

Regardless of the industry, optimum conditions for the best conducting of the technological process should be determined by technologists of a given production. The pinpointing of optimum conditions through experimental selection of combinations of the temperature and relative humidity of air in the area, which provide an appropriate specific moisture content of the treated material, is a fairly complicated problem.

This problem can be simplified using the value of the chemical potential of mass transfer of the substance in the hygroscopic region, whose gradient is exactly a driving force of mass transfer.

In the current study, such procedure is proposed for calculating optimum parameters of air for conditioning in production areas, public buildings (book depositories, archives, museums, etc.) and for storing hygroscopic materials in warehouse.

Having available data on the average specific isothermal mass capacity for various materials, which are presented in the study, it is possible to give recommendations for selecting individual components that enter into the system of contacting bodies or structures of wet materials and to choose the type and mode of the technological process with an objective of obtaining an optimum moisture-accumulating ability of the material, i.e., to produce the material with preset hygroscopic properties.

1

Forms of Moisture Binding to Materials

1.1 Classification of Forms and Energy of Moisture Binding to Materials

The most sound method of evaluation of the forms of moisture binding and their classification is the use of the energy of moisture binding to wet materials [1]. The energy of moisture binding to wet materials (the adsorption potential ε) was for the first time established by Polanyi [2–5].

It is known that forces of mutual attraction manifest themselves not only between molecules of the same substance but also between gas or vapor molecules and the surface of any solid body.

By Polanyi's theory of adsorption, whenever the vapor temperature is subcritical, the pressure of the adsorbate vapor in the immediate vicinity of the adsorbent surface is so high that its condensation (liquefaction) occurs, and the adsorbent surface is covered with a liquid layer.

If the temperature is fairly far (in the direction of decreasing values) from critical, the mass of the compressed vapor is insignificant in comparison with the mass of the adsorbate liquid. In this case, the liquid volume condensed on the surface of unit mass of the adsorbent can be approximately expressed as a quotient obtained when the adsorption u is divided by the liquid density ρ at an experimental temperature. Evidently, the liquid volume V is directly proportional to the average distance x of the liquid sur-

face from the adsorbent surface, i.e., $V = sx$, where s is the specific surface of unit mass of the adsorbent. Since real adsorbents are generally porous bodies with the specific surface unknown, there is no possibility of conversion from the volume of the liquid layer on the adsorbent surface to its thickness.

Therefore, the problem of Polanyi's theory was that of calculating the dependence $\varepsilon = f(x)$ from experimental data.

By definition, the adsorption potential ε expresses the work done by adsorption forces to move one mole of vapor from the volume, where adsorption forces are inoperative, over the distance x from the adsorbent surface. It is very easy to calculate this work of transfer to the surface of the liquefied layer, where the distance x represents its average thickness. Two assumptions are made here:

1) vapor in the gas phase obeys the ideal gas laws,

2) liquid in the adsorbed layer is incompressible.

The surface of the adsorbed liquid layer borders on the saturated vapor, whose pressure p_s is determined by an experimental temperature. Let v_s denote the volume of one mole of the saturated vapor at the pressure p_s and, correspondingly, v_u denote the volume of one mole of vapor at the equilibrium pressure in the gas phase p_u. The work done by adsorption forces at a constant temperature to move one mole of vapor from the gas phase to the surface of the liquid layer is equal to the work of its isothermal compression from the volume v_u to the volume v_s. This work represents the adsorption potential corresponding to the distance x, i.e.,

$$\varepsilon = RT \ln \frac{v_u}{v_s}, \tag{1.1}$$

where R is the universal gas constant and T is the absolute temperature.

Since at T = const the volumes are inversely proportional to pressure,

$$\varepsilon = RT \ln \frac{p_s}{p_u}. \tag{1.2}$$

It should be noted that corrections to the value of the adsorption potential that account for compressibility of the adsorbed liquid and for the departure of vapor from the ideal gas laws are insignificant, of the order of several percent [6], therefore, the error introduced by these assumptions in calculations of the adsorption potential is not great.

The calculated value of the potential corresponds to a certain magnitude of the vapor adsorption, when the average thickness of the liquid layer is x. Because the specific surface of the adsorbent is not known, the calculation of the distance x in absolute units is impracticable. Therefore the distance is expressed not in units of length but in units of volume of the liquid layer, which are directly proportional to length and are evidently different for various adsorbents (for the reason of dissimilar specific surface).

For each point of the measured isotherm it is possible to calculate corresponding values of ε and V. According to Polanyi's theory, the adsorption potential is approximately temperature-independent, consequently, $\varepsilon = f(V)$ is identical for all temperatures.

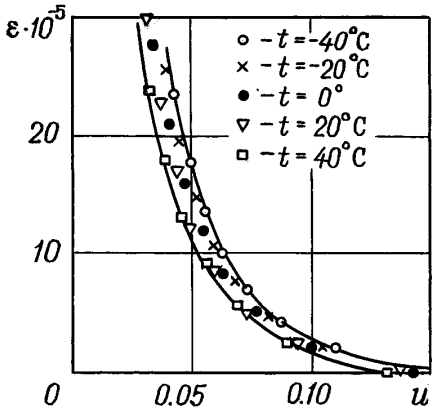

Fig. 1.1. Adsorption potential of mass transfer vs. specific moisture content at constant temperature for cork, γ = 200 kg/m^3.

Therefore, the $\varepsilon = f(V)$ curve is called a characteristic equation of adsorption. Figure 1.1 presents characteristic curves predicted from the sorption isotherms for cork with a specific weight of 200 kg/m^3 [7]. It is seen from Fig. 1.1 that, although the isotherms are markedly different (the temperature varies from –40 to 40°C), all points lie in a fairly narrow region, which is an experimental validation of the basic assumption of Polanyi's theory. According to [6], the potential theory is applicable to both monomolecular and polymolecular adsorption. It is the only theory of physical adsorption that quantitatively describes adsorption at sharply heterogeneous surfaces.

It is shown in [8] that the basic assertion of the potential theory about the existence of a characteristic curve that is invariant with respect to temperature does not require an identification of the adsorption phase with the free liquid. The condition for existence of unique, temperature-independent characteristic curve can be written in the form

$$\left(\frac{\partial \varepsilon}{\partial T}\right)_{uv} = 0\,, \tag{1.3}$$

where v is the specific volume of the adsorbed substance.

It is known [8] that in all cases the adsorption potential ε decreases with an increase in uv, i.e., $\partial\varepsilon/\partial uv < 0$. At u = const,

$$\frac{\partial \varepsilon}{\partial uv} = \frac{1}{u}\left(\frac{\partial \varepsilon}{\partial v}\right)_u = \frac{\left(\dfrac{\partial \varepsilon}{\partial T}\right)_u}{u\left(\dfrac{\partial u}{\partial T}\right)_u} < 0.$$

Assuming that the specific volume of the adsorption phase increases with temperature and therefore $(\partial v/\partial T)_u > 0$, we obtain $(\partial\varepsilon/\partial T)_u < 0$. The derivative, entering into this inequality, represents a variation in the differential entropy of adsorption ΔS.

Indeed [9], since the adsorption potential is a change in the free energy during the isothermal transfer of a mole of vapor from the volume, where adsorption forces are inoperative, to the adsorbent that is in equilibrium with the gas phase at the pressure p_u, then, by the Gibbs–Helmholtz equation for differential heat of adsorption at T = const

$$q_d = \varepsilon - T\left(\frac{\partial \varepsilon}{\partial T}\right)_u = \varepsilon - T\Delta S\,. \tag{1.4}$$

Therefore, the fulfillment of the basic equation of the potential theory (1.3) requires that the inequality $\Delta S < 0$ be satisfied. For $\Delta S < 0$, the inequality $q_d > \varepsilon$ is valid.

A temperature variation in ε at a constant rate of adsorption results from the thermal expansion of the compressed liquid laver at the adsorbent surface in desorption, which is accompanied by the energy absorption. Conversely, with a subsequent adsorption of fresh vapor portions, the liquid compression in the adsorbed layer increases, which leads to the heat release. Therefore the differential heat of adsorption is larger than the adsorption potential by the magnitude of this thermal effect.

Clearly, before adsorption the vapor molecules could freely move in the space of three dimensions, and therefore adsorption should involve a decrease in entropy ($\Delta S < 0$). As shown hereafter, each spontaneous process involves a decrease in the free energy of the system ($\Delta F < 0$). Since ΔS and ΔF are negative, the quantity ΔU — a variation in the internal energy during adsorption — should also be negative, by the Helmholtz equation. Hence, all adsorption processes are exothermic.

It is seen from Eq. (1.2) that, if $p_s = p_u$, i.e., in the presence of free moisture in the material, $\varepsilon = 0$. With removal of the entire free moisture and with a subsequent gradual decrease in the specific moisture content of the material u, the energy of moisture binding to the material increases continuously in the region of the adsorptively bound moisture and afterward increases stepwise in the transition to the chemically bound moisture, reaching a maximum at a minimum pressure of dissociation of a relevant hydrate. The indicated situation is consistent with experimental data of V. M. Kazanskii [10–14]. V. M. Kazanskii designed a recording electrocalorimetric device and developed the procedure of its application to determining the specific heats of moisture evaporation from capillary-porous colloidal bodies at various specific moisture contents of the latter.

Using this calorimetric device, experiments were conducted for determining the specific heats of evaporation of moisture adsorbed by various materials: silica gel MSM (fractions with a grain size of 0.25–0.5 mm), sand of cement rock of the same fractions, the Pyzhevsk bentonite and the Cherkassk mountain cork, and sand of silica gel KSM, KSK-3, and E. A combined analysis of the specific heats of moisture evaporation, energy diagrams, and curves of drying kinetics manifests that the specific heat of isothermal evaporation of moisture from capillaries of the porous structure of a body, especially of the adsorbed moisture of poly- and monomolecular adsorption, differs from the heat of phase transition of the free liquid to vapor. Here, a slight increase in the specific heats of evaporation for the capillary moisture is linked with a dissimilar

curvature of menisci in the adsorbent pores. An appreciable increase in the heats of evaporation of the adsorbed moisture of poly- and monomolecular adsorption is caused by the intensity of the energy of its binding to the solid phase of a body.

Four forms of moisture binding to materials are discerned (in the order of decreasing binding energy) [1]: chemically, adsorptivity, osmotically and capillary-bound moisture.

According to Academician P. A. Rebinder's scheme, the four forms of moisture binding to materials make up three classes, namely:

1. Chemical binding (stoichiometric), which is a binding in exact quantitative ratios. Here belongs ionic and molecular binding (the water of hydration).

2. Physico-chemical binding, which is a binding in various, not strictly definite ratios. To this class belong the adsorptively and osmotically bound moisture and moisture, immobilized in the formation of a body structure.

3. Mechanical binding, which is water retention in indefinite ratios (the binding in microcapillaries and macrocapillaries, and the wetting moisture).

The energy of chemical binding corresponds in magnitude with the heats of chemical reactions (tens of large calories per mole) [15].

The energy of molecular interactions is commensurable in magnitude with the specific heat of evaporation.

Below, characteristics of each form of moisture binding to materials are given. Since in the process of even deep drying of wet materials the chemically bound moisture is not removed, this question is not considered in detail in the current study.

1.2 Chemically Bound Moisture

The chemically bound moisture is taken to mean the hydrate water, bound in the form of hydroxyl ions, and the water of molecular compounds such as crystalline hydrates, which is bound much more weakly. The condition of formation of the ionic binding is a chemical reaction (hydration). A new body is formed here. Water as such disappears and becomes part of a new substance.

The binding is broken only as a result of the chemical interaction (sometimes as a result of calculation).

In the case of molecular binding, where crystallization from the solution occurs (a crystalline hydrate is formed), the body sharply changes its properties. Here, water enters into the crystal structure. The binding is broken with calcination.

At a certain temperature, the hydrate can exist only at a strictly definite pressure of water vapor in the surrounding space. This pressure is called dissociation pressure of the hydrate. It is individual for each hydrate and increases with temperature. Thus, for example, at 30^{o}C, the dissociation pressure of Glauber salt $Na_2SO_4 \cdot 10H_2O$ is 27 mm Hg, of copper sulfate $CuSO_4 \cdot 5H_2O$, 12.5 mm Hg and of barium chloride, as low as 4 mm Hg [16]. If the hydrate is in the air, where the pressure of water vapor is lower

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