

Influence of thermodynamic mechanism of interfacial adsorption on purifying air-conditioning engineering under intensification of electric field

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Abstract As a kind of mass transfer process as well as the basis of separating and purifying mixtures, interfacial adsorption has been widely applied to fields like chemical industry, medical industry and purification engineering in recent years. Influencing factors of interfacial adsorption, in addition to the traditional temperature, intensity of pressure, amount of substance and concentration, also include external fields, such as magnetic field, electric field and electromagnetic field, etc. Starting from the point of thermodynamics and taking the Gibbs adsorption as the model, the combination of energy axiom and the first law of thermodynamics was applied to boundary phase, and thus the theoretical expression for the volume of interface absorption under electric field as well as the mathematical relationship between surface tension and electric field intensity was obtained. In addition, according to the obtained theoretical expression, the volume of interface absorption of ethanol solution under different electric field intensities and concentrations was calculated. Moreover, the mechanism of interfacial adsorption was described from the perspective of thermodynamics and the influence of electric field on interfacial adsorption was explained reasonably, aiming to further discuss the influence of thermodynamic mechanism of interfacial adsorption on purifying air-conditioning engineering under intensification of electric field.

Keywords: Interfacial adsorption; Thermodynamics; Gibbs; Purifying air-conditioning engineering; Electric field

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1 Introduction

Purifying air-conditioning engineering is an important issue which guarantees that all parameters in clean operating department are under control. It requires the controlling of air temperature, humidity, dust, bacteria and concentration of hazardous gases, air flow distribution and pressure distribution in different regions of operating rooms and subsidiary rooms, thus to finally create a sterile environment in these rooms and reduce infection probability as well as improve the achievement ratio of operation. Generally, a purifying air-conditioning engineering is composed of air filter, circulating fan, surface air cooler, heating and humidifying units and other basic equipment. It filters the air through low efficient, medium efficient and highly efficient filter units to achieve required cleanness. However, such kind of filter unit has a short service life and high costs; moreover, its filtering effect needs further improvement. With the development of science and technology, interfacial adsorption has been extensively applied in environment and chemical industry production, such as purification of sewage, removal of toxic substances in air and separation or purification of mixtures, etc. [1–3]. Common methods of changing interfacial adsorption include changing temperature, intensity of pressure and types of adsorbent, increasing surface active agent, performing surface modification or applying physical field (such as electric field, magnetic field, etc.). Through these methods, adsorption efficiency can be enhanced or weakened, which can further satisfy the industrial demands [4–6]. However, during the operational process of these methods, such as using active carbon or silica gel to adsorb nitrogen or hydrogen, the adsorption temperature should be controlled at the boiling temperature of nitrogen or hydrogen, so that obvious adsorption effect can be realized because it is difficult for the adsorbents to absorb nitrogen or hydrogen at normal temperature. Moreover, the addition of surface active agent at the interface can affect the purity of products. By comparison, applying electric field can control the amount of adsorbed substances without introducing impurities; moreover, adsorbents can be reused which can save resources as well as costs, thus it is the optimal method which can be applied in chemical industry production [7–9].

Therefore, starting from the point of thermodynamics, this study analyzed the functional mechanism of electric field on interfacial adsorption. Main thermodynamic models used to describe interfacial adsorption at present are Gibbs adsorption model [10–11] and Guggenheim adsorption model [12]. In 1873–1878, American physicist and chemist Gibbs con-

cluded the classical thermodynamics and put forward the Gibbs adsorption isotherm as well as relevant models. In 1920's, a Chinese scientist Fu Ying studied adsorption phenomena and discovered the multilayer adsorption theory of aqueous adsorption; he also put forward methods of using the slope of the initial segment of adsorption isotherms to calculate adsorption standard free energy changes. Both models regard the system as a composition of two bulk phases and one interface. However, in Gibbs adsorption model the interface is regarded as a two-dimensional surface which has area but no volume; while in Guggenheim adsorption model, the interface is regarded as a boundary phase which has area as well as volume. Although the Guggenheim adsorption model is more similar to the actual system, it has a complex computational process and for that reason is not widely applied in engineering. On the contrary, Gibbs adsorption model reflects the relationship among interfacial tension, concentration and adsorbing capacity from the aspect of thermodynamics; it is the most basic formula used to analyze interfacial adsorption and the operation is simple; moreover, its results are close to the reality. Therefore, the Gibbs adsorption model was selected to study the influence of thermodynamic mechanism of interfacial adsorption on purifying air-conditioning engineering under intensification of electric field.

2 Definition of interfacial tension

Interfacial tension [13], also known as the surface tension of liquid, is the interfacial tension between liquid and air. Strictly speaking, the surface refers to the interface between liquid or solid and its saturated vapor; but customarily, the interface between liquid (solid) and air is called the surface of liquid (solid). Common interfaces include gas-liquid interface, gas-solid interface, liquid-liquid interface, liquid-solid interface, and solid-solid interface. The interfacial force between a kind of liquid and another kind of immiscible liquid is called the interfacial tension between two liquid phases. The interfacial force between a kind of liquid and a solid is called the interfacial tension between liquid and solid phases. The surface tension of liquid is also called the free energy of the liquid surface [14]. Taking the gas-liquid interface as an example, the attractive force from internal molecules of liquid is greater than that from the internal molecules of gas phase, thus the stress on interface is unbalanced and net attractive force is produced and interfacial tension is formed. The stress on molecules of interface is verti-

cal to the interface and points to the inside of liquid phase. The surface of liquid seems to have an elastic membrane which can allow liquid to be contractive. Interfacial tension is defined as the contraction force per unit length of straight line. Figure 1 shows a metal frame with a movable end where the tip is dipped with soap water. Suppose a force is applied to the end and the tip is pulled slowly rightwards; because the air bubble film on the end has two surfaces, the force applied on the liquid can be described as $F = 2\gamma l$, where γ refers to the interfacial tension. The work consumed during the process of liquid molecules moving to the interface is

$$dW_s = Fdx = \gamma dA . \quad (1)$$

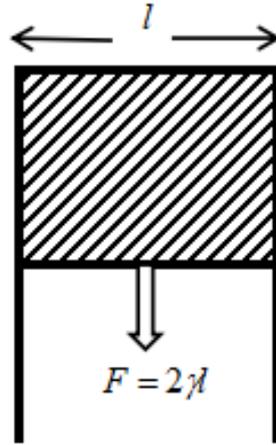


Figure 1: Interfacial tension and surface work.

Equation (1) indicates that, if the interfacial tension is big, then more energy should be consumed to increase the area of interface. From the law of thermodynamics we can know that, at constant temperature and under isopiestic pressure, the interfacial tension is equal to the surface energy of unit area, $\gamma = \partial G / \partial A_{T,p,n}$. In the equation, G refers to the Gibbs free energy and A is interfacial area, whereas the equation is the thermodynamic definition of interfacial tension. Moreover, all spontaneous thermodynamic states in nature are accompanied by changes of Gibbs free energy. Therefore, the solution interface can turn into a spherical shape automatically. Surface tension varies with temperature. Generally speaking, with the increase of temperature, surface tension decreases, and when the temperature

approaches the critical temperature, the surface tension decreases to zero. Surface tension of binary system is also related to components. Different pure substances have different interfacial tension. If a pure substance is added with a substance which can reduce the interfacial tension of pure solution, adsorption can occur on the interface of solution. The decrease of interfacial tension is the primary cause of interfacial adsorption.

3 Gibbs interface

Thickness of the interface layer of two contacted phases only equals to several molecular diameters, and only few molecules are in this region. When the specific value between superficial area and volume is high, the influence of surface effect on system property is significant. The property of interface layer changes from typical α phase to typical β phase (Fig. 2), which is uneven. The interfacial behavior analysis of this study on a single molecule tends to develop to general analysis reflecting the interface. Due to changes of the interaction between molecules, the average energy of molecules in interface zone is different from that of the molecules in a phase. The σdA refers to the work needed for the increase of interfacial area and σ refers to interfacial tension. Residual force fields are formed due to the asymmetric stress of molecules in superficial layer, and these unbalanced force fields can have absorption on surrounding mediums, thus the surface can adsorb substances that can reduce the surface energy. The adsorption which can make the surface concentration be bigger than the internal concentration of liquid is called the positive adsorption which can be generated by molecules migrating from the main phase to the interface to reduce interfacial energy or surface tension. In order to quantitatively describe the surface adsorption of liquid, the adsorbing capacity is introduced to represent the adsorption degree [15].

Concentration of substances on the adsorption surface changes continuously without a clear interface. In Gibbs thermodynamic model of adsorption, the interface is considered as a two-dimensional surface phase without thickness and the volume is negligible while other thermodynamic quantities are not zero. As shown in Fig. 2, the region above line A refers to the pure α phase and the region below line B refers to pure β phase. The area between A and B is the interface region where components and properties change instantaneously. According to the concept of surface phase of Gibbs, a two-dimensional geometric surface S with no thickness between

A and B is selected, which is called the surface phase σ . The symbol A refers to the area of surface phase and components of surface phase are described by surface excess amount n_i^σ ($i = 1, 2$). The n_i^σ is obtained by the actual mole number of i component in AB region subtracting the mole number of i component in phase boundary region AB when imaginary α and β phases evenly extend to the surface phase S according to bulk phase. Surface excess amount can be positive, negative or zero, which is related to the position of imaginary interface S.

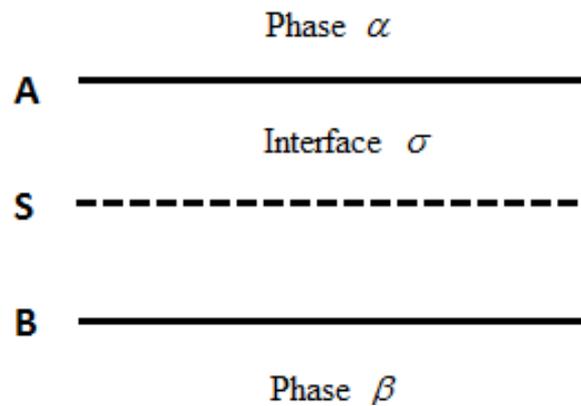


Figure 2: Illustration of the interface between two phases of Gibbs model.

4 Effect of electric fields on thermodynamics mechanism of adsorption

The adsorption thermodynamic model mainly refers to adsorption isothermal equation, which is used to describe the adsorption isotherm. The so called adsorption isotherm refers to the relation curve between equilibrium absorption capacity, Γ_e , of adsorbate on adsorbent and the equilibrium concentration, C_e , of adsorbate in liquid phase. The widely applied adsorption isothermal equations are Freundlich and Langmuir equations, in which the Freundlich equation is an empirical equation while the Langmuir equation is established on the basis of thermodynamic equilibrium theory.

Different factors can cause different changes of interface compositions in the interface layer. For example, substances transfer from the bulk phase to the interface or substances in the interface phase enter inside the bulk

phase. Therefore, the concentration in interface can increase as well as decrease. As shown in Fig. 3, the movement of these molecules explains the interfacial adsorption. Generally speaking, adsorption is the increase of substances in the interface phase; the amount of substances adsorbed on interface is described by surface excessive amount, Γ , i.e., the amount of adsorbed substances in unit area. When the temperature stays the same, the functional relationship between the surface excessive amount and intensity of pressure $\Gamma = pT$ or the functional relationship between surface excessive amount and concentration $\Gamma = cT$ is called adsorption isotherm (where p – pressure, c – condensation, T – temperature). Adsorption isothermal equations contribute to a better understanding of interfacial adsorption as well as the prediction of interfacial adsorption amount. Besides, the application of adsorption isotherms is extensive. For example, properties of interface and pores can be studied according to adsorption isotherms and specific surface area and pore size distribution can be calculated, etc. The adsorption caused by physical interaction is called the physical adsorption; a large number of adsorption isotherms are obtained through experiments according to physical conditions.

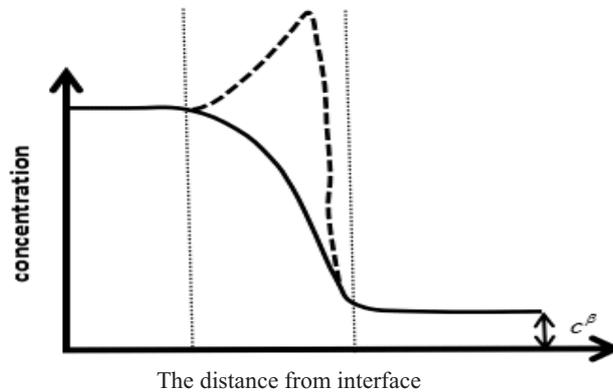


Figure 3: Change of concentration at the interface.

In general, the existence of interface can affect the thermodynamic parameters and properties of the whole system. To consider the thermodynamic properties of a system which contains an interface, the system is divided into three parts: two bulk phases whose volume is V^α and V^β , respectively, and an interface σ . In Gibbs adsorption model, two bulk phases are separated by a two-dimensional surface without thickness, i.e., Gibbs interface.

In Gibbs adsorption model, except volume, all other extensive quantities, such as internal energy, U , entropy, S , and the amount of substance, n , can be described as three parts, which form two bulk phases and one interface;

$$V = V^\alpha + V^\beta, \quad (2)$$

$$U = U^\alpha + U^\beta + U^\sigma, \quad (3)$$

$$S = S^\alpha + S^\beta + S^\sigma, \quad (4)$$

$$n_i = n_i^\alpha + n_i^\beta + n_i^\sigma. \quad (5)$$

Assume u^α , and u^β are internal energies of the unit volume of two bulk phases, respectively, and c_i^α , and c_i^β are concentrations of component, i , in two bulk phases, respectively, then the total amount of internal energies and substances of interface phase can be expressed as $U^\sigma = U - u^\alpha V^\alpha - u^\beta V^\beta$ and $n_i^\sigma = n_i - c_i^\alpha V^\alpha - c_i^\beta V^\beta$. Surface excessive amount is defined as

$$\Gamma = \frac{n_i^\sigma}{A}. \quad (6)$$

4.1 Traditional Gibbs adsorption isothermal equation

Under equilibrium state, the fundamental equation of thermodynamics when the system has a surface energy is expressed as $dU = TdS - pdV + \sum_i \mu_i dn_i + \gamma dA$, where all amounts in the equation are the total amount in the actual system. In Gibbs model, the internal energy of α phase and β phase is $dU^\alpha = TdS^\alpha - pdV^\alpha + \sum_i \mu_i dn_i^\alpha$ and $dU^\beta = TdS^\beta - pdV^\beta + \sum_i \mu_i dn_i^\beta$, respectively, and on the basis of Eqs. (2), (4) and (5), can be deduced

$$dU^\alpha = TdS^\alpha + \sum_i \mu_i dn_i^\alpha + \gamma dA. \quad (7)$$

The Gibbs function in imaginary surface phase of Gibbs model is defined as $G^\sigma = U^\sigma - TS^\sigma - \sigma A$ and the combination of its perfect differential and can obtain the differential equation $dG^\sigma = -S^\sigma dT + \sum_i \mu_i dn_i^\sigma - Ad\sigma$. On the basis of $G = \sum_i \mu_i n_i$, the Gibbs-Duhem function in σ phase is

$$S^\sigma dT + Ad\sigma + \sum_i n_i^\sigma d\mu_i = 0. \quad (8)$$

Under the same temperature and pressure, the following equation can be obtained

$$d\sigma = - \sum_i \left(\frac{n_i^\sigma}{A} \right) d\mu_i^\sigma = - \sum_i \Gamma_i d\mu_i^\sigma, \quad (9)$$

where $\frac{n_i^s}{A}$ refers to the surface excessive amount Γ_i , and the equation is the fundamental form of Gibbs adsorption equation.

The value of surface excessive amount is related to the position selection of interface S, and for any appointed component (such as solvent), there is one and only one position obeying $n_1^s = 0$. Taking two components as an example and selecting S to make $\Gamma_1 = 0$, then the above equation can be written as

$$d\sigma = -\Gamma_2 d\mu_2 \quad \text{or} \quad \Gamma_2 = - \left(\frac{\partial \sigma}{\partial \mu_2} \right)_T . \quad (10)$$

This is the expression of Gibbs adsorption equation in two components. According to the above analyses we discover that, the Gibbs adsorption isothermal equation can be obtained by the combination of Gibbs-Duhem function and the analysis methods of Gibbs surface phase, and the Gibbs adsorption isothermal equation under the effect of external field can be obtained by the combination of Gibbs-Duhem function and analysis methods of Gibbs surface phase under the external field effect.

4.2 Gibbs adsorption isothermal equation under the effect of electric field

On the basis of ideal Gibbs adsorption model theory and according to the energy axiom, the differential of any kind of energy can be expressed as the product of a basic intensive quantity, X , and a conjugated basic extensive quantity, x , differential, i.e., $dW = Xdx$, where, X , represents a kind of field quantity. The expression can also be expressed by work because the latter can transmit or change the internal energy of the whole system. For example, the analogy of surface work, $dW = \gamma dA$, can be the surface energy. Under the effect of electric field, substances can be polarized and the polarization degree can be represented by polarization energy. For the substance with even phases, the expression of its polarization energy under the effect of electric field can be expressed as

$$dW_c = EdP' , \quad (11)$$

where E refers to the applied electric field intensity and P' is the total electric dipole moment of substances.

According to the first law of thermodynamics, changes of internal energy of the system are simply caused by working or thermal transmission. Therefore, except thermal transmission, volume work, particle changes, and

surface energy – when the applied electric field acts on a three-phase system, the total internal energy of the system can be generalized as following according to the first law of thermodynamics and energy axiom [16]:

$$dU = TdS - p^\alpha dV^\alpha - p^\beta dV^\beta + \sum \mu_i dn_i + \gamma dA + EdP' . \quad (12)$$

For plane interface, $p^\alpha = p^\beta$, thus the above equation can be transformed as

$$dU = TdS - pdV + \sum \mu_i dn_i + \gamma dA + EdP' . \quad (13)$$

As for the intensive quantity, the chemical potential and electric field intensity of, such as temperature and one component, are uniform under the equilibrium state. Applying electric field in two-phase system, their internal energy can be expressed as

$$dU^\alpha = TdS^\alpha - pdV^\alpha + \sum \mu_i dn_i^\alpha + EdP'^\alpha , \quad (14)$$

$$dU^\beta = TdS^\beta - pdV^\beta + \sum \mu_i dn_i^\beta + EdP'^\beta . \quad (15)$$

Combining with the function $P' = P'^\alpha + P'^\beta + P'^\sigma$, $V = V^\alpha + V^\beta$, $U = U^\alpha + U^\beta + U^\sigma$, $S = S^\alpha + S^\beta + S^\sigma$ and $n_i = n_i^\alpha + n_i^\beta + n_i^\sigma$, the expression of internal energy of interface phase is

$$dU^\sigma = TdS^\sigma + \sum \mu_i dn_i + \gamma dA + EdP'^\sigma . \quad (16)$$

The above equation indicates that, the interface phase can be taken as a normal system and described directly using fundamental thermodynamic relation, except that the volume is neglected in Gibbs adsorption model. The Gibbs adsorption function describes the relationship between concentration changes and interfacial tension changes of a component in the interface phase. Initially, Gibbs deduces the change relation of interfacial tension and concentration of component through thermodynamic method, and on the basis of this Guggenheim and Adam [17] improved the method and put forward a simpler and more reliable method. Gibbs adsorption equation is the third basic equation of interfacial chemistry. For a multicomponent system, if its temperature, chemical potential, interfacial tension, and electric field intensity remain the same and its internal energy, entropy, superficial area and intensity of polarization increase from zero to a certain value, then the expression of total internal energy of interface phase is

$$U^\sigma = TS^\sigma + \sum \mu_i n_i^\sigma + \gamma A + EP'^\sigma . \quad (17)$$

The above equation is a generalized equation and through differential of the equation, the following function can be obtained:

$$dU^\sigma = TdS^\sigma + S^\sigma dT + \sum \mu_i dn_i^\sigma + \sum n_i d\mu_i^\sigma + \gamma dA + Ad\gamma + EdP'^\sigma + P'^\sigma dE . \quad (18)$$

Combining function (16) and (18), the Gibbs-Duhem function which has the interface phase under the influence of electric field can be:

$$S^\sigma dT + \sum n_i^\sigma d\mu_i + Ad\gamma + P'^\sigma dE = 0 . \quad (19)$$

For a multicomponent system, the total electric dipole moment can be expressed as $P' = \sum n_i^\sigma V_{m,i} P_i$, where $V_{m,i}$ refers to the partial molar volume, and P_i is the dipole moment of unit volume of i substance, which is also called the intensity of polarization, and its relation with electric field intensity is $P_i = \varepsilon_0(\varepsilon_{r,i} - 1)E$. Therefore, the polarization energy of dielectric medium in the system under the effect of electric field can be expressed as $P' dE = \sum n_i^\sigma d \left[\frac{1}{2} \varepsilon_0(\varepsilon_{r,i} - 1)E^2 V_{m,i} \right]$, where ε_0 and $\varepsilon_{r,i}$ are dielectric constant and relative dielectric constant in vacuum respectively. The equation is then substituted into Eq. (19) and under the condition of steady temperature the generalized Gibbs-Duhem under the effect of electric field can be simplified to:

$$\sum n_i^\sigma d\mu_i + Ad\gamma + \sum n_i^\sigma d \left[\frac{1}{2} \varepsilon_0(\varepsilon_{r,i} - 1)E^2 V_{m,i} \right] = 0 . \quad (20)$$

The above equation indicates that the interfacial adsorption not only includes the adsorption caused by changes of interfacial area, but also includes the effect of external electric field on adsorption. Combining Eq. (21) and definition of surface adsorption capacity, the Gibbs adsorption equation under the effect of electric field can be obtained:

$$d\gamma = - \sum \Gamma_i d \left[\mu_i + \frac{1}{2} \varepsilon_0(\varepsilon_{r,i} - 1)E^2 V_{m,i} \right] . \quad (21)$$

The above equation is the expression of Gibbs adsorption isothermal equation under the effect of electric field, which indicates the relationship between interfacial tension and adsorption capacity and electric field intensity [18]. When there is no extra electric field, the equation can be simplified as $d\gamma = -\Gamma_i d\mu_i$, which is the traditional Gibbs adsorption isothermal equation.

5 Application of Gibbs adsorption isothermal equation in two-component solution under the effect of electric field

In order to better understand the Gibbs adsorption isothermal equation under the effect of electric field, the simplest two components are taken as an example, which are solvent 1 and solute 2, respectively. Because the definition of surface adsorption capacity is related to the position of Gibbs interface, generally speaking, the position where the adsorption capacity of solvent 1 is zero [19], i.e., $\Gamma_1 = 0$ is defined as the Gibbs interface, then the Gibbs adsorption function of two components is

$$d\gamma = -\Gamma_2^1 d \left[\mu_2 + \frac{1}{2} \varepsilon_0 (\varepsilon_{r,2} - 1) E^2 V_{m,2} \right]. \quad (22)$$

According to the equation, the interfacial excessive amount of component 2 at Gibbs interface is

$$\Gamma_2^1 = - \left[\left(\frac{d\gamma}{d\mu^2} \right)^{-1} + \left(\frac{1}{1/2\varepsilon_0 (\varepsilon_{r,2} - 1) V_{m,2}} \frac{d\gamma}{dE^2} \right)^{-1} \right]^{-1}. \quad (23)$$

The chemical potential expression of ideal solution is $\mu_2 = \mu_2^0 + RT \ln(c_2/c_0)$, where c_0 refers to the concentration of ideal solution and R, T are the gas constant and absolute temperature, respectively. At constant temperature, the chemical potential is substituted into the above equation and the following equation can be obtained:

$$\Gamma_2^1 = - \left[\left(\frac{1}{RT} \frac{d\gamma}{d \ln c_2} \right)^{-1} + \left(\frac{1}{1/2\varepsilon_0 (\varepsilon_{r,2} - 1) V_{m,2}} \frac{d\gamma}{dE^2} \right)^{-1} \right]^{-1}. \quad (24)$$

If there is no extra electric field, then the second item of Eq. (24) is zero and the adsorption equation turns into the classic Gibbs adsorption equation $\Gamma_2^1 = -\frac{1}{RT} \frac{d\gamma}{d \ln c_2}$. If solvent is increased and the surface energy is decreased, then the surface tension decreases with the increase of concentration, i.e., $d\gamma/dc_2 < 0$; and the adsorption capacity is positive, $\Gamma_2^1 < 0$, i.e., positive adsorption and substances gather at interface. Otherwise, if the surface tension increases with the increase of concentration, $d\gamma/dc_2 < 0$, then the adsorption capacity is negative, $\Gamma_2^1 < 0$, i.e., negative adsorption and the

adsorption capacity of substances at the interface is negative. If the external field is applied, then the second item of Eq. (22) is not zero, thus the electric field can have an effect on the adsorption capacity of interface. Under certain concentration, according to former conclusions, the interfacial tension decreases with the increase of electric field intensity, $d\gamma/dc_2 < 0$, then $\Gamma_2^{1E} < \Gamma_2^1$, indicating that the application of electric field has an effect on interfacial adsorption, i.e., the interfacial adsorption capacity of solution decreases.

The purifying air-conditioning engineering discussed in this study can be applied to sterile laboratories, food clean workshops and hospital operating rooms. Electric field application has an impact on interfacial adsorption, i.e., the volume of interface absorption of solution decreases. Such conclusion can be applied to the purifying air-conditioning engineering, indicating that the thermodynamic mechanism of interfacial adsorption has an important effect on purifying air-conditioning engineering under intensification of electric field.

6 Conclusion

Gibbs model is the most classic model using thermodynamic methods for simplified treatment of interfacial phenomenon and the position selection of Gibbs interface determines the complexity of adsorption processing.

Therefore, firstly, this study introduces the selection of Gibbs interface, Gibbs adsorption model and traditional Gibbs adsorption equation. Then using the fundamental thermodynamic relation and energy axiom, the Gibbs adsorption isothermal equation under the effect of electric field is deduced, i.e., from the aspect of thermodynamics, the relationships between interfacial adsorption capacity and interfacial tension, concentration and electric field intensity are deduced. The obtained conclusions lay a theoretical foundation for the interfacial adsorption under the effect of electric field as well as a good understanding of the relationship among these four properties.

This study takes the simplest two components as an example to analyze the relationships among interfacial adsorption capacity and interfacial tension, concentration and electric field intensity at the same temperature and under the same pressure, and qualitatively analyzes the effect of interfacial tension and electric field intensity on interfacial adsorption respectively. In addition, it comes to a conclusion that, after the application of electric field,

the interfacial adsorption capacity of solution decreases.

Received 30 March 2016

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