Research Article

Distribution of Vapor Pressure in the Vacuum Freeze-Drying Equipment

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In the big vacuum freeze-drying equipment, the drying rate of materials is uneven at different positions. This phenomenon can be explained by the uneven distribution of vapor pressure in chamber during the freeze-drying process. In this paper, a mathematical model is developed to describe the vapor flow in the passageways either between material plates and in the channel between plate groups. The distribution of vapor pressure along flow passageway is given. Two characteristic factors of passageways are defined to express the effects of structural and process parameters on vapor pressure distribution. The affecting factors and their actions are quantitatively discussed in detail. Two examples are calculated and analyzed. The analysis method and the conclusions are useful to estimate the difference of material drying rate at different parts in equipment and to direct the choice of structural and process parameters.

1. Introduction

Generally speaking, the food vacuum freeze-drying equipments of which the freeze-drying area is equivalent to or larger than $50 \, \text{m}^2$ are classified as big vacuum freeze-drying equipments [1]. In this type of vacuum freeze-drying equipment, there always exists a phenomenon that the drying rates of materials are uneven at different positions [2]. The reason is commonly charged upon the uneven temperature distribution of material plate [3, 4]. But, the uneven distribution of vapor pressure in vacuum chamber is usually ignored. In fact, the big vacuum freeze-drying equipment always adopts the form of multilayer of big area material plates (shelves) in order to improve the production efficiency and output. And in order to enhance the volume utilization rate of the vacuum chamber, the space between material plates is relatively narrow, and the evacuating port of the vacuum chamber is always on one side of it. All these structural characteristics accordingly lead to the result that

a flexural, long, and narrow flow transfer passageway is formed from the evaporating surfaces of the materials on the plates to the evacuating port of the vacuum chamber and can produce a flow resistance that cannot be ignored. And in the big vacuum freeze-drying equipment the water vapor flow in the sublimating drying stage is rather large, so in the long and narrow passageway there is a significant water vapor pressure difference, and this finally leads to the result that the drying rate of the materials is different on different plates or even in different positions of the same plate in the big vacuum freeze-drying equipment.

Analyzing the distribution of vapor pressure in the big vacuum freeze-drying equipment during the process of vacuum freeze-drying is significant to the correct evaluation of the inhomogeneity of the drying rate and its reduction. The former research on the mass transfer of vapor in the freeze-drying equipment mostly focused on the diffusion process and average drying rate in the materials to be dried [5–7]. But there is less study on the process of the vapor flowing from the surfaces of the materials to the evacuating port of the freeze-drying chamber and the difference of the drying rate in different parts of the materials caused by flowing resistance. This paper develops a mathematic model which describes the flow of the vapor outside the materials in the freeze-drying equipment. The distribution of vapor pressure and the relationship of parameters of each influencing factor are determined. And it also puts forward a quantitative analysis method of drying rate in different positions of the material and the theoretical designing basis of the key structure and processing parameter of the freeze-drying equipment.

2. Structure and Simplified Model

As shown in Figure 1, the material plates and pallets in the big freeze-drying equipment are usually arranged as the form of multilayer matrix, several or more than ten layers vertically aligned to form a group. There is a equidistant passageway between each two plates. One group, two groups or even more groups are arranged in parallel in the vacuum chamber. On both sides of each group and between two groups, there is an evacuating channel. The evacuating port of the vacuum chamber is often on the back of it opposite to the evacuating channels between groups (position a in Figure 1) or on the top or the bottom (position b or b′ in Figure 1). It also can be on the lateral side of the chamber (position d and d′, or position c in Figure 1). This form can have two evacuating ports.

In the freeze-drying process, the vapor sublimated from the material firstly flows to the evacuating channel between groups through the passageway along the plates, and then flows to the evacuating port along the evacuating channel. Obviously, in the same plate, the vapor sublimated from the material close to the evacuating channel can easily enter the evacuating channel between groups. But because the vapor sublimated from the material in the center of the plate is far from the evacuating channel, not only its transfer distance is long, but also it would merge the vapor sublimated from the material passing by and then flow to the evacuating channel together therefore, the flow resistance is bigger; and this results in that the vapor pressure component in the center of the passageway along the plate is obviously higher than that on the edge of the plate. Similarly, the vapor entering into the evacuating channel would also merge the one flowed out from other plates and then flow together to the evacuating port of the vacuum chamber. And this results in the uneven distribution of vapor pressure in the channel between plate groups. The final result is that the vapor pressure on the edge of the plate close to the evacuating port is the lowest where the material dries quickly, while the vapor pressure in the central of the plate far from the evacuating port is the highest
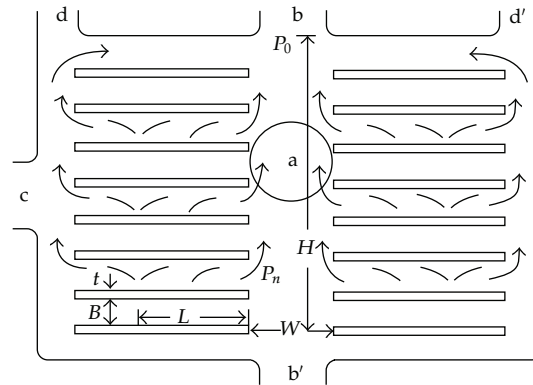
Figure 1: Structure sketch map of multilayer matrix in big-scale freezing equipment.

where the material dries slowly. This is just the significant reason for the uneven distribution of the drying rate of the material in the big vacuum freeze-drying equipment.

In this paper, a mathematic model is firstly developed for calculating the vapor flow and the pressure distribution in the unilateral passageway between plates in order to discover how the equipment's structure and process parameters affect the distribution of vapor pressure. And then the result is extended to the evacuating channel between plate groups and a method is consequently put forward to analyze the distribution of vapor pressure in the whole freeze-drying equipment.

The simplified model of the vapor flow in the unilateral passageway between plates is shown in Figure 2. The coordinate axis X is established along the midline of the width of the passageway, taking the midpoint of the plate as original point, that is, the effective length L of the passageway equals to half of the width of the plate. When the evacuating channels on both sides of the plate are largely different, the original point should be at the demarcation point where the vapor flows towards two sides, respectively. As to the plate which has the evacuating channel on one side only while its other side is closed, the original point should be on the closed termination and the effective length L of the passageway should be the full width of plate. The coordinate axis Y is established along the height direction of the passageway. The bottom of the upper plate is in the position where $Y = B/2$. The upper surface of material on this plate is in the position where $Y = -B/2$, and the vapor sublimates from this surface to the passageway between plates. The length direction of plate vertical to the XOY plane is regarded as to be infinite, so only the unit length of the plate is studied.

3. Hypothesis and Mathematical Equations

In order to simplify the calculation, the following hypotheses are made:

  (1) in the whole passageway, the vapor sublimates homogeneously from the surface of the material with constant rate. It is not influenced by the distribution of vapor pressure. The outgassing rate G_m of the material's surface can be calculated through the total dehydration quantity for some time in the freeze-drying process divided by the area of plate and time;
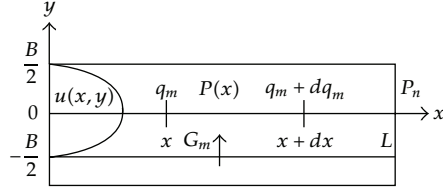
Figure 2: Simplified model of the vapor flow in the unilateral passageway between plates.

(2) both the influence of the permanent gas in the freezing chamber and the flow of vapor along the Y direction in the passageway are ignored. Attention is only focused on the one dimensional flow along the X direction and it is supposed that the flow is steady lamellar flow and the distribution of its flow velocity can be calculated according to the Poiseuille's flow between flat planes;

(3) the relationship between the vapor density and the pressure obeys to the perfect gas equation.

Some parameters and their units are defined as follows: B is the height of passageway between plates, m; L is the unilateral length of the passageway, m; u is the flow velocity of vapor, m/s; μ is the kinematical viscosity of vapor, Pa·s; $P(x)$ is the vapor pressure somewhere in the passageway between plates, Pa; q_m is the mass flow through the passageway section, kg/s; G_m is the outgassing rate of the material's surface, kg/(m²s); M is the molar mass of the vapor, 0.018 kg/mol; R is the universal gas constant, 8.3145 J/(mol. K); T is the Kelvin temperature of vapor, K; P_n is the vapor pressure at the outlet of the passageway, Pa.

The mathematical equation which describes the vapor flow in the passageway between plates is as follows: distribution of the vapor flow velocity is

$$u(x,y) = -\frac{1}{2\mu} \cdot \frac{dp}{dx} \cdot \left[\left(\frac{B}{2} \right)^2 - y^2 \right].$$ (3.1)

The mass flow through any passageway section X is

$$q_m(x) = \int_{-B/2}^{B/2} \rho \, u(x,y) \cdot dy = -\frac{MB^3}{12\mu RT} \cdot p \cdot \frac{dp}{dx}.$$ (3.2)

In the above equation, the perfect gas equation has been used as

$$\rho = \frac{pM}{RT}.$$ (3.3)

Considering the law of conservation of mass, the mass increment in the segment $x \sim x + dx$ is

$$dq_m = -\frac{MB^3}{12\mu RT} d\left(p\frac{dp}{dx} \right) = G_m \cdot dx$$ (3.4)

Accordingly, the control equation can be obtained which describes the distribution of vapor pressure

$$\frac{d^2}{dx^2}\left(p^2\right) = -\frac{24\mu RTG_m}{MB^3}. \tag{3.5}$$

Using the terminal condition

$$x = 0, \qquad \frac{dp}{dx} = 0, \qquad x = L, \qquad p = p_n, \tag{3.6}$$

there will be

$$p(x) = p_n\sqrt{1 + \alpha - \alpha\left(\frac{x}{L}\right)^2}, \tag{3.7}$$

where, the characteristic coefficient of the passageway α is defined as

$$\alpha = \frac{12\mu RTG_m \cdot L^2}{MB^3 p_n^2}. \tag{3.8}$$

And the mass flows either through any passage section x and at the outlet of the passageway where $x = L$ are respectively obtained as

$$q_m(x) = G_m \cdot x, \qquad q_m(L) = G_m \cdot L. \tag{3.9}$$

The analytical method and the calculated result above can be extended to the calculation of the pressure distribution in the evacuating channel between plate groups. Referring to the mark symbol in **Figure 1**, the width of the evacuating channel between plate groups W is substituted for the width of the passageway between plates B. The average distance H from the farthest plate to the evacuating port is substituted for the unilateral length L of the passageway between plates. The coordinate axis z is established to express the distance from a certain plate to the farthest plate. The total outgassing quantity of each plate at its outlet G_mL is uniformly spread in the width $B + t$ of space between plates. And it is considered that the outgassing appears on both sides of the evacuating channel between groups. Then, the outgassing rate G_z corresponding to the evacuating channel between groups is

$$G_z = \frac{2G_mL}{B + t}. \tag{3.10}$$

If P_0 is defined to express the vapor pressure at the terminal end of the evacuating channel between groups, that is, at the evacuating port of the vacuum chamber, then the formula

calculating the distribution of vapor pressure $p_n(z)$ in the evacuating channel between groups is

$$p_n(z) = p_0\sqrt{1 + \beta - \beta\left(\frac{z}{H}\right)^2},\tag{3.11}$$

where, the characteristic coefficient of the evacuating channel between groups β is defined as

$$\beta = \frac{12\mu RTG_z \cdot H^2}{MW^3 p_0^2}.\tag{3.12}$$

4. Discussion about the Calculating Results

(1) Using the above result, the vapor pressure at any appointed position in the vacuum freeze-drying equipment can be conveniently calculated. Firstly, P_0 must be estimated, which is the lowest vapor pressure at the evacuating port and is produced by the vacuum system and the ice condenser. Then, based on the related structural parameters of freeze-dryer such as H, W, B, t, and L, besides the processing parameters G_m and P_0, the vapor pressure P_n at the outlets of the passageway in any plates can be calculated by formulae (3.10), (3.11), and (3.12). At last, the vapor pressure $P(x)$ in any position of the passageway of the plate can be worked out by formulae (3.7) and (3.8). For example, in general case, the central part ($x = 0$) of the farthest plate ($z = 0$) is the place where vapor pressure is the highest in the vacuum chamber, and the value of maximum vapor pressure is

$$p_{\max} = p_0\sqrt{1 + \beta}\ \sqrt{1 + \alpha}.\tag{4.1}$$

(2) In the passageway of a certain plate, the maximal pressure difference between the central part and the edge part is

$$\Delta p = p(0) - p_n = p_n\left(\sqrt{1 + \alpha} - 1\right).\tag{4.2}$$

The deviation ratio of maximal pressure in the passageway of this plate is defined as

$$\delta = \frac{\Delta p}{p_n} = \sqrt{1 + \alpha} - 1.\tag{4.3}$$

The relation curve of δ-α is shown in **Figure 3**. From the figure, it is found that if you want the deviation ratio of pressure δ to be small, correspondingly the characteristic coefficient of the passageway between plates α should also be small. Similarly, the deviation ratio of maximal pressure in the evacuating channel between groups can be defined. Its relationship to the characteristic coefficient of the evacuating channel
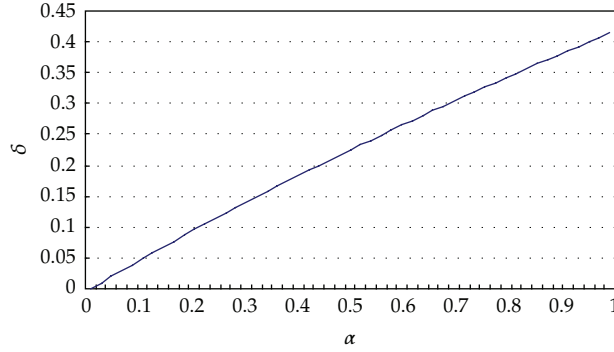
Figure 3: The relation of the deviation ratio of maximal pressure to the characteristic coefficient.

between groups is the same to formula (4.3). The deviation ratio of maximal pressure for the whole vacuum chamber should be

$$\delta_{\max} = \frac{p_{\max} - p_0}{p_0} = \sqrt{1 + \alpha} \sqrt{1 + \beta} - 1. \tag{4.4}$$

If you want $\delta_{\max}$ to be less than a certainvalue, you have to control the value of α and β simultaneously.

(3) The distribution of vapor pressure along the passageway of a certain plate is only related to the value of α. The pressure variation $p(x)/p_n \sim x/L$ is shown in **Figure 4**. From the definition formula (3.8) of α, it is known that the influencing factors include not only the geometrical structure parameters of passageway B and L, but also the processing parameters G_m and P_n. The difference of the power of each parameter indicates that they have different effects on the vapor pressure distribution. The relationship of the characteristic parameter α to the length L and height B of the passageway is expressed as $\alpha \propto L^2/B^3$. This does not obey to the proportional similarity criterion. For example, if both L and B are reduced by one time, the value of α will increase one time but not keep invariant. Therefore, in order to reduce the value of α, that will reduce the pressure difference between the central part and the edge part of the passageway, appropriately increasing the height B is more effective than reducing the length L. The relationship of the characteristic parameter α to the processing parameters G_m and P_n is $\alpha \propto G_m/p_n^2$. It indicates that when the outgassing quantity of the material surface G_m increases, the α will increase with direct proportion, and the pressure difference between the central part and the edge part of the passageway will also increase. On the other hand, when the outlet pressure P_n decreases, though the characteristic coefficient α and the relative deviation ratio δ is increasing sharply, the maximal pressure difference Δp is not increasing quickly and its value will not exceed $\Delta p_{\max} = p_n \sqrt{\alpha}$.

(4) Similarly, the distribution of vapor pressure along the evacuating channel between groups is only related to the value of β. The pressure variation $p_n/p_0 \sim z/H$ is also shown in Figure 4. Its influencing factors contain geometrical structure parameters and processing parameters. The restrictive relationship among them and their relation to β are the same as the results above.
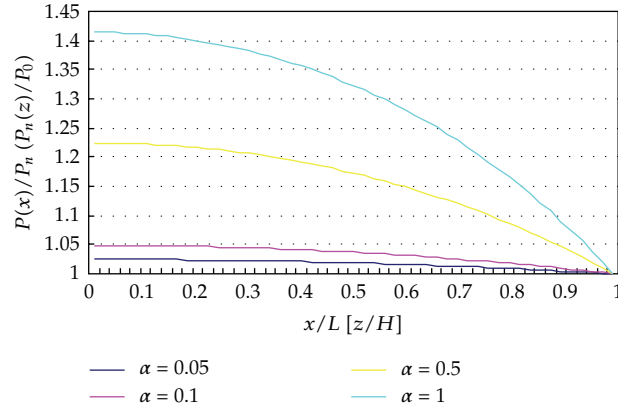
Figure 4: Vapor pressure distribution in the passageway channel between plates.

(5) Using the calculated result of vapor pressure distribution, the approximate quantitative analysis of the material drying rate in different positions of plates in big freeze-drying equipments can be made. The research shows that the relation of the sublimation rate G_m of the moisture in material to the vapor pressure P on the upper surface of material is approximately like this: $G_m \propto \sqrt{p_v - p}$. Where the P_v is the saturated vapor pressure on the sublimation interface and is approximated as a constant during the freeze-drying process. Then the relative difference of the drying rate in different position can be estimated by substituting values of vapor pressure P.

(6) The above result can be the theoretical reference for the design of structure and processing parameters of freeze-drying equipment. For instance, it is usually chosen that the characteristic coefficient α of the passageway in the farthest plate equals to the characteristic coefficient β of the evacuating channel between groups. If the value of deviation ratio of maximal pressure in the whole vacuum chamber is determined, then the relationship of the structural parameters should be

$$\frac{LW^3(B+t)}{2H^2B^3} = 1 + \delta_{\max}. \tag{4.5}$$

5. Calculation Example

Using the above calculation method, the checking computations of vapor pressure distribution in two types of food vacuum freeze-drying equipments are respectively made. The first example is a type of the food vacuum freeze-drying equipment LG-50 with the shelf area of $50\,\mathrm{m}^2$. The arrangement layout of its plate system is the same as the one shown in Figure 1. Two plate groups are lateral symmetry and each group has sixteen radiation heating plates. Between each two adjoining radiation heating plates, there hangs a material pallet. The total width between each pallet and the plate are all $0.6\,\mathrm{m}$. Between each two groups and on both sides of them, there is a wide evacuating channel which leads to the evacuating port towards the ice condenser. The evacuating port is on the bottom of the central channel between groups (b′ in the Figure 1), and its shape is a whole strip along the length direction

Table 1: The structural, processing, and calculated parameters of example 1.

Structural parameters	Processing parameters	Calculated parameters
$B = 0.025\,\text{m}$ $t = 0.065\,\text{m}$ $L = 0.3\,\text{m}$ $W = 0.38\,\text{m}$ $H = 1.6\,\text{m}$ $n = 1 \sim 15$	$p_0 = 50\,\text{Pa}$ $G_m = 1\,\text{kg}/(\text{m}^2\text{h}) = 2.78 \times 10^{-4}\,\text{kg}/(\text{m}^2\text{s})$ $T = 310\,\text{K} = 37°\text{C}$ $\mu = 8.8 \times 10^{-5}\,\text{Pa·s}$ $G_z = 1.85 \times 10^{-3}\,\text{kg}/(\text{m}^2\text{s})$	$\beta = 5.2 \times 10^{-3}$ $\alpha = 0.097$ $\delta_{\max} = 0.050$ $p_{\max} = 52.5\,\text{Pa}$ $\Delta p_{\max} = 2.5\,\text{Pa}$

of plates (vertical to the paper in the Figure 1). So the vapor flow of each plate can flow down directly from the central and bilateral evacuating channel to the evacuating port.

The structural parameters, determined processing parameters, and calculated results of the pressure distribution are shown in Table 1. It indicates that the value of the characteristic coefficient β is very small. This shows that the vapor can flow in the wide evacuating channel between groups favorably and will not produce measurable pressure difference on the two sides of the plates. The distribution of vapor pressure in the whole drying chamber is quite even. The deviation ratio of maximal pressure $\delta_{\max}$ is only 4.7% and the factual value of pressure difference is 2.5 Pa. The existence of pressure difference is for that the characteristic coefficient α is a little big that is, the pressure difference mainly exists in the central and on the edge of the plates. Furthermore, by adjusting the position of the pallet between two plates to increase the value of B, the value of α can be decreased, therefore, both the deviation ratio of pressure and pressure difference between plates may be decreased. For example, when $B = 0.03\,\text{m}$ and $t = 0.06\,\text{m}$, the maximal pressure difference is $\Delta p_{\max} = 1.5\,\text{Pa}$.

The second example is a big food vacuum freeze-drying equipment of which the area is approximately $200\,\text{m}^2$. The arrangement layout of its plate system is the same as the first example, as shown in Figure 1. There are evacuating channels between groups with the same width in the center and on the two sides of each two plates. The two evacuating ports are respectively on the top of bilateral channels (d and d′ in Figure 1). But, because the space between the top radiation heating plate and the top of vacuum drying chamber is too small, the vapor flowing from the two groups of plates cannot flow to the bilateral evacuating ports through the central evacuating channel. It is only discharged through the bilateral evacuating channels. And this equals to only the plates having evacuating channel on one side. At the same time, the evacuating port is set in the center of the freeze-drying chamber of which the length is 8 m (vertical to the paper in Figure 1). So the vapor from the plates on the two ends of chamber need to transfer about 4.5 m along the bilateral evacuating channels before it arrives at the evacuating port. The cross-section area of the evacuating port is much smaller than that of the evacuating channel, so the closer to the evacuating port, the bigger the converted outgassing rate of the channel between groups is. And this makes the average outgassing rate G_z to be about five times of the normal value. These factors all enlarge the uneven level of pressure distribution in the whole drying chamber.

During the calculation of the pressure distribution in the drying chamber, the attention is focused on the vapor flowing process from the plates on two ends of the chamber to the bilateral evacuating channels. The structural parameters, processing parameters and calculated results are shown in Table 2. It indicates that the deviation ratio of maximal pressure in the chamber is close to 18.8% and the maximal pressure difference is $\Delta p_{\max} = 9.4\,\text{Pa}$. It has exceeded the allowable deviation range of normal work. And it has been found that the drying extent of the freeze-dried food is uneven from the experiment. This indirectly

Table 2: The structural, processing, and calculated parameters of example 2.

Structural parameters	Processing parameters	Calculated parameters
$B = 0.03\,\text{m}$		$\beta = 0.196$
$t = 0.065\,\text{m}$	$p_0 = 50\,\text{Pa}$	$\alpha = 0.181$
$L = 0.6\,\text{m}$	$G_m = 1\,\text{kg}/(\text{m}^2\text{h}) = 2.78 \times 10^{-4}\,\text{kg}/(\text{m}^2\text{s})$	$\delta_{\max} = 0.188$
$W = 0.38\,\text{m}$	$T = 310\,\text{K} = 37°\text{C}$	$p_{\max} = 59.43\,\text{Pa}$
$H = 4.5\,\text{m}$	$\mu = 8.8 \times 10^{-5}\,\text{Pa·s}$	$\Delta p_{\max} = 9.43\,\text{Pa}$
$n = 1 \sim 19$	$G_z = 8.78 \times 10^{-3}\,\text{kg}/(\text{m}^2\text{s})$	

indicates the correctness of the calculated results. Afterwards, through increasing the number of evacuating port and properly arranging them, the pressure distribution in the drying chamber tends to be even and the drying extent becomes uniform.

6. Conclusion

(1) The calculation indicates that there exists an uneven vapor pressure distribution in big vacuum freeze-drying equipments due to flowing resistance.

(2) Though the position of the evacuating port and the structure of the evacuating channel are properly arranging, the uneven pressure distribution can be improved, and the internal pressure difference can be decreased.

(3) The checking computations indicate that when the values of characteristic coefficient α and β exceed 15% and the value of deviation ratio of maximal pressure exceeds 15%, it can be obviously found that the drying extent is uneven in the freeze-dried food.

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