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Optimization of Drying Process of Lithium Battery Pole Piece Based on Simulation Technology

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Abstract: - After electrode pulping and coating of lithium battery, it is necessary to dry the pole pieces, but there is a contradiction between drying efficiency and drying quality. In the process of rapid drying, the binder components are easy to migrate, which reduces the adhesion of the pole pieces, leading to the increase of internal resistance of the pole pieces and subsequent manufacturing defects. Therefore, this paper puts forward an optimization scheme of pole piece efficient drying process. Firstly, based on the analysis of the transfer process of heat and solvent quality in the drying process, the calculation method of drying curve is put forward, and the optimal design criteria of drying parameters are established. Furthermore, a multiphase flow and heat transfer model of drying process is constructed, and the oven structure and parameters are optimized based on multiphase flow simulation. The experimental results of drying process optimization of positive pole pieces show that the mass production speed of 51 Ah positive pole is increased by 25% after process optimization. The adhesion of the A surface of the pole piece is increased by 6.5%, and the difference between the A and B surfaces is decreased by 91%. The average resistance of the pole piece decreases by 12%, which meets the high requirements for the quality and efficiency of pole piece drying for automotive lithium-ion batteries.

Keywords: Lithium Battery, Positive Plate, Drying, Analog Simulation.

I. INTRODUCTION

After electrode pulping and coating, it is necessary to dry the pole piece, but there is a contradiction between drying efficiency and drying quality[1]. In the process of rapid drying, the binder components are easy to migrate, which reduces the adhesion of the pole piece, leading to the increase of internal resistance of the pole piece and subsequent manufacturing defects[2]. Therefore, this paper puts forward an optimization scheme of pole piece efficient drying process. Firstly, based on the analysis of the transfer process between heat and solvent quality in drying process, the calculation method of drying curve is put forward, and the optimal **design criteria of drying parameters are established. Furthermore, the multiphase flow and heat transfer model of drying process** is constructed, and the structure and parameters of oven are optimized based on multiphase flow simulation[3].

II. ANALYSIS AND OPTIMIZATION METHOD OF DRYING PROCESS

A. *Drying Process Analysis*

This paper mainly focuses on the optimization of drying process of positive pole pieces. The main problems in drying the positive plate are: low adhesion of the plate, whether the oily solvent NMP is fully dried, high resistance of the plate, low production efficiency and so on[4]. On the basis of ensuring drying, firstly, powder and glue are required to be evenly distributed in the pole piece. When the drying speed is too fast, glue and conductive agent will float up, resulting in low adhesion and high resistance of the pole piece, so in theory, the slower the drying speed, the better[5]. There are two transfer processes in the pole piece drying process:

1) *Heat transfer process:* The schematic diagram of heat transfer during pole piece drying is shown in Figure 1.

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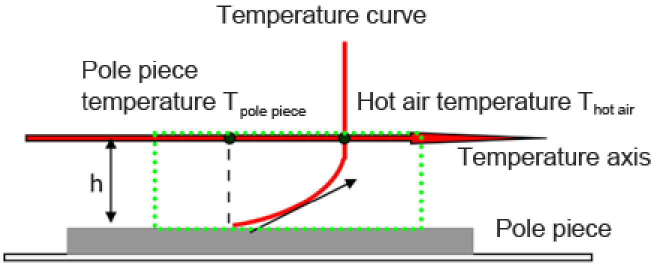

Figure 1: Schematic Diagram of Heat Transfer in the Drying Process of Pole Piece

Heat transfer rate between hot air and pole piece

$$a=k\cdot\frac{(T_{air}-T_{pole})}{h} \tag{1}$$

In order to obtain higher heat transfer efficiency, K and △t should be as large as possible and H should be as small as possible. However, k is a constant and cannot be increased, while the temperature of the pole piece is constant. Therefore, we can only raise the temperature of hot air and reduce the thickness of boundary layer as much as possible. The thickness of the boundary layer can be reduced by increasing the wind speed.

2) Quality transfer process: The mass transfer process during pole piece drying refers to the process of NMP volatilization from pole piece to hot air[6]. Comprises two stages, namely, NMP is transferred from the inside of the pole piece to the surface of the pole piece and from the surface of the pole piece to the hot air. The schematic diagram of the volatilization process of NMP from the surface of the pole piece to hot air is shown in Figure 2.

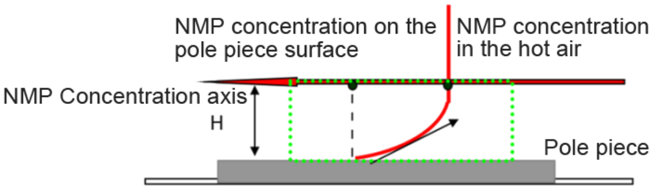

Figure 2: Schematic Diagram of the Evaporation Process of Nmp (Moisture) From the Surface of the Electrode to Hot Air

The concentration difference of NMP is

$$\Delta n = n_{es} - n_{ha} \tag{2}$$

n_{elec} represents the concentration of NMP on the surface of the polarizer. n_{ha} represents the concentration of NMP in the hot air. The transfer rate of NMP between the hot air and the polarizer is:

$$a = k\cdot\frac{n_{elec}-n_{ha}}{h} \tag{3}$$

With the volatilization of NMP on the surface of the pole piece, the NMP inside the pole piece will diffuse to the surface of the pole piece along the micro-holes formed by the volatilization of NMP on the surface, and then volatilize from the surface of the pole piece into the hot air. The diffusion speed of NMP inside the pole piece to the surface directly affects the adhesion of the pole piece. If the speed is too fast, the glue in the pole piece will be brought to the surface of the pole piece with NMP, which will cause the adhesion of the pole piece to deteriorate. At the same time, too fast drying will lead to uneven hole-making inside the pole piece. In theory, the slower the diffusion process is, the better, but it is impossible in actual production, so a reasonable drying parameter should be taken between the quality of the pole piece and the production efficiency.

B. Calculation Method of Drying Curve

By calculating the actual drying amount of the pole piece in each temperature zone, the drying ratio of the pole piece in each temperature zone can be calculated, and the drying curve can be made. The drying curve can be changed by adjusting the drying parameters, and the adhesion and resistance of the pole piece corresponding to different drying curves can be tested to find the most suitable drying parameters[7]. In the test, the known parameters are fresh air speed V, fresh air temperature T, fresh air duct cross-sectional area S, and exhaust NMP concentration n%.

$$v_t = v + s \tag{4}$$

$$v_{ss} = v_t \times \frac{273.15}{273.15 + t_{vss}} \quad (5)$$

vss represents the wind speed under standard conditions. Because the NMP concentration tested by the gas sensor is the percentage of the NMP concentration with the lower explosion limit of LEL% of NMP, and the lower explosion limit of NMP is LEL% of 0.99%, the actual NMP concentration in the exhaust air is zero, and this concentration is the volume concentration. The volume of NMP discharged per unit time under standard temperature and pressure.

$$\mathrm{v} = v_{ss} \times \mathrm{n}_{real} = v_t \times \frac{273.15}{273.15 + \mathrm{t}} \times \mathrm{n}\% \times LEL\% \quad (6)$$

nreal represents the actual concentration of NMP.

From the ideal gas state equation

$$PV = \mathrm{n}RT \quad (7)$$

The number of moles of NMP discharged per unit time

$$\mathrm{n} = \frac{PV}{RT} = P \times v_t \times \frac{273.15}{273.15 + t} \times n\% \times LEL\% \times \frac{1}{RT} \quad (8)$$

Where p is the standard atmospheric pressure and r is the constant of 8.314; The Kelvin temperature when t is 0 degrees Celsius is 273.15 K.. Therefore, the mass of NMP discharged per unit time is the product of "the number of moles of NMP" and "the molar mass of NMP".

$$\mathrm{n} = \frac{PV}{RT} = P \times v_t \times \frac{273.15}{273.15 + t} \times n\% \times LEL\% \times M_{NMP} \times \frac{1}{RT} \quad (9)$$

Note: MNMP is about 99 g/mol.

C. Optimum Design of Drying Parameters

According to the existing oven drying process, the eight temperature zones of the oven are divided into the following four zones[8], and the temperature zones and the direction of the pole pieces are shown in Figure 3.

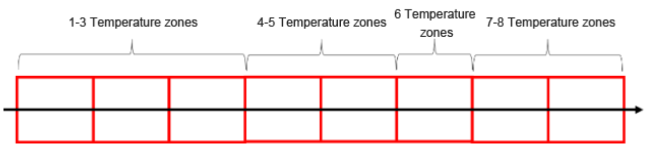

Figure 3: Pole Piece Transport Direction

1) 1-3 temperature zone: the pre-drying zone of pole pieces. When the pole pieces enter the oven, the temperature is low, so the drying temperature can be increased as much as possible.

2) 4-5 temperature zone: the accelerated drying zone of the pole piece. After entering this zone, the temperature of the pole piece is already very high, and NMP in the pole piece will volatilize rapidly in this zone. It is necessary to reduce the drying speed in this area. Because there is a lot of NMP evaporated in this area, the concentration of NMP in the oven is too high, which will lead to safety accidents. Therefore, it is necessary to control the drying temperature.

3) 6 temperature zone: the transition zone of pole piece drying; After the pole piece enters this area, most NMP on the pole piece has been volatilized, so the drying temperature can be raised appropriately.

4) 7-8 temperature zone: electrode plate deceleration drying zone: only a little NMP remains after the electrode plate enters this zone. In order to make the NMP residue in the electrode plate low, it is necessary to increase the drying temperature to make it volatilize as much as possible.

From the above drying process setting, it can be seen that slowing down the drying speed of the pole piece can make the glue and conductive agent inside the pole piece volatilize more evenly, thus improving the adhesion of the pole piece and reducing the resistance. The curve of A-side coating and drying will continue to move to the later temperature zone, so that the seventh and eighth temperature zones will also undertake certain drying tasks, and the A-side pole piece will be dried again when it passes through layer B, which can solve the problem of excessive NMP residue. According to this idea, the drying effect can be optimized by changing the drying parameters.

III. OPTIMIZATION OF OVEN STRUCTURE AND PARAMETERS BASED ON MULTIPHASE FLOW SIMULATION

In order to study the drying characteristics of pole pieces under limited experimental conditions and low cost, explore the influence of different factors on drying evaporation characteristics, and find out the drying process with guiding significance for production, computational fluid dynamics software is selected to simulate the drying characteristics of pole pieces. The physical model and numerical simulation of the oven and pole piece are built. On the basis of verifying the calculation model, the experimental results of the bench are simulated and compared.

A. *Multiphase Flow Heat Transfer Model*

NMP undergoes phase change in the oven, which belongs to phase change heat transfer, so the multiphase flow model is used to solve it[9]. The software STAR-CCM+ mainly provides users with three multiphase flow models, namely Euler separated flow model, VOF model and Euler mixed model[10]. Euler's mixed model is a simplified version of Euler's model, and its computational cost is low. This model is suitable for simulating suspended multiphase flow. In this model, the computational cost is reduced by assuming that the suspended phase is a uniform single-phase system. The limitation of this model is that one and only one of the mixed phases is compressible.

In the Eulerian multiphase model, the mass flux of each component I is conserved at the interface between gas and liquid film, so:

$$\rho Y_{\mathrm{i}}(\nu-\dot{h})-\rho D_i \frac{dY_i}{dy}=\rho_f Y_{f,i}(\nu_f-\dot{h})-\rho_f D_{f,i}\frac{dY_i}{dy}\bigg|_f \tag{10}$$

At the critical state, the calculation yields, ρ and ρ_f are the densities of gas and liquid film, Y_i and $Y_{f,i}$ are the mass fractions of gas and liquid film, v and v_f are the normal velocity components of gas and liquid film, D_i and $D_{f,i}$ are the molecular diffusion coefficients of gas and liquid film, and h is the change rate of liquid film thickness.

The expression of total evaporation rate is derived, which is valid under all conditions:

$$\dot{m}_\nu=\frac{\dot{Q}_\nu+\sum_{\mathrm{i}}^{N_\nu}\Delta H_i^{vap}\rho D_i\frac{dY_i}{dy}}{\sum_{\mathrm{i}}^{N_\nu}\Delta H_{\mathrm{i}}^{\mathrm{vap}}Y_{\mathrm{i}}} \tag{11}$$

B. *Finite Element Calculation Simulation Verification*

STAR-CCM+ was selected to simulate the flow and heat transfer in pole piece drying. For the convenience of calculation, the following assumptions are made: assume that the gas in the drying box is incompressible, assume that the gas in the drying box is Newtonian fluid, ignore the influence of small parts in the drying box on the thermal environment in the box, and the wall of the drying box is insulated. The speed inlet and pressure outlet are selected at the inlet and outlet of the drying box respectively. In the physical model of liquid film, NMP and cathode material are regarded as slurry. The mass fraction of liquid phase in liquid film is monitored, and when the monitored value reaches stability, iteration is regarded as convergence.

According to the control experiment, the calculation conditions are selected: the initial temperature of the drying oven is 80℃, the temperature of the pole piece is 25℃, the evaporation temperature of the drying oven for 1-8 sections is 60℃, 60℃, 70℃, 110℃, 110℃, 90℃, 80℃ and 80℃ respectively, the drying air flow rate is 1 m/s, and the relative speed of the pole piece is 20 m/min. The evaporation solvent is NMP, and the material library of NMP is constructed in STAR-CCM+. The oven is modeled in STAR-CCM+ software, and the structure of the oven is shown in Figure 3(a).

Before the simulation begins, the initial temperature of the pole piece and the oven are set at 25℃ in the model, the rotating speed of the air inlet fan is 0.5 m/s, and the temperatures of eight ovens are 60℃, 60℃, 80℃, 100℃, 100℃, 90℃, 80℃ and 80℃ respectively, and the drying time of each oven is 5 seconds. During the pole piece drying process of the oven, In the simulation process, due to the influence of the structure of the upper and lower chambers of the oven and the arrangement position of the nozzles, the temperature of the pole pieces at different positions varies with time with the increase of time, and the simulation results are shown in Figure 4. The pole piece in the middle position of the oven has a higher temperature than other pole pieces. When the simulation time is 10 s, after two drying processes at 60℃, the temperature difference between the pole piece in the middle position and the pole piece in the edge position of the oven is about 10℃. When the simulation time is 20 s and 30 s, the average temperature reaches the highest after passing through four and six ovens respectively, and the temperature difference between the pole piece in the middle position and the pole piece in the edge position of the oven can reach about 30℃ and 50℃ respectively. When the simulation time is 40 s, the oven temperature drops and the temperature difference drops to about 25℃. It can be seen that the temperature of pole pieces in different positions in the oven is obviously different after being heated by hot air.

However, when the oven structure leads to different temperatures on the surface of the pole piece at different positions, the change of NMP mass fraction in the pole piece is shown in Figure 4. From the simulation results, it can be seen that after the 40 s drying process is completed, the NMP content in the pole piece at the center and the pole piece at the edge of the oven is 0.08 and 0.01 respectively, and the difference is very obvious. This may be one of the reasons for the performance difference of battery pole pieces.

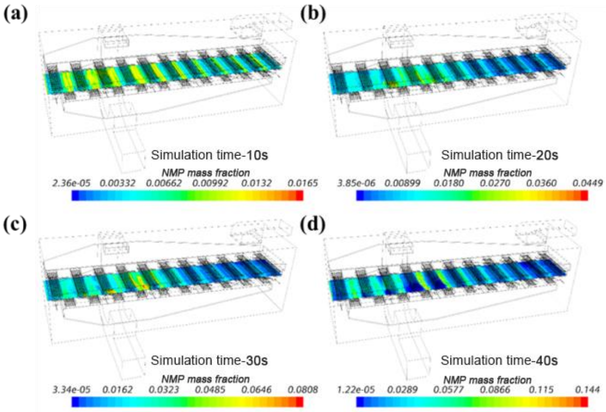

Figure 4: Pole Piece Transport Direction(Wind Speed a~d)

In addition to the position of the pole piece in the oven, the fan speed at the air inlet of the oven will lead to the change of air flow in the oven, which will have different effects on the temperature of the pole piece at different positions[11]. The influence of different fan speeds on the air distribution in the oven is shown in Figure 5. It can be concluded that the higher the inlet fan speed, the better the average drying effect, but the worse the drying uniformity, the higher the fan speed is, the better.

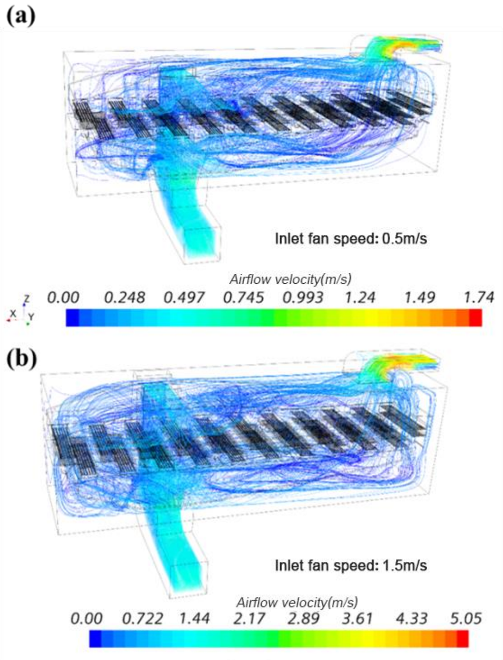

Figure 5: Pole Piece Transport Direction(Mode a+ Mode b)

In addition to the pole piece position and fan speed, the initial temperature of the oven is also one of the factors that affect the pole piece drying effect[12]. The simulation results show that the lower the initial temperature of the oven and the slower the fan speed, the slower the evaporation mass fraction of NMP decreases and the lower the average surface evaporation rate. On the contrary, the higher the initial temperature of the oven and the faster the fan speed, the faster the evaporation mass fraction of NMP decreases and the higher the average evaporation rate on the surface. When the evaporation mass fraction of NMP decreases to a certain extent, the surface evaporation rate begins to decrease obviously, and at this time, the obvious drying effect cannot be obtained by continuing to increase the oven temperature or fan speed[13].

IV. Optimization scheme and verification of pole piece drying process.

According to the solvent evaporation results of drying experiment, the evaporation ratio of each temperature zone is shown in Figure 6. Scheme 1 moves the drying curve to the later temperature zone on the basis of mass production scheme, and Scheme 2 moves the drying curve to the later temperature zone on the basis of Scheme 1, and a small amount of NMP still evaporates in the 8 temperature zone of the pole piece, which makes the drying speed of the pole piece slower.

After comparing the drying effect of each temperature zone, the performance of the dried pole piece is shown in Table 1. Among them, the adhesive force of the A and B surfaces of the pole piece is tested by universal tensile machine, and the internal resistance of the pole piece is tested.

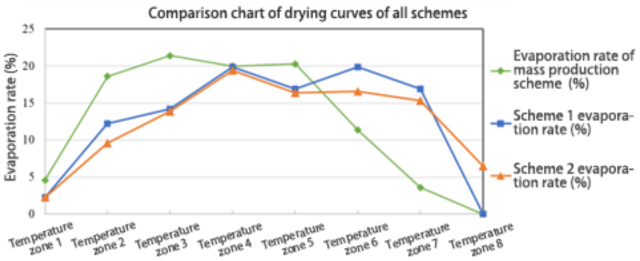

Figure 6: Comparison Chart of Drying Curves of All Schemes

Table 1: Comparison of Pole Piece Performance of Two Schemes

Cpmparison	A(gf)	B(gf)	IR(Ω)
Production	38.6	45.9	21.3
Scheme 1	42.7	46.5	22.6
Scheme 2	46.6	45.6	21.7

It can be seen from the test results that as the drying curve of the pole piece moves to the back temperature zone, the drying of the pole piece in each temperature zone is more balanced, the adhesion level of the pole piece is improved, and there is no obvious difference in the resistance of the pole piece.

From the results, the production efficiency increased by 25% after the speed increase, the resistance of the pole piece decreased obviously, and the adhesion force did not change obviously. Therefore, the optimization of drying parameters and the improvement of coating speed can obviously improve the performance of battery pole pieces and greatly improve the production efficiency.

V. Conclusions

In this paper, in view of the high requirements for the drying quality and efficiency of automotive lithium-ion batteries, firstly, the transfer process of heat and solvent quality in the drying process is analyzed, the calculation method of drying curve is put forward, and the optimal design criteria of drying parameters are established. Secondly, the multiphase flow and heat transfer model of drying process is established, and the structure and parameters of the oven are optimized based on multiphase flow simulation. Finally, the drying process optimization experiment of anode pole piece was carried out. The results showed that the mass production speed of 51 Ah anode was increased by 25% after the process optimization. The adhesion of the A surface of the pole piece is increased by 6.5%, and the difference between the A and B surfaces is decreased by 91%. The average resistance of the pole piece decreased by 12%.

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