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A mathematical model of spray drying granulation process in formulation

Kagami, H.
School of Medicine, Fujita Health University, Toyoake, Japan

*E-mail of the corresponding author: kagami@fujita-hu.ac.jp

Abstract
In this study, we apply the drying model of polymer solution coated on a flat substrate to the spray drying granulation process in formulation. In order to apply the former model to the drying of this spherical object, we consider this spherical object to be a stack of solution films applied on the spherical shell and discuss the drying process of each solution film. As a result, we see that the smaller the radius of curvature of the droplet, the more the fine particles tend to be unevenly distributed on the surface of the droplets during drying.

Keywords: *model, numerical simulation, spray drying granulation, spherical, droplets*

1. Introduction

Drying process of polymer solution coated on a substrate is very important in various industrial applications such as fabricating flat polymer thin films [1] and inkjet printing [2, 3]. Then we have proposed and modified a model of drying process of polymer solution coated on a flat substrate for flat polymer film fabrication [4 – 14]. Then we have proposed the method of thickness control of a thin film after drying [15 – 17]. And we have clarified dependence of distribution of polymer molecules on a flat substrate after drying on various parameters based on analysis of many numerical simulations of the model.

On the other hand, the demand that a flat polymer film should be formed not only on a flat substrate but also on a three-dimensional uneven substrate after drying a polymer liquid film coated on the substrate has been increasing with the advancement in microfabrication technology. Therefore, we expanded the above-mentioned model into the drying model of a polymer liquid film coated on three-dimensional structure. Then we analyzed dependence of distribution of polymer molecules on the three-dimensional structure on various parameters through many numerical simulations. The results were reported at IDS 2016 [18].

Then, in this study, we apply the drying model to the spray drying granulation process in formulation. Spray drying granulation is a type of wet granulation method in formulations. In the spray drying granulation, the slurry raw material is made into minute droplets and dried. Therefore, the object to be dried is spherical. In order to apply the former model to the drying of this spherical object, we consider this spherical object to be a stack of solution films applied on the spherical shell and discuss the drying process of each solution film. The difference in area between the upper surface and the lower surface in the minute volume of each solution film can be represented by the radius from the center of the spherical shell and is related to the difference in diffusion in the vertical direction.

As a result of numerical simulation of this modified model, it was found that the smaller the radius of curvature r of the droplet, the more the fine particles tend to be unevenly distributed on the surface of the droplets during drying.

2. Model

2.1. Theory and basic equations

As mentioned in previous papers, there are two dynamic models of the drying process of polymer solution coated on a flat substrate, namely, an evaporation model and a transport model for a non-equilibrium polymer solution (Kagami et al., 2002; Kagami, 2011; Kagami and Kubota, 2011). The latter can be divided into two basic types of transport, that is, the following two diffusion paths (Kagami et al., 2002):

(1) Diffusion of solvent containing solutes (polymers) in the direction of evaporation of the solvent

(2) Change in concentration in a solution

Because details of theory and the basic equations have been reported in previous papers (Kagami et al., 2002; Kagami, 2011; Kagami and Kubota, 2011; Kagami, 2014a; Kagami, 2015), only the basic equations are shown here.

First, we consider the following evaporation model (Kagami et al., 2002).

$$G = \gamma(1 - \beta C) \tag{1}$$

Here, G is the evaporation rate, C is the concentration of the solution, and β is a constant. Furthermore,

$$\gamma = K\sqrt{\frac{M}{2\pi RT}}P_0 \tag{2}$$

is a correction factor, where P_0 is the vapor pressure, M is the molecular weight, R is the gas constant, T is temperature, and K is a correction factor for the theoretical evaporation rate (Hickman and Trevoy, 1952).

Then, the two diffusion models are formulated as follows. First, the diffusion equation for solvent containing solutes is written as

$$\frac{\partial V}{\partial t} = K_v \nabla^2 V \tag{3}$$

where V is the volume of solvent containing solutes included in a space and K_v is the diffusion coefficient of the solvent (Kagami, 2011). An evaporation term (Eq. (1)) is added to Eq. (3), which describes the interface between liquid and gas (vacuum), so Eq. (3) is modified to (Kagami, 2011)

$$\frac{\partial V}{\partial t} = K_v \nabla^2 V - \gamma(1 - \beta C) \tag{4}$$

Next, the diffusion equation governing the change in concentration in solution is written as (Kagami, 2014a; Kagami and Kubota, 2011)

$$\frac{\partial N}{\partial t} = K_C \nabla^2 N + \frac{N}{V}(K_V - K_C)\nabla^2 V - \frac{2K_C}{V}\nabla N \cdot \nabla V \tag{5}$$

where N denotes the number of solute molecules included in a space and K_C is the diffusion coefficient of the solution.

In this study, we consider a solution containing one type of solute and one type of solvent for simplicity. Therefore, we build our model mainly using Eqs. (1)–(5).

The diffusion coefficient, K_C, of the solution changes with time. As mentioned in previous studies,   is written as (Kagami, 2011)

$$K_C = \frac{k_B T}{6\pi R \eta_0 \{[\eta]C + 1\}} \qquad (6)$$

where k_B is the Boltzmann constant, η_0 is the viscosity of the solvent, and $[\eta]$ is the intrinsic viscosity.

2.2. Improvement of the avobe model in the case of spherical dry solution

Now consider the drying of spherical solution as in the case of sray drying granulation. In order to estimate the diffusion in each direction in the minute region in the spherical solution, consider the minute region S in the spherical solution between the distance r and $r + dr$ from the center O of the sphere.

For the sake of simplicity, let us consider a circle C of a cross section of this sphere cut by a plane passing through the center O of the sphere. Considering the cross section $\mathrm{S_C}$ of the micro region S cut off at the deviation angle θ in the circle C, it is as shown in Fig. 1, and the length in the circumferential direction and the length in the radial direction of the cross section $\mathrm{S_C}$ can be expressed as shown in Fig. 1.

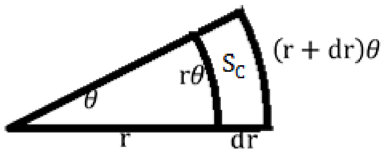

Fig. 1 The cross section $\mathrm{S_C}$ of the micro region S cut off at the deviation angle θ in the circle C

Now, if $r\theta = dq$, the length of the circular arc of radius r and the length of the circular arc of radius $r + dr$ in cross section $\mathrm{S_C}$ can be represented as dq and $dq\left(1 + \frac{dr}{r}\right)$, respectively. Assuming now that $dr = dq$, only the length of the arc of the radius $r + dr$ among the lengths in the circumferential direction and the radial direction of the cross section $\mathrm{S_C}$ is other $\left(1 + \frac{dr}{r}\right)$ times.

By replacing the deviation angle θ with the solid angle Θ and doing the similar consideration, only the area of the spherical surface with the radius $r + dr$ among the areas of each surface formed by cutting the minute region S at the solid angle Θ is other

$\left(1+\frac{dr}{r}\right)^2$ times. Namely, in diffusion in the spherical object, it is estimated that the ratio of the diffusion coefficients of the upper surface and the lower surface is $\left(1+\frac{dr}{r}\right)^2$.

3. Results and Discussion

3.1. Condition of numerical simulation

In order to discuss the drying of the solution film of thickness dr coated on the surface of spherical body with curvature radius r, r is regarded as a parameter and $dr = 0.1[mm]$. Solvent vaporizes only on interface between liquid and gas (vacuum) and boiling is out of imagination. Volume of solvent V and the number of solute N in solution are homogeneous except for surface coming in contact with gas (vacuum). Concerning surface, 10% unevenness about N is given only to the upper surface of the solution film by homogeneous random number.

Simulation is ended when total volume of solvent become less than a fixed value, for it is thought that drying is completed, that is, solutes can move in solution no more then.

Initial values of parameters are set as follows; $K_C = 1.8750\times10^{-8}$[m^2/s], $K_v = 1.2500\times10^{-8}$[m^2/s], $\gamma = 5.7000\times10^{-14}$[m^2/s].

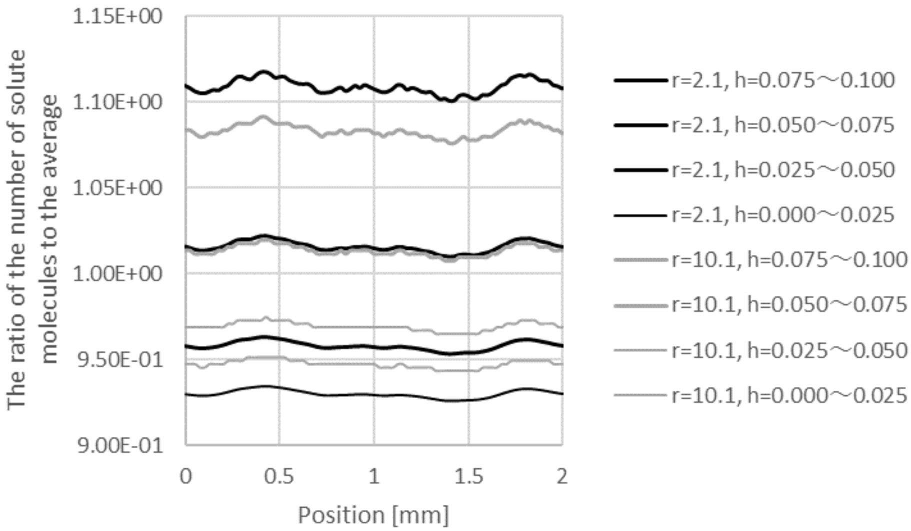

Fig. 2 The r dependence of the amount of solute present in each layer of height from the bottom of the solution film after drying

3.2. Results of numerical simulation

Figure 2 shows the r dependence of the amount of solute present in each layer of height h from the bottom of the solution film after drying. From this result, it can be seen that as the radius of curvature r is smaller, the uneven distribution of the solute (or the fine particles) on the surface of the solution (or the slurry) film after drying becomes conspicuous.

4. Conclusions

In this study, we apply the drying model to the spray drying granulation process in formulation. In order to apply the former model to the drying of this spherical object, we consider this spherical object to be a stack of solution films applied on the spherical shell and discuss the drying process of each solution film.
As a result, we see that the smaller the radius of curvature of the droplet, the more the fine particles tend to be unevenly distributed on the surface of the droplets during drying.

5. References

[1] Bornside, D. E., Macosko, C. W. and Scriven, L. E. Spin coating: One - dimensional model. J. Appl. Phys., 1989, 66, 5185-5193.

[2] de Gans, B. J. and Schubert, U. S. Inkjet Printing of Well-Defined Polymer Dots and Arrays. Langmuir, 2004, 20, 7789-7793.

[3] Soltman, D. and Subramanian, V. Inkjet-Printed Line Morphologies and Temperature Control of the Coffee Ring Effect. Langmuir, 2008, 24, 2224-2231.

[4] Kagami, H., Miyagawa, R., Kawata, A., Nakashima, D., Kobayashi, S., Kitano, T., Takeshita, K., Kubota, H. and Ohmi, T. A model of coating and drying process for the flat polymer film fabrication. Proc. SPIE., 2002, 4754, 252-259.

[5] Kagami, H. A modified model of drying process of a polymer liquid film taking effects of latent heat and heat conductivity into account. Proc. SPIE., 2003, 5130, 220-227.

[6] Kagami, H. The influence of spatio-temporal variation of temperature distribution in a polymer solution on a flat substrate on formation of polymer film's thickness distribution during drying process - based on results of simulation of the modified model. Proc. SPIE., 2004, 5446, 135-142.

[7] Kagami, H. An additional modified model of drying process of a polymer liquid film – influence of an abrupt drop of vapor pressure peculiar to polymer solution –. The 9th Asia Pacific Physics Conference Proceedings, 2004, 10-10C.

[8] Kagami, H. Nonlinear dynamics that appears in the dynamical model of drying process of a polymer solution coated on a flat substrate. Proc. SPIE., 2007, 6417, 641704-1 - 8.

[9] Kagami, H. A modified dynamical model of drying process of polymer blend solution coated on a flat substrate. Proc. SPIE., 2008, 7028, 702825-1 - 9.

[10] Kagami, H. Characteristic three-dimensional structure of resist's distribution after drying a resist solution coated on a flat substrate : analysis using the extended dynamical model of the drying process. Proc. SPIE., 2009, 7273, 727335-1 - 8.

[11] Kagami, H. A modified dynamical model of drying process of a polymer solution having plural solvents coated on a flat substrate for a flat and homogeneous polymer film fabrication. Proc. SPIE., 2010, 7716, 771625-1 - 9.

[12] Kagami, H. and Kubota, H. Applying the dynamical model of drying process of a polymer solution coated on a flat substrate to effects of bumpy substrate. Proc. SPIE., 2010, 7764, 77640T-1 - 8.

[13] Kagami, H., Kubota, H. A dynamical model of drying process of a polymer solution having plural solvents and plural solutes (polymers) coated on a flat substrate for a flat and homogeneous polymer film fabrication. Physica Status Solidi, C 8, 2011, No. 2, 586-588.

[14] Kagami, H. Impact of the Marangoni effect on the thin film thickness profile after drying polymer solution coated on a flat substrate. JPS Conf. Proc., 2014, 1, 015087-1 - 5.

[15] Kagami, H. Thickness control of a thin film after drying through thermal and evaporative management in drying process of a polymer solution coated on a flat substrate: application of the dynamical model of the drying process. Proc. SPIE., 2011, 8068, 806813-1 - 6.

[16] Kagami, H. More minute thickness control of a thin film after drying through temperature, evaporation and concentration management in drying process of a polymer solution coated on a flat substrate. Proceedings of the 19th International Drying Symposium, Lyon, August 24-27, 2014, ISBN 978-2-7598-1631-6.

[17] Kagami, H. Control of the Marangoni Effect in the Drying Process of a Polymer Solution Coated on a Flat Substrate through Temperature, Evaporation and Solute Concentration Management. Athens Journal of Technology & Engineering, 2015, Volume 2, Issue 1, 45-53.

[18] Kagami, H. A model of drying process of a polymer liquid film coated on three-dimensional structure. In Proceedings of 20th International Drying Symposium, Gifu, Japan, August 8-10, 2016; A-1-4.