Simulation of Microencapsulation Avocado Seeds Oil by Spray Drying

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Abstract. Drying air inlet temperature is one of the critical variables in the microencapsulation process by spray drying. However, when spray drying is carried out at inappropriate drying air inlet temperature, it can impact the particle produced. This study presents a simulation of spray drying from a mathematical model was developed to determine the effect of drying air inlet temperature on moisture content, particle diameter, particle density, and drying air outlet temperature in the microencapsulation process of avocado seeds oil as core materials and gum arabic as wall materials. For this aim, the mathematical model was developed then simulated using a matrix laboratory (Matlab) computer program with Euler numerical method for drying air inlet temperatures of 160, 180, and 200 °C. The selected model was validated with Cotabarren's experimental results indicating the model was acceptable. The particles' moisture contents predicted from simulation results are 1.170, 1.049, and 0.933 kg water/kg solid for 160, 180, and 200 °C, respectively. On the other hand, the predicted particle diameters are 29.73, 29.49, and 29.23 μm for 160, 180, and 200 °C, respectively. The predicted particle densities are 1215.72, 1225.21, and 1233.25 kg/m³ for 160, 180, and 200 °C, respectively. The prediction of drying air outlet temperatures was 39.76, 41.94, and 43.89 °C for inlet air temperatures of 160, 180, and 200 °C, respectively. The proposed models' simulation results show that the higher temperatures caused lower particle moisture content, smaller particle diameter, and higher particle density. Also, the outlet drying air temperatures were always the same as the outlet particle temperatures.

Keywords: Avocado seeds oil, drying air inlet temperature, Matlab, microencapsulation, spray drying

1. Introduction

Microencapsulation is the widely used technique to protect core material from damage caused by hazardous environmental conditions by entrapping core in shell material and forming small particles with a diameter of 1–1000 μm. Spray drying is one of the technologies that has been developed successfully to produce small particles powder, which has advantages of relatively simple, rapid, and reproducible, allowing easy scale-up compared to other encapsulation techniques, justifying its preferred use in the industrial sector, flexible process, and lower production costs [1]. However, encapsulation using spray drying is a very sensitive thermal process, and drying inlet temperature is one of the main parameters leading to variations in particle size, process yield, particle morphology, crystalline state, moisture content, and density, among others. Therefore, it would be disadvantageous to find these particles characterization by trial and error, which needs time, high costs, large waste generation, and operations distant from the optimal points, and considerable difficulties in ensuring the product's quality [2].
developed mathematical models are applied as solutions to these problems to predict the characteristics of the particles produced, and it is advantageous to determine suitable operating conditions to obtain the desired particle characteristics.

Generally, spray drying has four operation stages. They are atomization of liquid feed into a spray chamber, contact between the spray and the drying medium, moisture evaporation, and separation of dried products from airstream [3]. On the other side, emulsion droplets undergo two drying stages. Firstly, the droplet with excess liquid is subjected to a stream of drying gas and gains sensible heat, and vaporization begins. The evaporation results in droplet shrinkage and a simultaneous increase of solids concentration near the droplet surface. In turn, it leads to the precipitation of solid components at the droplet surface, and, eventually, all the surface becomes covered with a solid layer called the crust. At this moment, the droplet turns into a wet particle, and the second stage of drying commences. In the second stage, the drying process is hindered by additional resistance to the crust layer's evaporation. The second drying stage continues until the particle moisture content reduces to equilibrium with the drying medium. After this point, the drying process stops, and the particle is heated up to the equilibrium temperature [4]. These drying kinetics can be used as underlying the mathematical models to determine the effect of drying air inlet temperature.

A previous study has reported a steady-state mathematical model for a co-current spray dryer is developed to give a more phenomenological insight in the production of inhalable particles, ciprofloxacin hydrochloride (CIP), and then implemented in the gPROMS Environment [2]. However, the droplets were studied has no emulsion content. Furthermore, it is still an important challenge to understanding the drying kinetics of single droplets containing emulsion. Therefore, this study's objective was to combine mathematical models that apply to single droplets containing emulsion, which in this study avocado seeds oil as core material and gum arabic as wall material.

2. Methodology

This research aims to simulate a mathematical model for avocado seed extract's microencapsulation process with spray drying using Matlab.

2.1 Mathematical Model Assumptions

The mathematical model is carried out with a 1-D simulation approach. In the 1-D simulation approach, mass, heat, and momentum balance is carried out at the individual droplet level following the drying time. The drying kinetics incorporated in this approach allows the prediction of the drying rate profile.

1. The assumptions used in this mathematical model as follows [2]:
2. The drying medium is preheated air, and the solvent of the sprayed solution is water.
3. The drying air and the atomized liquid are co-current.
4. Steady-state for both air and droplets/particles phases is applied.
5. Droplets are spherical.
6. Plug flow for both air and droplet/particles phases (variations in the radial direction of mean droplet/particle size, droplet and air temperature, content water droplets, and air, and droplet and air velocities are negligible).
7. The evaporation force is the difference between air humidity, considering the droplet surface's saturation and bulk air's relative humidity.
8. The water content and temperature profiles in the droplets are neglected.
9. Agglomeration and breakage of droplets are neglected.
10. Heat transfer between droplets and air is considered.
11. The spray dryer undergoes heat exchange with ambient air through the wall chamber.
12. Chamber is considered to be a cylinder.
13. The diameter of the avocado seed extract in the droplet emulsion is 6 μm.
14. Consider only the distance function, i.e., axial direction or axial function distance, and do not consider the time function.
2.2 Mathematical Model Equations

This model is used to determine the optimum conditions/parameters for the microencapsulation of avocado seeds by spray drying, where the avocado seed extract in a droplet emulsion is assumed to be 6 μm [5]. The illustration of the drying of the microencapsulation is shown in Figure 1.

The variables used in this study were the inlet drying temperature of the water with variations of 160, 180, 200 °C.

The humidity of the sample is calculated using the following equation:

$$\frac{dW_p}{dz} = -\frac{\pi d_p^2 m_v}{v_p m_s} W_p(z_0) = W_{p0}$$  \hspace{1cm} (1)

The evaporation rate is obtained from the equation:

$$m_v = \beta (Y_{sat} - Y_b)$$ \hspace{1cm} (2)

$$Y_{sat} = \frac{P v M_w}{(P - P_v) M_a}$$ \hspace{1cm} (3)

β calculated as a function of the heat transfer coefficient (α):

$$\beta = \frac{\alpha d_p D_{eff}}{k_a}$$  \hspace{1cm} (4)

One droplet or isolated particle assumption is used to calculate the heat transfer coefficient [6-8]:

$$Nu = \frac{\alpha d_p}{k_a} = 2 + 0.6 Re^{0.5} Pr^{0.33}$$ \hspace{1cm} (5)

$$Re = \frac{\rho_a d_p (v_p - v_a)}{\mu_a}$$ \hspace{1cm} (6)

$$Pr = \frac{c p_a \mu_a}{k_a}$$ \hspace{1cm} (7)

Figure 1. The mechanism of the microencapsulation process in spray drying.
\[ v_a = \frac{\dot{M}_a}{\rho_a A_c} \]  

(8)

All air properties were calculated as an air temperature function (Ta) and calculated \([7]\) and \(D_{eff}\), which is calculated at 298 K.

For the droplet temperature calculated by the equation:

\[ \frac{dT_p}{dz} = \frac{\pi d_p^2}{v_p m_s (c_p + W_p c_{pw})} \left[ a(T_a - T_p) - m_s \Delta H_{ev} \right], \quad T_p(0) = T_{p0} \]  

(9)

when \(W_p \geq W_{pc}\)

\[ \frac{dd_p}{dz} = \frac{d_{p0} \pi d_p^2}{3 m_s v_p} \left( \frac{\rho_{p0} - \rho_w}{\rho_p - \rho_w} \right)^{-2/3} \rho_{p0} - \rho_w \left( 1 - \frac{\rho_p}{\rho_w} \right)^{2/3} \rho_s \left( 1 + \frac{\rho_p}{\rho_w} W_p \right)^2 d_p(0) = d_{p0} \]  

(10)

\[ v_a = \frac{d \rho_p}{dz} = - \frac{m_s \pi d_p^2}{m_s v_p} \rho_s \frac{1 - \rho_p}{\rho_w} \left( \frac{\rho_p}{\rho_w} \right)^2, \quad \rho_p(0) = \rho_{p0} \]  

(11)

when \(W_{peq} < W_p < W_{pc}\)

\[ \frac{d d_p}{dz} = 0 \]  

(12)

\[ \frac{d \rho_p}{dz} = - \frac{6 m_s}{d_p v_p} \]  

(13)

when \(W_p < W_{peq}\)

\[ \frac{d \rho_p}{dz} = 0 \]  

(14)

The change in droplet velocity \((v_p)\) is calculated through its momentum balance. The total force generated is balanced with the droplet density to produce

\[ \frac{dv_p}{dz} = g \left( \frac{\rho_p - \rho_a}{\rho_p v_p} \right) - \frac{3}{4 d_p \rho_p v_p} \left( v_a - v_p \right)^2, \quad v_p(0) = v_{p0} \]  

(15)

where \(g\) is the acceleration due to gravity and \(C_D\) is the drag coefficient, which is calculated as follows

\[ C_D = \frac{24}{Re} \]  

(16)

For moisture content, the equation as follows:

\[ \frac{dY_b}{dz} = N_t \frac{m_s \pi d_p^2}{M_a v_p}, \quad Y_b(0) = Y_{b0} \]  

(17)

The number of droplets \((Nt)\) that enter the chamber, which is calculated by

\[ N_t = \frac{\dot{M}_t}{V_{po} \rho_{p0}} \]  

(18)
The energy balance for the drying medium is given as follows

$$\frac{dT_a}{dz} = -\frac{N_t \pi d_p^2 (n_v c_p v + \alpha) (T_a - T_p)}{v_p M_a (c_p a + X_b c_p v)} + \frac{U (T_a - T_p) \pi D_c}{M_a (c_p a + X_b c_p v)}$$  \(T_a(0) = T_{a0}\)  \((19)\)

U represents the global heat transfer coefficient, contributing to conduction loss through the chamber wall. The initial air temperature \(T_{a0}\) is calculated based on the intake temperature of the drying air and atomization, \(D_c\) is the chamber diameter, and \(c_p v\) is the steam heat capacity calculated by:

$$\overline{c_p v} = \frac{1}{T_p - T_a} \int_{T_a}^{T_p} c_p v dT$$  \((20)\)

This research was conducted in a simulation using matrix laboratory software (Matlab) using Euler’s numerical method to perform microencapsulation modeling by spray drying. The flow of the simulation is shown in Figure 2.

**Figure 2.** Simulation flowchart

### 2.3 Material Properties and Operation Condition

In this study, the materials used for validation were CIP solution with the properties shown in Table 1. The same model was also applied to study the microencapsulation in spray drying using the avocado seed extract as the core material, gum arabic as the wall material, and water as the solvent. In this study, the new droplets that entered the chamber had a composition, namely gum arabic, avocado seed extract, and water. So that the density can be calculated as follows [8]:

$$\rho_{p0} = x_{GA} \rho_{GA} + x_{AE} \rho_{AE} + x_w \rho_w$$  \((21)\)
As for the solid-state of the particles, the composition is gum arabic and avocado seed extract. The avocado seed extract is considered a solid because it does not evaporate as it does in water. So that the solid density and heat capacity can be calculated as follows:

$$\rho_s = x_{GA}\rho_{GA} + x_{AE}\rho_{AE}$$  \hspace{1cm} (22)$$

$$c_{ps} = x_{GA}c_{pGA} + x_{AE}c_{pAE}$$  \hspace{1cm} (23)$$

The initial condition, the operating condition, and parameter data used in the simulation are depicted in Table 2.

**Table 1.** Material properties for validation and research

| No | Parameter                              | Ciprofloxacin hydrochloride (CIP) | Avocado Extract | Gum Arabic |
|----|----------------------------------------|----------------------------------|-----------------|------------|
| 1  | Density (kg/m³)                        | 1000 (CIP 10)                    | 1100 (CIP solid)| 1000       |
| 2  | Heat capacity (J/kg.K)                 | 1568 (CIP solid)                | 1670*           | 1500**     |

* Avocado seed extract heat capacity uses the heat capacity vegetable oil because avocado seed extract could not be found.
** The heat capacity gum arabic value uses the maltodextrin heat capacity value because the value for gum arabic cannot be found. Maltodextrin is chosen because it is commonly used as wall material and is often combined with gum arabic to become a better wall material.

**Table 2.** The initial condition, the operation condition, and determined parameter for simulation

| No | Parameter                              | Validation | Research |
|----|----------------------------------------|------------|----------|
| 1  | Initial temperature drying air (K)     | 383        | 433, 453, 473 |
| 2  | Initial temperature droplet (K)        | 298        | 298      |
| 3  | Initial droplet moisture content (kg air / kg solid) | 0.95 | 2.33 |
| 4  | Initial droplet diameter (μm)          | 32         | 32       |
| 5  | Initial droplet density (kg/m³)        | 1000       | 1081     |
| 6  | Initial humidity drying air (kg air / kg udara) | 0.0015 | 0.0015 |
| 7  | Initial velocity droplet/particle (m/s) | 1.897x10⁻⁴ | 1.897x10⁻⁴ |

**Operation Condition**

1. Total solid concentration in emulsion 10 (mg/mL) 30 (% mass)
2. Core material concentration in solid - 10 (% mass)
3. Flowrate feed liquid (m³/s) $3 \times 10^{-6}$ 9.72 x 10⁻⁸
4. Flowrate atomization air (m³/s) $667 \times 10^{-12}$ -
5. Flowrate drying air (m³/s) $3600$ 0.0097 0.0203
6. Delta z $10^6$ $10^6$

**Determined Parameter**

1. Thermal glass conductivity (W/mK) 0.8 0.8
2. Air molecular weight (g/mol) 28.97 28.97
3. Water molecular weight (g/mol) 18.02 18.02
4. Atmospheric pressure (Pa) 101325 101325
5. Gas pressure (Pa) 101325 101325
6. Water critical temperature (K) 647.3 647.3
7. Environment temperature (K) 298 298
8. Thickness of the surrounding air barrier layer(m) $8.97 \times 10^{-4}$ $8.97 \times 10^{-4}$
9. Chamber diameter (m) 0.2304 0.5
10. Diameter nozzle feed liquid (m) $0.77 \times 10^{-3}$ $0.77 \times 10^{-3}$
11. Chamber wall thickness (m) 0.0025 0.0025
12. Gravitation acceleration (m/s$^2$) 9.8 9.8
13. Gas constant (J/molK) 8.314 8.314
14. Global heat transfer coefficient (W/m$^2$K) 1.5144 1.5144

3. Results and Discussion

This study aims to create a simulation of a mathematical model for spray drying microencapsulation and determine the effect of drying air inlet temperature on particle moisture content, particle diameter, particle density, and drying air outlet temperature. The variable used in this simulation is the drying air inlet temperature ($T_a$), which is 160, 180, 200°C.

3.1 Mathematical Model Validation

In this study, validation was carried out by comparing the simulation results with the simulation and experimental results from the Cotabarren experiment and simulation [2]. The validation aims to examine whether the selected mathematical model is suitable for existing experimental data. The system's boundary condition was determined to be the distance traveled by the particles in the chamber starting from the nozzle's tip to the point where the particles leave the chamber (assumed 0 to 0.5 meters). The validation data simulation results with experiment and simulation by Cotabarren [2] consisted of particle moisture content profiles, particle diameter, particle density, and the drying air outlet temperature to the distance traveled by the particles in the chamber were shown in Figure 3.

Figure 3(a) shows that the final value of particle moisture content only reached 0.156, while in the Cotabarren experiment and simulation, both reached 0.03, so the error is 422%. Likewise, in Figure 3(c), the particle density changed very little compared to the reference results, with a final density of 1010.22 kg/m$^3$ and 1275 kg/m$^3$, so the error is 21.07% and 17.87%. For the final particle diameter, in Figure 3(b), the final result is 3.53 μm, exceeding the reference simulation data of 11 μm and experimental data of 7 μm, so the error of 67.89% and 49.54% is obtained. Figure 3(d) shows that the drying air temperature is close to the reference results at 330.55 K. For the simulation and experimental data have a discrepancy of 2.33% and 3.94% at the temperature of 323 K and 318 K, respectively.
Figure 3. The result of mathematical model validation for (a) particle moisture content, (b) particle diameter, (c) particle density, and (d) drying air temperature.

The trendline simulation results are the same as Cotabarren's simulation results. However, if it is seen from the error percentage, moisture content gives a high deviation, diameter and density results are good enough because the error is not too high, and for drying air temperature, the results are good because the error is deficient. The simulation results also do not show critical points and constant lines on moisture content, diameter, and density. These deviations are probably caused by the difference in the simulation software used. The data from the reference used are the simulation result data that have the most significant error.

3.2 Effect of drying air inlet temperature on particle characteristics

Figure 4(a) shows the effect of drying air inlet temperature on the particle's moisture content. It can be seen that increasing the temperature of drying air can increase the release of moisture from the droplets, resulting in particles with lower moisture content. It happened due to an increase in the driving force from drying at higher temperatures. The first drying stage is shorter at 200°C compared to 160 and 180°C. The higher temperature of drying air increases the evaporation rate, thereby accelerating crusts' formation and causing the critical point to be reached faster. However, from the simulation results, the lowest final moisture content only reaches 0.933 at 200°C. It indicates that the moisture content of the final particles is still a lot, even almost half. It might happen because the equilibrium moisture content formula for CIP was used instead of gum arabic.
Figure 4(b) shows the effect of the drying air's inlet temperature on the particle's diameter. It can be seen that increasing the temperature of the drying air results in lower diameter particles. The droplet volume, which initially contains much water, will evaporate, leaving particles with much less water content, so the final particles' volume also shrinks, which is related to the shrinking particle diameter. Furthermore, since the highest temperature resulted in the lowest moisture content, automatically the diameter is also the smallest. However, from the simulation results, it can also be seen that the shrinkage of the particle diameter is not too significant, where the smallest diameter that can be achieved is only up to 29.232 μm at a temperature of 200 °C or only 3 μm less than the initial one. It might happen because the critical moisture content achieved is still high so that the shrinkage of the particle size is not significant.

Figure 4(c) the effect of drying air inlet temperature on the density of the particle. It can that increasing the drying air temperature results in a higher particle density. As the diameter shrinks, the particles' volume also shrinks, or there is volume lost due to evaporation, so their density increases. Compared with theory, this increase in density should only occur at the beginning of evaporation until the moisture content reaches its critical point. After that, although the droplet diameter is constant, the porosity increases, so the density decreases. Nevertheless, from the results, there is no drop in density. It might happen due to the final moisture content, which is still large, causing the particles obtained are still wet, so the porosity does not increase.

Figure 4(d) shows the temperature trend for drying air as well as particles. From the simulation results, it can be seen that the drying air has decreased in temperature, while the particles have increased in temperature. It is caused by the transfer of heat from the drying air to the particles used to evaporate the water. So the output of the drying air and the particles fall at the same temperature.

From the simulation results above, if the graph is zoomed in, it will show that the trend for moisture content and diameter is following the single droplet drying kinetics as shown before, where the shape of the graph is divided into four parts. In the first part, changes are taking place very slowly. The heat from the drying air still heats the particles' surface before starting to evaporate the moisture. There is a drastic change caused by moisture evaporation from the particles' surface in the second part. The first and second parts are called the first drying stage, which occurs when the droplets enter the chamber until they reach their moisture content's critical point. In the third part, critical moisture content has been reached, and a transition has begun to occur towards its equilibrium point, marked by a sloping slope. This section is called the second drying stage. Then in the fourth part, the slope is linear, where the particles do not change anymore.
4. Conclusion

A simulation of a mathematical model for microencapsulation with spray drying has been made. The simulation has been validated resulted in the particle moisture content with a very high deviation of 422%. The particle diameter and density of simulation results have an acceptable deviation of 49.54% and 17.87%, respectively, from Cotabarren's experimental results. Whereas the air temperature with a very low deviation of 3.95%. By varying the drying air inlet's temperature, higher temperatures produce lower particle moisture content, smaller particle diameter, and higher particle density. Moreover, drying air and particles has the same temperature at the outlet.

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References

[1] Bakri A M, Abbas S, Ali B, Majeed H, Abouelwafa M Y, Mousa A, and Liang L 2016 Comprehensive Reviews in Food Science and Food Safety 15 143
[2] Cotabarren I M, Bertin D, Razuc M, Ramirez-Rigo M V, and Pina J 2018 Chemical Engineering Research and Design 132 1091
[3] Kuriakose R and Anandharakrishnan C 2010 Trends Food Sci Tech 21 383
[4] Mezhericher M, Levy A, and Borde I 2014 Drying Technology: An International Journal 4
[5] Boger B R, Georgetti S R, and Kurozawa L E 2018 Food Sci. Technol. 3
[6] Ranz W E and Marshall W R 1952 Chem. Eng. Prog. 48 141
[7] Negiz A, Lagergen R, and Cinar A 1995 Ind. Eng. Chem. Res. 34 3289
[8] Pinto M A, Kemp I, Beringham S, Hartwig T, and Bisten A 2014 Chem. Eng. Res. Des. 92 619
[9] Welty J R, Wicks C E, Wilson R E, and Rorrer G L 2008 Wiley & Sons 572
[10] Shamaei S, Seidilou S, Aghbashlo M, and Valizade H 2017 Food and Bioproducts Processing 101 103