Numerical investigation of droplet evaporation phenomena in the spray methods

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Abstract. Physical phenomena of the droplet evaporation occurred in the spray pyrolysis reactor tube has been studied. This model simulation was expected to predict the process occurred in the reactor tube and predicted the size and morphology of particle. The dynamical model of particles influenced by macroscopic parameters such as reactor wall temperature, flow rate carrier gas and concentration of precursor have been successfully simulated. The size of the particle, the number of aqueous vapors around droplet, air temperature around the droplet, temperature of droplet and solution concentration profile of particle in the reactor tube were simulated. The desired final particle can be achieved by controlling these factors.

1. Introduction
The spray method is one of the nanoparticle synthesis methods that are now being developed by researchers. The spray is the generation of small droplets originating from the liquid phase medium. The method commonly used in the synthesis of nanostructured particle solids is spray drying and spray pyrolysis. The method is initiated from a solution of metal nitrate or other atomized solution, and then after going through a heating system, the solid particles will be formed. Spray methods have many advantages, including: the resulting particles are in the form of spherical, the diameter distribution is uniform and can be controlled from micrometers to sub-micrometers; the purity of the particles produced is very high, the process takes place continuously [1–5].

Simulation is a bridge between theory and experiment that has been accepted as one of the methods of material research and development. An experiment material can be preceded by computer simulation to determine the conditions needed or predict the results. Computer simulations can function to complement experiments. The advantage of computer simulations is easy to perform. The results obtained from computer simulations can be a benchmark for the experimental methods carried out.

A computational approach was used to investigate the spray drying process. In spray drying experiments, there are several parameters such as carrier gas flow (Q), reactor wall temperature (Tₐ), initial concentration of droplet solution (C₀), the number of concentrations of droplets entering the
reactor \((N_o)\) and initial droplet \((d_{p_o})\) diameter size that affects the reactor length needed for droplets to be dry particles, the size of the final droplet, the amount of water vapor concentration on the droplet surface, the amount of water vapor concentration around the droplet, the temperature on the droplet surface and the air around the droplet. Therefore computer simulations can be varied so that the parameters can predict the results obtained and can be used as a reference for experiments spray drying.

In the current study, the models presented by Jayanthi [6] and Kodas [7] were used. This research aims to simulate physical phenomena (particle dynamics) of droplets in a tube reactor using the spray drying methods and to get a deep understanding of mechanisms involved in the particle formation to obtain desired final particles. Relationships between the operating conditions and final properties of particles are investigated.

2. Experimental procedure

Study on droplet evaporation and particle formation in the reactor are depicted in figure 1. The model describes changes in droplet diameter and continuous particle morphology based on multiple evaporations of droplets along the tubular aerosol reactor and together with the distribution of solution concentrations in the droplet [1]. The droplet (monodispersed droplets) assumed has diameter \(d_p\), droplet concentration \(N_o\) and solution concentration \(C_o\). The droplet is transported into the reactor using the average carrier gas flow \(Q\), relative humidity \(RH^o\) and initial temperature \(T_w\). The reactor has length \(L\) and diameter \(2R\) and temperature \(T_w\). As the solvent evaporates, the size of the droplet decreases and the concentration of the solution inside increases while moving inside the reactor [1]. The concentration of the solution in the droplet is in the critical area during the nucleation of the solution, precipitation, decomposition and solid processes that occur in the particles.

![Figure 1. Illustration of simulation model.](image-url)
Evaporation of solvents causes the concentration of a solution to increase on the droplet surface and form a concentration gradient which is the result of diffusion of the solution. At critical supersaturation (CSS), if the concentration of the solution at the center of the droplet \( (\frac{c}{R} = 0) \) is higher than the equilibrium concentration (ES) \([1,8,9]\), the solid phase will nucleate and produce a solid particle. Meanwhile, if the concentration of the solution in the center of the droplet is smaller than the equilibrium concentration (ES), hollow particles will be formed \([1]\).

The modeling is equated with limiting the problem. Changes in diameter \( d_{p} \) (residence time) along the reactor can be stated as follows:

\[
\frac{dd_{p}}{dt} = \frac{4D_{v}m_{l}}{\rho_{p}d_{p}N_{A}n(n - n_{s})} (1)
\]

with \( D_{v} \) (cm\(^2\)/s) is the vapor diffusion coefficient in air at \( T \), \( m_{l} \) (g/mol) is the mass of the water molecule, \( n \) is the concentration of vapor (molecule / cm\(^3\)) in the air at \( T \), \( n_{s} \) is the concentration of vapor (molecule / cm\(^3\)) at the droplet surface at \( T_{s} \) and \( \rho_{p} \) are droplet density, which is a function of particle size.

Changes in droplet surface temperature \( (T_{s}) \) over the residence time along the reactor is given by latent heat of evaporation of water and heat transfer from the air surrounding it \([1]\).

\[
\frac{dT_{s}}{dt} = \frac{1}{C_{p}d_{p}} \left[ 3H_{L} \frac{dd_{p}}{dt} + \frac{6h_{s}}{\rho_{p}}(T - T_{s}) \right] (2)
\]

With \( C_{p} \) is droplet heat capacity, \( H_{L} \) is latent heat from evaporation of water and \( h_{s} \) is heat transfer coefficient at the droplet. Changes in the concentration of vapor with respect to residence time are given by:

\[
\frac{dn}{dt} = \left[ -2\pi d_{p}D_{v}N_{o}(n(n - n_{s})) - \frac{2K_{m}(n(n - n_{w}))}{R} \right] (3)
\]

\( N_{o} \) (no / cm\(^3\)) is the number of droplet concentrations, \( n_{w} \) a is the concentration of vapor at the reactor wall, \( K_{m} \) is vapor mass transfer coefficient for laminar flow, \( R \) is the reactor tube radius. The first term of equation (3) states the concentration of water vapor entering the air due to evaporation of droplets. The second term states the vapor lost due to the diffusion of water vapor into the reactor wall. Air temperature along the residence time due to the transfer of heat from the air to the droplet and from the reactor wall \([10]\):

\[
\frac{dT}{dt} = \frac{1}{FC_{pa}} \left[ -\pi^{2}R^{2}d_{p}^{2}N_{o}h_{s}(T - T_{s}) + 2\pi Rh_{w}(T_{w} - T) \right] \left( \frac{dz}{dt} \right) (4)
\]

with \( F \) is the molar flow rate of air, \( C_{pa} \) is the heat capacity of surrounding gas and \( h_{w} \) is the heat transfer coefficient at the reactor wall. By using the ideal gas law, aerosol residence times along the reactor can be stated as follows:

\[
\frac{dt}{dz} = \frac{0.06\pi R^{2}/T_{o}}{0.06\pi R^{2}/T}(1 - yw) \left( 1 - yw_{o} \right) (5)
\]

\( t \) is residence time (s), \( z \) (cm) is the coordinate distance of the reactor, \( Q \) is the carrier gas flow rate (cm\(^3\)/s), \( T \) is the air temperature (K), \( y_{w} \) is the mole fraction of vapor in surrounding gas, \( y_{w_{o}} \) is the mole
fraction of vapor in surrounding gas at \( z = 0 \). The correction factor \( \frac{(1 - yw)}{(1 - yw_o)} \) is calculated for changes vapor contained in the air flow as a result of evaporation of droplets. Assuming \( n_w = 0 \), the fraction of moles of water vapor in the air can be expressed as:

\[
yw = \frac{n}{n + n_s}
\]  

(6)

whereas the mole fraction of water vapor in air at \( z = 0 \) can be expressed as:

\[
yw_o = \frac{n_o}{n_o + n_s}
\]  

(7)

Equations (1) to (5) are coupled differential equations. The solutions to these equations are done simultaneously using the 4th order Runge-Kutta method. Order 4 of the Runge-Kutta method is one method for finding solutions to the first-order differential equation. The constants used in the model are obtained from Reid et al. [11] and some are calculated. The boundary condition of the mass and energy transfer on the reactor wall are determined by the reactor system. In the case of diffusion drying, the reactor tube is wrapped in a drying agent, so the wall behaves like a sink that is perfect for steam \( (n_{w}) \) and constant wall temperatures.

The initial conditions at the reactor inlet \( (z = 0) \) are:

\[
t = 0, \quad d_p = d_{po}, \quad n = n_o, \quad T_s = T = T_o.
\]

The initial vapor concentration with relative humidity in surrounding gas \( (RH^o) \) is:

\[
n_o = \frac{RH^o p_s^o}{k T_o}
\]  

(8)

with \( p_s^o \) is the equilibrium vapor pressure to the temperature \( T_o \) and \( k \) is the Boltzmann constant. The vapor concentration at the droplet surface is given by:

\[
n_s = \frac{p_s}{k T_s}
\]  

(9)

\( p_s \) is the droplet surface vapor pressure at temperature \( T_s \). The presence of dissolved substances in water, resulting in the lowest balance of vapor pressure through pure solution rather than through pure solvent \( p_s^o(T_s) \) is:

\[
p_s^o(T_s) = a p_s^o(T_s)
\]  

(10)

with \( a \) is water activity in droplet solution which is influenced by the molality of the solution \( m \) and the osmotic coefficient of water \( \phi \), with the equation:

\[
a = \exp(-0.036 m \phi)
\]  

(11)

with \( \phi \) is a function that alternates from \( m \) obtained from Pitzer and Mayorga [12]. By assuming the ideal mixed solution of dissolved and water, the droplet density is given by:

\[
\rho_p = \rho_L + \left( \frac{d_{ps}}{d_p} \right)^3 (\rho_s - \rho_L)
\]  

(12)
with $\rho_w$ is the density of pure water, $\rho_a$ is the dissolved density and $d_{ps}$ is the equivalent diameter of a complete drying droplet. The average molecular droplet is given:

$$b = \frac{1000\rho_s}{M_s\rho_l[(d_p/d_{ps}) - 1]}$$ (13)

with $M_s$ is the soluble molecular weight (NaCl).

In relation to the higher transport resistance for diffusion of the solution in the droplet compared to the evaporation of solvents on the droplet surface, the concentration profile spreads in the droplet with the highest solution concentration on the surface. Assuming $d_p(dd_p/dt)$ is constant, then the concentration of solute in the radial direction $r$ of the droplet center is [7]:

$$C = C_o \exp \left[ \frac{K}{4D_s} \left( \frac{r}{r_p} \right)^2 \right]$$ (14)

with $C_o$ is the solute concentration at the center of the droplet, $r_p = d_p/2$ is the radius of the droplet, $D_s$ is the coefficient of diffusion of the solution while $K$ is the mass balance on the surface of the droplet:

$$K = \frac{2D_s m_l(n_s - n)}{\rho_o N_A}$$ (15)

3. Results and discussion

The results obtained refer to some variations in the value of flow rate ($Q$), variations in the value of reactor wall temperature ($T_w$) and variations in the value of the initial concentration ($C_o$) of NaCl solution. The three variations carried out in the program simulation, and in the experimental process, then the size of the droplet simulation results and experimental results are compared.

3.1. Variation in $Q$ (flow rate)

In this simulation, the flow rate was varied in three values: 1 l/min, 3 l/min, and 5 l/min. The effect of the average gas flow on the evolution of the size of droplets along the reactor can be seen in figure 2.

Figure 2. Effect of flow rate on the evolution of the size of NaCl droplets throughout the reactor.

Figure 3. The effect of the average gas flow on air temperature.
Based on figure 2, the droplet diameter size evolved from $2.14 \times 10^{-4}$ cm (the initial size of the droplet calculated on the ultrasonic nebulizer frequency at 1.7 MHz) up to $4.60 \times 10^{-5}$ cm. The result shows that the size of the final droplet diameter is not influenced by variations in $Q$ (flow rate) because the final droplet size remains the same ($4.60 \times 10^{-5}$ cm). However, the achievement of the last droplet size on the length of the reactor is different. The reactor length needed to reach dry particles (perfect evaporation process) is 11.3563 cm for $Q = 1$ l/min, while for $Q = 3$ l/min is 34.069 cm and for $Q = 5$ l/min is 56.7816 cm. The difference in reactor length needed to reach the final size of the droplet is due to the difference in the carrier gas flow rate carrying the droplet. The greater the $Q$ value, the faster the carrier gas flow so that with the same evaporation time, the reactor length needed to reach the final size is different. The evaporation time needed by droplets for one evaporation process is 0.002 seconds. Therefore with the same evaporation time and different flow rates, the required reactor length is different. Therefore, the greater the $Q$ value, the longer the reactor length for the droplet to completely evaporate the solvent and become a dry particle. The heat required by the droplet originating from the furnace that flows radiatively to the reactor wall, the heat reactor wall then flows through the air. The profile of the air temperature around the droplet can be seen in figure 3.

In figure 3, it can be seen that the value of air temperature rises from 299 K (initial air temperature, obtained from the measurement results in the experiment) up to 500 K. The difference in temperature transition rate towards stable values occurs for variation in $Q$. For $Q = 1$ l/min the air temperature reaches a stable value around the reactor length of 2.5 cm so that the air temperature has the same value with the reactor wall temperature. For $Q = 3$ l/min the reactor length needed to reach a stable temperature is around 5 cm while at $Q = 5$ l/min the length of the reactor required to achieve a stable value is about 10 cm. The difference in the transition temperature rate is due to differences in the velocity of the carrier gas. The smaller the $Q$ value, the slower the speed of the carrier gas, the more time it takes for heat to flow from the reactor wall to the air, which will result in faster temperature transition speeds. In the other side, if the value of $Q$ increased, the rate of the carrier gas will become faster so that the time needed for the heat to flow from the reactor wall into the air will reduce the speed of the transition temperature. The heat contained in the air would be flowed by gas particles to the droplet surface so that the heat is used by droplets to vaporize the solvents contained in the droplets. Heat can flow because of the temperature difference between gas and droplets. Temperature profiles on droplets can be seen in figure 4.

![Figure 4](image_url)  
**Figure 4.** Effect of average gas flow on droplet temperature.

![Figure 5](image_url)  
**Figure 5.** The effect of the average gas flow on the concentration of water vapor in the air around the droplet.
The result shows that the droplet temperature values increase from 299 K (initial air temperature, obtained from the measurement results) to 500 K. At the final condition (at the time of evaporation) the droplet temperature reaches 499.9212 K. This value is smaller than the air temperature (499.9595 K). The droplet temperature is quite enough to evaporate the solvent in the droplet because in this case water is used as the solvent and it requires temperatures of around 100°C or 373 K to evaporate completely. The droplet temperature transition rate is the same as the air temperature profile. Because the solvent in the droplet evaporates, the concentration of water vapor in the carrier gas increases. The profile of water vapor concentration in the gas around the droplet is shown in figure 5.

In figure 5, the increase in the concentration of water vapor rate is different for each value of $Q$. The smaller the value of $Q$, the faster the concentration of water vapor increases. This is because the flow velocity of the carrier gas was slow enough so that the time needed for heat to flow to the droplet is getting longer, as a result, the concentration of water vapor rate is getting faster. However, the amount of water vapor in the carrier gas during the evaporation time is the same. Meanwhile, if the value of $Q$ is getting bigger, then the concentration of water vapor rate will increase. During the evaporation time the droplet concentration of water vapor in the carrier gas increases. The solvent in the droplet that evaporates into water vapor not only diffuses into the surrounding gas but also distributes to the droplet surface. The water vapor concentration profile on the droplet surface can be seen in figure 6.

![Figure 6](image1.png)  ![Figure 7](image2.png)

**Figure 6.** Effects of the average gas flow on the concentration of water vapor on the surface of the droplet.

**Figure 7.** Effect of average gas flow on solution concentration on droplets.

Figure 6 shows that the concentration of water vapor on the droplet surface decreases with respect to the length of the reactor. This is because the heat on the droplet surface is used by water vapor particles to increase its kinetic energy and moved from the droplet surface to the air with the carrier gas. During the spray drying process, the concentration of the solution is changed gradually. The concentration profile of the solution on droplets could be seen in figure 7. It can be seen that the concentration of the solution on the droplet is higher when it gets closer to the surface. This is because when the solvent in the droplet underwent an evaporation process, the solvent carries dissolved particles on the droplet to the surface through diffusion which causes the concentration of the solution on the droplet surface to become higher than the solution concentration at the center of the droplet. However, the difference in the $Q$ does not affect the magnitude of the concentration distribution in the droplet as the difference in $Q$ only affects the gas flow rate and the droplet heating time. However, because the temperature used is constant, the difference in $Q$ does not change the concentration distribution in the droplet, because the concentration distribution in the droplet is influenced by the temperature on the droplet and the initial
3.2. Variation in Tw (wall temperature)

Value of reactor wall temperature is also an essential element in physical phenomena that occur in the reactor. The value of temperature was varied at 400 K, 500 K, and 600 K. The profile of the droplet size of the reactor temperature variation can be seen in figure 8. The result showed that the size of the droplet diameter evolved from $2.14 \times 10^{-5}$ cm (the initial size of the droplet obtained from the measurement of frequency (at 1.7 MHz) from the ultrasonic nebulizer) to a certain size which varies in each reactor wall temperature variation. In the temperature range of 400-600 K, the linearly increasing temperature profile will result in a longer reactor length needed to reach the final size of the droplet. This is because of the higher the reactor wall temperature, the higher the heat given to the air around the reactor. Therefore, the kinetic energy of gas particles increases so that the gas velocity of the droplet particle carrier is getting fast and makes the required reactor length become longer. This will cause less heating time so that the droplet particles dry more entirely and cause the droplet size to be higher as the temperature increases. At 400 K the last droplet size obtained was $2.2 \times 10^{-4}$ cm, at 500 K the final droplet size obtained was $4.60 \times 10^{-5}$ cm, while at 600 K the last droplet size obtained was $4.93 \times 10^{-5}$ cm. The effect of the reactor wall temperature on the air temperature around the droplet can be seen in figure 9.

Figure 9 shows that the air temperature was transitioned from 299 K to 400 K, 500 K and 600 K according to the variation in the reactor wall temperature. The higher the temperature of the reactor wall, the higher the air temperature around the droplet. The air temperature profile will affect the droplet temperature profile. The droplet temperature profile can be seen in figure 10. As with air temperature, the higher the reactor wall temperature, the higher the droplet surface temperature in its steady state. The temperature transition rate is the same since the $Q$ value in this simulation is the same.

The profile of water vapor concentration in the air around the droplet can be seen in figure 11. The result shows that the smaller the reactor wall temperature, the more water vapor is produced, result in a faster water vapor concentration rate. At 400 K the concentration of the final water vapor produced is $8.2235 \times 10^{20}$ molecules / cm$^3$, at 500 K the concentration of the final water vapor produced is $8.13142 \times 10^{20}$ molecules / cm$^3$ and at 600 K the final water vapor concentration produced is $8.04199 \times 10^{20}$ molecules / cm$^3$.

![Figure 8](image1.png)  **Figure 8.** Effect of reactor wall temperature on the evolution of the size of NaCl droplets throughout the reactor.

![Figure 9](image2.png)  **Figure 9.** Effect of reactor wall temperature on the air temperature around the droplet.
Figure 10. Effect of reactor wall temperature on the temperature of the droplet.

Figure 11. Effect of reactor wall temperature on the concentration of water vapor around the droplet.

The water vapor concentration profile on the droplet surface can be seen in figure 12. The result shows that the higher the reactor wall temperature is given, the less the amount of water vapor concentration attached to the droplet surface. This is because as the temperature increases, the kinetic energy of water vapor attached to the droplet surface become higher so that the water vapor can diffuse toward the surface into the air. The concentration profile of the solution on droplets can be seen in figure 13. The result shows that the concentration on the surface of the droplet is higher as the reactor wall temperature increases since the higher the reactor wall temperature, the higher the droplet temperature. As the droplet temperature increases, the diffusion coefficient between the solvent (water) and the gas around the droplet becomes increase, causing more solvents to become water vapor particles and causing the concentration of the solution on the droplet surface increases.

Figure 12. Effect of reactor wall temperature on water vapor concentration on the droplet surface.

Figure 13. Effect of reactor wall temperature on solution concentration in droplets.
3.3. Variation in Co (initial concentration)

The initial concentration of the solution are varied by 0.1 mol/l, 0.5 mol/l, 1 mol/l, and 2 mol/l. The simulation results of this different solution concentration can be seen in figure 14. In the figure, it can be seen that the concentration of the solution in the droplet is different for each condition as the initial concentration were varied.

![Figure 14](image1.png)  ![Figure 15](image2.png)

**Figure 14.** Effect of the initial concentration of the solution on the concentration of the solution on the droplet.

**Figure 15.** Effect of initial concentration of solution on the evolution of the size of NaCl droplets throughout the reactor.

In figure 14, changes in the solution concentration at the droplet center or at the surface of the droplet are not visible. However, this is not the case since from the center of the droplet until the surface of the droplet distribution the concentration of the solution changes very little. While the effect of variations in solution concentration on droplet size can be seen in figure 15. In the picture, it can be seen that each variation in the value of the initial solution concentration does not affect the final particle size and does not affect the length of the reactor required by the droplet to reach its final size.

![Figure 16](image3.png)  ![Figure 17](image4.png)

**Figure 16.** Effect of the initial concentration of the solution on the air temperature in the reactor tube.

**Figure 17.** Effect of the initial concentration of the solution on the temperature on the droplet.
Similarly, the air temperature, droplet temperature, water vapor concentration in the air around the droplet, and the concentration of water vapor on the droplet surface, all of which do not change with variations in the concentration of the solution. The profile can be seen in figure 16-19.

**Figure 18.** Effect of the initial concentration of the solution on the concentration of water vapor on the surface of the droplet.

**Figure 19.** Effect of the initial concentration of the solution on the concentration of water vapor in the air around the droplet.

### 4. Conclusion

The dynamics of the droplets have been successfully simulated by the influenced of reactor wall temperature ($T_w$), the flow rate of carrier gas ($Q$), and initial concentration ($C_0$). Variation $Q$ affects the flow rate of the carrier gas carrying the droplet and the length required for the droplets to reach their final shape, but that variation does not affect the final size of the particle. $T_w$ variations affect the flow rate of carrier gas carrying droplets and the size of droplets. $C_0$ variation does not affect the flow rate of the carrier gas transporting the droplet so that the length of the reactor required by droplet to reach its final size is fixed for each variation. In this modeling $C_0$ variation seems to have no effect on the droplet size.

### Nomenclature

- $b$: average molecular droplet (mol/g)
- $C_0$: initial solution concentration or solute concentration at the center of the droplet (mol/l)
- $C_{pa}$: heat capacity of surrounding gas (J/mol K)
- $C_p$: heat capacity of droplet (J/mol K)
- $CSS$: critical supersaturation concentration (mol/l)
- $C$: solute concentration at a radius $r$ (mol/l)
- $d_{p0}$: initial droplet diameter (cm)
- $d_{ps}$: equivalent diameter of a complete drying droplet (cm)
- $D_L$: solute diffusion coefficient (cm/s)
- $D_v$: solvent vapor diffusion coefficients (cm/s)
- $ES$: equilibrium saturation concentration (mol/l)
- $F$: average molar flow of air (mol/s)
- $H_L$: latent heat of vaporization (J/g)
- $h_s$: heat-transfer coefficient at the droplet surface (J/cm² s K)
\( h_w \) heat-transfer coefficient at the reactor wall (J/cm² s K)
\( K \) constant, cm²/s
\( K_m \) vapor mass transfer coefficient for laminar flow (cm/s)
\( L \) reactor length (cm)
\( m_i \) mass of water molecule (g/mol)
\( M_s \) solute molecular weight (g/mol)
\( N_0 \) vapor concentration at the droplet surface (no./cm³)
\( N_A \) Avogadro constant (no./mol)
\( n \) vapor concentration in surrounding gas (molecules/cm³)
\( n_s \) vapor concentration at the droplet surface (molecules/cm³)
\( p_s \) droplet surface vapor pressure (dyne/cm²)
\( p_s^0 \) equilibrium vapor pressure (dyne/cm²)
\( Q \) carrier gas flow rate (l/min)
\( R \) reactor tube radius (cm)
\( r \) radial coordinate (cm)

\( RH^0 \) relative humidity in surrounding gas (-)
\( r_p \) droplet radius (cm)
\( R_{p0} \) initial droplet radius (cm)
\( t \) time (s)
\( T \) temperature of surrounding gas (K)
\( T_0 \) initial temperature of reactor inlet (K)
\( T_w \) temperature at the reactor wall (K)
\( T_S \) temperature at the droplet surface (K)
\( T_{wo} \) vapor mole fraction in surrounding gas (-)
\( T_w^0 \) vapor mole fraction in surrounding gas at the reactor inlet (-)
\( z \) Reactor coordinate distance (cm)
\( \rho_L \) density of the pure water (g/cm³)
\( \rho_S \) dissolved density (g/cm³)
\( \rho_v \) density of the droplet (g/cm³)

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