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<th>数値解析手法を用いた粒子間の親付着力評価その1: 試験結果とその考察について</th>
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<td>菅 裕之</td>
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Numerical Analysis of Particle-Particle Adhesion by Dynamic Liquid Bridge

Hiroyuki KAN

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Doctoral Thesis at Osaka Prefecture University
Acknowledgements

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Chapter I

Introduction
1. General introduction

In many powder handling processes, particle adhesion is a critical phenomenon, which determines performance of the process and quality of the final products. Generally, the interparticle adhesion forces are classified into three types: van der Waals force, electrostatic force, and liquid bridge force [1-3]. Among these forces, the liquid bridge force can be dominant in many cases, because magnitude of the liquid bridge force is much higher than the other forces by a few orders [2]. Therefore, understanding of the adhesion phenomenon mediated by a liquid bridge becomes an important issue. Although numerous studies have been conducted from a century ago, many issues regarding the liquid bridge adhesion remain to be elucidated.

In many industrial areas, the liquid bridge adhesion can cause many troubles. For example, in a powder mixing process, undesirable adhesion between particles and between particle and equipment wall can often occur due to the liquid bridge. This largely reduces the yield and quality of the final powder mixture. On the other hand, there are many powder handling processes where the liquid bridge adhesion is positively utilized. This is known as a wet granulation, in which the liquid bridge adhesion is employed to produce assembly of original fine particles. A wet granulation is used in so many industrial areas such as chemical, food, agricultural, and pharmaceutical to improve the flowability, handling efficiency, and uniformity of the active ingredients. The granules are produced by means of various types of processes, including a fluidized bed, high-shear mixer, and rotating drum [3, 4]. Fig. 1-1 shows schematic illustration of particle agglomeration in a typical process (fluidized bed spray granulation). Binder droplets are firstly adhered to original particles, followed by wetting and nucleation. The particles then collide with each other through binder droplets, and the particles are adhered by a liquid bridge. By repeating these processes, the original particles coalesce with each other to promote granule growth. Subsequently, a liquid bridge is transformed to a solid bridge after drying of liquid, and dry granules are finally produced. The effect of operating parameters of the process and physicochemical properties of both binder liquid and particle on the physical properties of granules such as size distribution and structure have been experimentally investigated.
(e.g., [5-13]). These physical properties of granules determine quality of the final products. Therefore, understanding of how the agglomerated particles are formed from original primary particles is very important. In a wet granulation process, the particle-particle collision mediated by a binder droplet and subsequent particle-particle adhesion by the liquid bridge is the most fundamental phenomenon. Thus, elucidation of mechanisms for the particle-particle adhesion by the liquid bridge is essential to control the wet granulation and would open up new directions in the next-generation wet granulation technologies.

Historically, numerous studies on the liquid bridge adhesion force have been reported. A lot of theoretical and analytical models for the liquid bridge force have been proposed. Most of the conventional theories and analytical models cover the “static” liquid bridge, where shape of the liquid bridge does not temporally deform, i.e., steady-state liquid bridge. However, in an actual wet granulation process, the particles continuously move due to mechanical external forces, resulting in deformation of the liquid bridge (e.g., compression, elongation, and rupture) as shown in Fig. 1-2. This implies that the liquid bridge is not static but dynamic in an actual process. The “dynamic” liquid bridge means that shape of the liquid bridge temporally deforms, i.e., unsteady-state liquid bridge. There are some theoretical and experimental studies on the dynamic liquid bridge. However, these previous studies focused on the liquid bridge force and deformation of the liquid bridge. Therefore, the particle-particle adhesion, i.e., whether the particles are adhered by the dynamic liquid bridge, has not been investigated, although the particle adhesion phenomenon should be investigated for the comprehensive understanding of particle growth in wet granulations. This is because that it is too difficult to theoretically and experimentally analyze the microscopic particle-particle adhesion phenomenon (primary scale particle adhesion phenomenon as shown in Fig. 1-2) due to their complexities.

In such a case, a numerical simulation can be a promising approach. To simulate the particle-particle adhesion by a dynamic liquid bridge, a multi-phase flow including solid-liquid-gas three-phase flow should be calculated. Recently, the dynamic liquid bridge has been analyzed using a direct numerical simulation. However, the microscopic
particle-particle adhesion phenomenon has not been studied.

This thesis is devoted to a numerical modeling of particle-particle adhesion by a dynamic liquid bridge and elucidation of its physical mechanism. The particle adhesion phenomenon can be affected by various factors, including physical properties of particle and liquid, mode and intensity of particle-particle collision. In this thesis, effects of particle collision velocity, particle wettability, binder droplet size, and collision angle on particle-particle adhesion by the dynamic liquid bridge were analyzed and role of these factors in the particle adhesion was theoretically elucidated.
Fig. 1-1. Particle agglomeration process in wet granulation.
Fig. 1-2. Deformation of liquid bridge in actual wet granulation process.
2. Review of previous works

Historically, numerous studies on the liquid bridge between particles have been conducted. First of all, modeling and experimental studies on the static liquid bridge are reviewed. Second, modeling and experimental studies on the dynamic liquid bridge are reviewed. Finally, direct numerical simulations of the multi-phase flow including the dynamic liquid bridge are reviewed.

2.1. Configurations of liquid bridge between particles

Configurations of the liquid bridge between particles are classified by a degree of liquid saturation [4], as shown in Fig. 1-3. In a pendular state (Fig. 1-3 (a)), each liquid bridge exists only between two particles. In a capillary state (Fig. 1-3 (c)), voids between particles are fully filled with liquid and the surface liquid is drawn back into the pores due to capillary force. A funicular state (Fig. 1-3 (b)) is a transition state between pendular and capillary where voids between particles are partially filled with liquid. In a slurry state (Fig. 1-3 (d)), voids between particles are filled with liquid and particles are fully covered with liquid. In this thesis, the pendular liquid bridge between two particles was focused, because the particle-particle adhesion by the pendular liquid bridge is the most fundamental phenomenon in a wet granulation.
Fig. 1-3. Configurations of liquid bridge between particles.
2.2. Modeling and experimental studies on static liquid bridge

There are numerous studies on the static liquid bridge. The static liquid bridge force acting on a stable particle has been analyzed theoretically and experimentally. Generally, the static liquid bridge force is derived from a sum of two forces: (i) surface tension force and (ii) capillary pressure force. The surface tension force acts to reduce its surface area at a gas-liquid interface. The capillary pressure force is derived from the pressure difference between inside and outside of the liquid bridge. Mathematical expressions of the static liquid bridge force have been derived by the following two methods [14]:

Boundary method
\[
F_{LB} = 2\pi (R_p \sin \alpha_{hf}) \sigma \sin(\theta + \alpha_{hf}) + \pi (R_p \sin \alpha_{hf})^2 \Delta P
\]  \hspace{1cm} (1-1)

Gorge method
\[
F_{LB} = 2\pi R_2 \sigma + \pi R_2^2 \Delta P
\]  \hspace{1cm} (1-2)

where \(F_{LB}\) is the static liquid bridge force, \(R_p\) is the particle radius, \(\alpha_{hf}\) is the half-filling angle, \(\theta\) is the contact angle, \(\sigma\) is the surface tension coefficient, \(\Delta P\) is the pressure difference between inside and outside of the liquid bridge, and \(R_2\) is the neck radius of the liquid bridge. These symbols are shown in Fig. 1-4. In Eqs. (1-1) and (1-2), the first and second terms of the right-hand side indicate the surface tension force and capillary pressure force, respectively. In the boundary method (Eq. (1-1)), the liquid bridge force is estimated at the contact line (i.e., gas-liquid-solid three-phase interface). In the Gorge method (Eq. (1-2)), the liquid bridge force is estimated at the neck of the liquid bridge. Among these two methods, the boundary method (Eq. (1-1)) is often used, because effect of the contact angle can be considered. The pressure difference \(\Delta P\) is described by the Young-Laplace equation as follows:
\[ \Delta P = \sigma \left( \frac{1}{r_1} - \frac{1}{r_2} \right) \]  
\hspace{1cm} (1-3)

\[ r_1 = \sqrt{1 + \left( \frac{dy}{dx} \right)^2} \frac{3}{2} \]  
\hspace{1cm} (1-4)

\[ r_2 = y \left( 1 + \left( \frac{dy}{dx} \right)^2 \right)^{1/2} \]  
\hspace{1cm} (1-5)

where \( r_1 \) and \( r_2 \) are radius of curvature of the gas-liquid interface, \( x \) and \( y \) are coordinates along the gas-liquid interface, shown in Fig. 1-4. Generally, this non-linear Young-Laplace equation is difficult to be solved analytically. Therefore, approximate or numerical approaches were adapted to solve the Young-Laplace equation. In most cases, the effect of gravity was not taken into account.

Table 1-1 shows summary of theoretical studies on the static liquid bridge. Modeling study of the liquid bridge force was started from Haines [15] and Fisher [27] in one hundred years ago. As an approximation approach, a toroidal approximation, which assumes shape of the liquid bridge (gas-liquid interface) as a circular arc (red line in Fig. 1-5), has been adapted. Many researchers adapted this approximation and analyzed the properties of the static liquid bridge [15-26]. However, it was pointed out that the toroidal approximation cannot express the capillary pressure inside the liquid bridge with sufficient accuracy [35]. To more accurately approximate the shape of the gas-liquid interface, a nodoidal approximation (blue line in Fig. 1-5) has been adapted. In the nodoidal approximation, curvature of the gas-liquid interface is treated as a constant \( R_1 \), which is defined at the neck of the liquid bridge. Using this approximation method, properties of the static liquid bridge have been analyzed [27-31]. However, despite the nodoidal approximation, accuracy of the liquid bridge force is deteriorated at higher liquid bridge volume and larger separation distance between particles [34].
Fig. 1-4. Schematic of liquid bridge between spherical particles.
**Table 1-1 Theoretical studies on static liquid bridge**

<table>
<thead>
<tr>
<th>Author</th>
<th>Treatment of liquid bridge shape</th>
<th>Subject</th>
<th>Ref. No.</th>
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<td>Approx. (Toroid)</td>
<td>Liquid bridge force</td>
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</tr>
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<td>Pietsch and Rumpf (1967)</td>
<td>Approx. (Toroid)</td>
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<td>16</td>
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<td>Erle et al. (1971)</td>
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<td>Liquid bridge force</td>
<td>17</td>
</tr>
<tr>
<td>Mehrotra and Sastry (1980, 1985)</td>
<td>Approx. (Toroid)</td>
<td>Liquid bridge force</td>
<td>18, 19</td>
</tr>
<tr>
<td>Israelachvili (1991)</td>
<td>Approx. (Toroid)</td>
<td>Liquid bridge force</td>
<td>20</td>
</tr>
<tr>
<td>Rabinovivh et al. (2002, 2005)</td>
<td>Approx. (Toroid)</td>
<td>Liquid bridge force</td>
<td>21, 22</td>
</tr>
<tr>
<td>Butt and Kapple (2009)</td>
<td>Approx. (Toroid)</td>
<td>Liquid bridge force</td>
<td>23</td>
</tr>
<tr>
<td>Chen et al. (2011)</td>
<td>Approx. (Toroid)</td>
<td>Liquid bridge force, liquid bridge volume</td>
<td>24</td>
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<tr>
<td>Rose (1958)</td>
<td>Approx. (Toroid)</td>
<td>Liquid bridge volume, surface area</td>
<td>25</td>
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<tr>
<td>Dai and Lu (1998)</td>
<td>Approx. (Toroid)</td>
<td>Rupture distance</td>
<td>26</td>
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<tr>
<td>Fisher (1926)</td>
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<td>27</td>
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<td>Pitois et al. (2002)</td>
<td>Approx. (Nodoid)</td>
<td>Liquid bridge force, rupture energy</td>
<td>28</td>
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<tr>
<td>Simons et al. (1994)</td>
<td>Approx. (Nodoid)</td>
<td>Rupture energy</td>
<td>29</td>
</tr>
<tr>
<td>Melrose (1966)</td>
<td>Approx. (Nodoid)</td>
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<td>30</td>
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<td>Bisschop and Rigole (1982)</td>
<td>Approx. (Nodoid)</td>
<td>Liquid bridge shape</td>
<td>31</td>
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<td>Pepint et al. (2000)</td>
<td>Approx. (Parabolic curve)</td>
<td>Liquid bridge shape</td>
<td>32</td>
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<td>Heady and Cahn (1970)</td>
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<td>Hotta et al. (1974)</td>
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<td>Mazzone et al. (1986)</td>
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<td>Liquid bridge force, liquid bridge shape</td>
<td>34</td>
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<td>Lian et al. (1993, 2016)</td>
<td>Numerically solved</td>
<td>Liquid bridge force, rupture distance</td>
<td>37, 38</td>
</tr>
<tr>
<td>Mikami et al. (1998)</td>
<td>Numerically solved</td>
<td>Liquid bridge force, rupture distance</td>
<td>39</td>
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<tr>
<td>Soulie et al. (2006)</td>
<td>Numerically solved</td>
<td>Extended from Mikami et al. (1998)</td>
<td>40</td>
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<tr>
<td>Adams et al. (2002)</td>
<td>Numerically solved</td>
<td>Liquid bridge force, rupture distance</td>
<td>41</td>
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<tr>
<td>Pakarnen et al. (2005)</td>
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<td>Liquid bridge force</td>
<td>42</td>
</tr>
<tr>
<td>Orr et al. (1975)</td>
<td>Numerically solved Approx. (Toroid)</td>
<td>Liquid bridge shape, curvature</td>
<td>43</td>
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<tr>
<td>Kousaka et al. (1992)</td>
<td>Numerically solved Approx. (Toroid)</td>
<td>Liquid bridge shape</td>
<td>44</td>
</tr>
<tr>
<td>Endo et al. (1992)</td>
<td>Numerically solved</td>
<td>Liquid bridge shape, liquid bridge volume</td>
<td>45</td>
</tr>
</tbody>
</table>
Fig. 1-5. Approximation shape of liquid bridge.
Therefore, the nodoidal approximation can be adapted when the liquid volume is small and/or when the separation distance is small. To overcome this limitation, the Young-Laplace equation has been numerically solved without any shape approximations [14, 33-45]. In these approaches, shape of the gas-liquid interface can be expressed with high accuracy regardless of the liquid bridge volume and separation distance.

By solving the Young-Laplace equation, properties of the liquid bridge including liquid bridge force acting on a particle, liquid bridge shape, volume and surface area, rupture distance, and rupture energy have been analyzed. Many researchers focused on modeling of the liquid bridge force [14-24, 27, 28, 33-42]. Erle et al. [17] calculated the liquid bridge force obtained from the condition where the surface energy of the gas-liquid interface becomes the minimum value. Mehrotra and Sastry [18, 19] modeled the liquid bridge force between unequal-sized spherical particles. Israelachvili [20] modeled the liquid bridge force as a function of particle diameter, contact angle, and separation distance from the total energy of the liquid bridge. It was pointed out that the surface tension force was not considered in Israelachvili’s model [22]. Rabinovich et al. [22] formulated the liquid bridge force from a total liquid bridge energy with solving the Young-Laplace equation. In their model, surface tension effect was added to the equation proposed by Israelachvili [20], and they indicated that surface tension force does not play a significant role in the case of a small liquid volume. Endo et al. [36] modeled the liquid bridge force by numerically integrating the Young-Laplace equation. Their equation was same as Fisher’s equation which was obtained using a shape approximation. Mikami et al. [39] numerically solved the Young-Laplace equation under various conditions. They modeled the liquid bridge force between equal-sized spherical particles, which was expressed by a liquid bridge volume, contact angle, and surface distance between particles, by regression analysis using numerical solutions of the Young-Laplace equation. Soulie et al. [40] extended Mikami model [39] to adapt for unequal-sized particles. Table 1-2 shows representative models of the static liquid bridge force. $\tilde{F}_{LB}$, $\tilde{V}_{LB}$, $\tilde{h}$ and $\tilde{\tilde{h}}$ are the dimensionless liquid bridge force ($= F_{LB}/\pi R_p \sigma$), dimensionless liquid bridge volume ($= V_{LB}/R_p^3$), surface distance between particles, and dimensionless distance between particles ($= h/R_p$), respectively.
Comments on the accuracy of the estimated liquid bridge force are also described. These three models are regarded as reliable models and frequently used for estimation of the liquid bridge force between particles.

Liquid bridge shape, volume, and surface area are also analyzed [24, 25, 30-32, 34-36, 43-45]. Bisschop and Rigole [31] formulated a nodoidal shape of the gas-liquid interface as a function of separation distance, particle diameter, contact angle, and liquid bridge volume. Orr et al. [43] solved the Young-Laplace equation in terms of elliptic integrals for all possible types of liquid bridge between sphere and flat substrate. They analyzed the liquid bridge shape, curvature, and area and volume of the liquid bridge. Mazzone et al. [34] considered the effect of gravity by Bond number, which is the ratio of gravity force to surface tension. They indicated that gravity has a significant effect on the liquid bridge shape, when the Bond number \((= \Delta \rho g R_p^2/\sigma)\) is of the order unity and larger. Kousaka et al. [44] and Endo et al. [45] theoretically analyzed the mechanism of liquid bridge formation due to condensation by solving the Young-Laplace equation combined with Kelvin equation.

Rupture distance of the static liquid bridge is also important property [37, 39-41]. Lian et al. [37] numerically solved the Young-Laplace equation by using an Euler method and modeled the rupture distance of the liquid bridge. They simply modeled the rupture distance as a function of contact angle and liquid bridge volume. Their equation is often used in modeling of the static liquid bridge. Mikami et al. [39] proposed a similar equation with Lain et al. [37]. Adams et al. [41] formulated the rupture distance considering the effect of gravity by Bond number. Rupture energy of the liquid bridge was calculated by Simons et al [29] and Pitois et al. [28]. Simons et al. [29] obtained the rupture energy integrating the liquid bridge force proposed by Fisher [27]. Pitois et al. [28] added the effect of viscous force to the rupture energy proposed by Simons et al. [29].

The static liquid bridge force was also experimentally measured. Table 1-3 shows summary of experimental studies on the static liquid bridge. Mason and Clark [46] and Clark et al. [47] measured the liquid bridge force between particles and between particle and flat substrate using a hydrometer. They used two immiscible liquids and the liquid
bridge was formed in water. Measured liquid bridge force was similar with that of theoretical value obtained by Fisher’s model [27]. Willett et al. [49] measured the liquid bridge force and separation distance by recording microbalance force and displacement of the actuator, respectively. They investigated effects of the liquid volume and particle size on the liquid bridge force as a function of separation distance. Rabinovich et al. [21, 22] measured the liquid bridge force between individual glass spheres with 20 to 50 µm in diameter by using an atomic force microscopy (AFM). They compared experimental results with their theoretical model and both results were found to agree. Rossetti et al. [50] used a micro force balance (MFB) developed by Fairbrother and Simons [53] to analyze shape and force of the liquid bridge. Lambert et al. [51] observed the liquid bridge shape using a CCD camera and simultaneously measured the liquid bridge force using an AFM. They also compared experimental results with theoretical results to validate the previous theoretical models [20, 22].

Numerous investigations of the static liquid bridge have been conducted by many researchers. However, in actual powder handling processes, the liquid bridge formed between particles shows dynamic deformation due to moving particles. Therefore, to understand the particle adhesion phenomenon in actual processes, the dynamic liquid bridge should be studied.
Regression analysis was conducted using solutions of the Young-Laplace equation in the range of \( V_{LB} \) = 2.0×10\(^{-3} \) – 2.0×10\(^{-2} \), \( \theta = 0 \) – 50 deg. Within these ranges, estimated liquid bridge force is consistent with an experimental result with high accuracy regardless of the separation distance between particles.

At the small liquid bridge volume, estimated liquid bridge force is consistent with an experimental result. At the high liquid bridge volume, the accuracy decreases for small and large separation distance.

At the small liquid bridge volume, estimated liquid bridge force is consistent with an experimental result. At the high liquid bridge volume, the liquid bridge force is overestimated compared with an experimental result.

<table>
<thead>
<tr>
<th>Author</th>
<th>Equation</th>
<th>Accuracy</th>
<th>Ref. No.</th>
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<tr>
<td>Mikami et al. (1998)</td>
<td>( \hat{F}_{LB} = \exp(A \hat{h} + B) + C )</td>
<td>Regression analysis was conducted using solutions of the Young-Laplace equation in the range of ( V_{LB} ) = 2.0×10(^{-3} ) – 2.0×10(^{-2} ), ( \theta = 0 ) – 50 deg. Within these ranges, estimated liquid bridge force is consistent with an experimental result with high accuracy regardless of the separation distance between particles.</td>
<td>39</td>
</tr>
<tr>
<td>Israelachvili (1991)</td>
<td>( F_{LB} = \frac{2\pi R_p \sigma \cos \theta}{1 + h/2d} )</td>
<td>At the small liquid bridge volume, estimated liquid bridge force is consistent with an experimental result. At the high liquid bridge volume, the accuracy decreases for small and large separation distance.</td>
<td>20</td>
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<tr>
<td>Rabinovich et al. (2005)</td>
<td>( F_{LB} = \frac{2\pi R_p \sigma \cos \theta}{1 + h/2d} )</td>
<td>At the small liquid bridge volume, estimated liquid bridge force is consistent with an experimental result. At the high liquid bridge volume, the liquid bridge force is overestimated compared with an experimental result.</td>
<td>22</td>
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<table>
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<tr>
<th>Table 1-3 Experimental studies on static liquid bridge force</th>
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<tr>
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<tr>
<td>Mason and Clark (1965)</td>
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<td>Clark et al. (1968)</td>
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<td>Gillespie and Settineri (1967)</td>
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<td>Willet et al. (2000)</td>
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<td>Rabinovich et al. (2002, 2005)</td>
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<td>Rossetti et al. (2003)</td>
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<td>Lambert et al. (2005, 2008)</td>
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2.3. Modeling and experimental studies on dynamic liquid bridge

There are a few studies on the dynamic liquid bridge. Table 1-4 shows summary for theoretical investigation of the dynamic liquid bridge. Deformation of the liquid bridge derived from moving particle and dynamic liquid bridge force acting on a particle have been analyzed analytically and experimentally.

In a case of the dynamic liquid bridge, influence of the viscous force is included as a liquid bridge force. Ennis et al. [54] modeled simplified solution of dynamic liquid bridge force based on a lubrication theory and toroidal approximation of the liquid bridge shape. They indicated that magnitude of the dynamic liquid bridge force exceeds the static one by over an order of magnitude, although liquid bridge shape was relatively similar between dynamic and static cases. Balakin et al. [55] also modeled the dynamic liquid bridge force as sum of the static liquid bridge force proposed by Rabinovich et al. [22] and viscous force derived from a lubrication force. They analyzed the particle behavior during approach and separation of two particles by solving the equation of motion which considered the dynamic liquid bridge force as an external force. These are major approaches for modeling of the dynamic liquid bridge force.

Valsamis et al. [56] and Darabi et al. [57] used different approaches. Valsamis et al. [56] proposed a mechanical model. In their model, the liquid bridge force is modeled by a Kelvin-Voigt model, consisting of a spring, a dashpot, and a mass in parallel. Parameters for these mechanical elements were provided by an analytical approximation based on simplifications of the Navier-Stokes equation. Darabi et al. [57] proposed a simplified model for the liquid bridge shape by using a parabolic approximation. They analyzed the quasi-static elongation of the liquid bridge between two spherical particles and rupture distance of the liquid bridge.

Ennis et al. [58] also studied critical condition of particle adhesion due to a dynamic liquid bridge force. They proposed a model based on a lubrication theory, in which the particle-particle adhesion by the dynamic pendular liquid bridge was simplified as an adhesion phenomenon between particles completely covered with a thin liquid film as shown in Fig. 1-6. In their model, the critical velocity for particle adhesion was obtained by following equation:
Table 1–4 Theoretical studies on dynamic liquid bridge

<table>
<thead>
<tr>
<th>Author</th>
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<th>Subject</th>
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<tbody>
<tr>
<td>Ennis et al. (1990)</td>
<td>Sum of static liquid bridge force and viscous force</td>
<td>Liquid bridge force</td>
<td>54</td>
</tr>
<tr>
<td>Balakin et al. (2013)</td>
<td>Sum of static liquid bridge force and viscous force</td>
<td>Liquid bridge force</td>
<td>55</td>
</tr>
<tr>
<td>Valsamis et al. (2013)</td>
<td>Mechanical model</td>
<td>Liquid bridge force</td>
<td>56</td>
</tr>
<tr>
<td>Darabi et al. (2010)</td>
<td>Parabolic approximation</td>
<td>Liquid bridge shape</td>
<td>57</td>
</tr>
<tr>
<td>Ennis et al. (1991)</td>
<td>Lubrication theory</td>
<td>Critical collision velocity for particle adhesion</td>
<td>58</td>
</tr>
<tr>
<td>Liu et al. (2000)</td>
<td>Lubrication theory</td>
<td>Critical collision velocity for particle adhesion</td>
<td>59</td>
</tr>
</tbody>
</table>

Fig. 1–6. Schematic of model used by Ennis et al. [58].
\[ v_c = \frac{3\pi \mu R_p^2}{2m_p} \left( 1 + \frac{1}{e} \right) \ln \left( \frac{z}{z_a} \right) \]  

(1-6)

where \( \mu \), \( R_p \), \( m_p \), \( e \), \( z \) and \( z_a \) are the liquid viscosity, particle radius, particle mass, restitution coefficient between solid particles, liquid film thickness, and surface roughness, respectively. Liu et al. [59] extended this model by considering the particle plastic deformation during the particle-particle collision. These two models are useful for considering coalescence of the particles, because the particle adhesion phenomenon due to the dynamic liquid bridge can be quantitatively explained. However, deformation of the liquid was not taken into account. Therefore, the particle-particle adhesion by the dynamic liquid bridge has not been modeled yet.

Experimental studies on the dynamic liquid bridge between spherical particles have been conducted [60-65]. Table 1-5 summarizes experimental studies on the dynamic liquid bridge. Major analysis of the dynamic liquid bridge was liquid bridge force acting on a particle and rupture behavior for the liquid bridge between vertically or horizontally arranged two particles. Mazzone et al. [60] and Ennis et al. [54] analyzed the dynamic liquid bridge force between oscillating particles. They indicated that magnitude of the dynamic liquid bridge force exceeds that of static liquid bridge force by an order of magnitude. Mazzone et al. [60] also analyzed rupture behavior of the liquid bridge between millimeter scale particles. They indicated that the shape and rupture point of the dynamic liquid bridge were quite different from those of the static liquid bridge. Simons and Fairbrother [62] observed rupture behavior of the liquid bridge between 10 to 100 \( \mu m \) radius glass ballotini and measured dynamic liquid bridge force. Some researchers analyzed the liquid bridge between multiple spherical particles [63-65]. Murase et al. [63] and Lievano et al. [65] measured the dynamic liquid bridge force acting on a moving particle between millimeter-scale three spheres.

In wet granulation processes, collisions of particles with other particles play an important role. From this background, collisions between a particle and a thin liquid film were analyzed by experimental studies [66-70]. Collisions and rebound for small metal and plastic balls impacting a flat substrate covered with a thin viscous liquid film.
<table>
<thead>
<tr>
<th>Author</th>
<th>Experimental system</th>
<th>Measured properties</th>
<th>Ref. No.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mazzone et al. (1987)</td>
<td>Liquid bridge between vertically arranged two particles</td>
<td>Liquid bridge force, rupture behavior</td>
<td>60</td>
</tr>
<tr>
<td>Ennis et al. (1990)</td>
<td>Liquid bridge between vertically arranged two particles</td>
<td>Liquid bridge force</td>
<td>54</td>
</tr>
<tr>
<td>Zhuang and Ju (2015)</td>
<td>Liquid bridge between vertically arranged two particles</td>
<td>Liquid bridge force</td>
<td>61</td>
</tr>
<tr>
<td>Simons and Fairbrother (2000)</td>
<td>Liquid bridge between horizontally arranged two particles</td>
<td>Liquid bridge force, rupture behavior</td>
<td>62</td>
</tr>
<tr>
<td>Murase et al. (2004, 2008)</td>
<td>Liquid bridge between three particles</td>
<td>Liquid bridge force</td>
<td>63, 64</td>
</tr>
<tr>
<td>Lievano et al. (2017)</td>
<td>Liquid bridge between two or three particles</td>
<td>Liquid bridge force</td>
<td>65</td>
</tr>
<tr>
<td>Davis et al. (2002)</td>
<td>Collision between particle and liquid film</td>
<td>Wet restitution coefficient</td>
<td>66</td>
</tr>
<tr>
<td>Kantak et al. (2004)</td>
<td>Collision between particle and liquid film</td>
<td>Wet restitution coefficient</td>
<td>67</td>
</tr>
<tr>
<td>Antonyuk et al. (2009)</td>
<td>Collision between particle and liquid film</td>
<td>Wet restitution coefficient</td>
<td>68</td>
</tr>
<tr>
<td>Sutkar et al. (2015)</td>
<td>Collision between particle and liquid film</td>
<td>Wet restitution coefficient</td>
<td>69</td>
</tr>
<tr>
<td>Ma et al. (2016)</td>
<td>Collision between particle and liquid film</td>
<td>Wet restitution coefficient</td>
<td>70</td>
</tr>
</tbody>
</table>
was experimentally analyzed by Davis et al. [66]. A wet restitution coefficient as a function of Stokes number was obtained from experimental results. Kantak et al. [67] and Ma et al. [70] treated oblique collisions between spheres and wetted surface and separately measured normal and tangential wet restitution coefficients. Some theoretical models of wet restitution coefficient were proposed by Antonyuk et al. [68] and Sutkar et al. [69]. Antonyuk et al. [68] solved the equation of translational motion of a spherical particle and obtained simulation results of wet restitution coefficient. Sutkar et al. [69] developed an expression for wet restitution coefficient through dimensional analysis and an energy analysis.

In the analytical approaches, the dynamic liquid bridge force was obtained based on the static liquid bridge force and viscous force. The static liquid bridge force was calculated assuming that the liquid bridge is in a steady-state at the moment, while the viscous force was calculated from the liquid viscosity and not considered deformation of the liquid bridge. Therefore, unsteady deformation of the liquid bridge was not strictly taken into account. In the experimental approaches, one particle was basically fixed and particle scale was larger than the particles which were used in an actual wet granulation due to experimental limitations. Therefore, the previous studies for the dynamic liquid bridge still have a gap from the actual particle-particle adhesion phenomenon. In the analysis of the microscopic particle-particle adhesion by the dynamic liquid bridge, a direct numerical simulation can be a powerful approach. A direct numerical simulation can strictly track the unsteady deformation of the liquid bridge derived from the particle motion, and the particles can freely move like an actual wet granulation process.
2.4. Direct numerical simulation of multi-phase flow

To simulate the particle-particle adhesion by a dynamic liquid bridge, motions of gas, liquid, and solid which compose multi-phase flow are needed to be solved. Especially, deformation of gas-liquid interface should be exactly tracked. In such a case, a direct numerical simulation (DNS) can be used. In a direct numerical simulation of a multi-phase flow, a discrete element method (DEM), which is originally proposed by Cundall and Strack [71], is adapted to simulate the solid particle motion, while a computational fluid dynamics (CFD) is adapted to simulate the fluid flow.

A simulation of the solid-fluid two-phase flow has been conducted using a DEM-CFD coupling model. It this simulation, how to consider the interaction between the solid and fluid phase is important. Generally, an immersed boundary (IB) method [72-74] is used. Using these methods, some direct numerical simulations of the solid-fluid two-phase flow have been conducted [75-81]. Pan et al. [75] simulated the fluidization behavior of spheres and confirmed that their simulation results showed good agreement with an experimental result. Deen et al. [76, 77] simulated the fluidization behavior of sphere particles considering heat transfer in a pseudo two-dimensional fluidized bed. Feng et al. [78] also investigated particle fluidization with heat transfer in a liquid-solid bed. In these simulations, the number of particles was from a few hundred to a thousand, because the calculation load of the direct numerical simulation is too high to simulate a large amount of particles.

In a direct numerical simulation of gas-liquid two phase flow, a volume of fluid (VOF) method proposed by Hirt and Nichols [82] and a level set (LS) method proposed by Osher and Serthian [83] are used to track deformation of gas-liquid interface. A coupled level set and volume of fluid (CLSVOF) method was also proposed by Sussman and Puckett [84]. Using these methods, gas-liquid/liquid-liquid two phase flow was simulated [85-88]. Ohta et al. simulated behavior of a gas bubble rising in viscous liquid [85, 86] and deformation and breakup of a droplet rising in an immiscible liquid [87, 88]. Their simulation can express a coalescence and breakup behavior of bubbles. Yokoi [89] used CLSVOF method to simulate droplet impact to a thin liquid layer. In their simulation, behavior of droplets scatter was well simulated. Simulations of a
gas-liquid flow including solid have also been conducted. Collisions between a droplet and a flat substrate including oblique condition [90-94], between a droplet and a particle surface [95], between a dry particle and a thin liquid film [96] were simulated. Bussmann et al. [90, 91] simulated wetting behavior of droplet on a flat substrate by using the VOF method. Their simulation results showed good agreement with experimental results. Pasandideh-Fard et al. [92] added effects of heat transfer and phase-change to the VOF method proposed by Bussmann et al. [90] and simulated wetting behavior of a molten metal droplet on an oblique substrate. Mitra et al. [95] simulated collision between a water droplet and heated brass particle and consequently wetting behavior of the droplet.

A constrained interpolation profile (CIP) method proposed by Yabe et al. [97-99] is also used to track deformation of gas-liquid interface. The CIP method is used to calculate an advection equation for gas-liquid interface with high accuracy. Some researchers simulated solid-liquid-gas three-phase flow by using a CIP method [100-104]. Yabe et al. [100] simulated behavior of a thin aluminum cylinder which is thrown over water surface. Their simulation results were qualitatively similar with experimental results. Nishiura et al. [101, 102] used a CIP method combined with a DEM to simulate drying process of suspensions containing fine solid particles. They investigated the relationship between the morphology of granules and drying conditions. Washino et al. [103] simulated liquid bridge behavior between two static parallel walls and droplet impingement on the solid wall as a fundamental simulation for development of direct numerical simulation model of a wet granulation. Washino et al. [104] also proposed a calculation model of capillary force for a DEM-CIP coupling method. They simulated droplet penetration into a particle bed and collision behavior between a droplet and particles in a high-shear mixer.

Recently, a direct numerical simulation of the dynamic liquid bridge has also been simulated. Darabi et al. [57] simulated quasi-static elongation and rupture behaviors of liquid bridge between particles by using two-dimensional VOF method. They analyzed effects of dimensionless numbers (capillary, Weber, and Bond numbers) on the rupture distance of the liquid bridge. Deganello et al. [105, 106] simulated deformation and
rupture behaviors of a liquid bridge between flat substrates by using a LS method. Their simulation results showed good agreement with experimental results. Ido et al. [107] used a moving particle semi-implicit (MPS) method [108], which is one of the Lagrangian methods, to simulate behavior of the liquid bridge. They simulated rupture behavior of the liquid bridge between spherical particles by using a DEM-MPS method. Their simulation result was relatively similar with an experimental result. However, it was pointed out that stability of their simulation model decreased, when distance between particles become small. Therefore, particle-particle collision mediated by a liquid cannot be calculated in their model. Wu et al. [109, 110] simulated liquid transport between equal-seized and unequal-sized particles by using the VOF method. They extracted the position of the interface from results of DNS simulation and built a dynamic model for predicting bridge volume and the liquid remaining on the particle surface. Sun and Sakai [111] simulated behavior of the liquid bridge between spherical particles by using the VOF coupled with an IB method. They simulated quasi-static stretching and rupture behavior of the liquid bridge between two spherical particles and compared with previous analytical and experimental results. They also calculated the liquid bridge force between multiple (three and four) spherical particles.

Many researchers have investigated the dynamic liquid bridge between particles using a direct numerical simulation. However, the previous studies focused on deformation behavior and liquid bridge force. If the dynamic liquid bridge force is mathematically formulated, whether the particles are adhered by the dynamic liquid bridge can be estimated without solving unsteady deformation of the liquid bridge. However, the mathematical formulation of the dynamic liquid bridge force is too complicated, because various parameters contribute magnitude of the dynamic liquid bridge force and the force is temporally varied unlike a static liquid bridge force. Therefore, to analyze whether the particle-particle adhesion occurs mediated by the dynamic liquid bridge, it is necessary to solve the particle motion which freely move by receiving the dynamic liquid bridge force, like in an actual wet granulation.
3. Scope of this thesis

The purpose of this thesis is to develop a numerical simulation model for a microscopic particle-particle adhesion by the dynamic liquid bridge and elucidate the effect of many critical parameters on mechanism of particle adhesion phenomenon. In this thesis, the pendular liquid bridge between two particles was focused. This thesis consists of six chapters and each content is listed below.

Chapter I describes general background of this thesis. In the first part of this chapter, the general introduction is presented. The second part reviews previous studies related to this thesis. The review consists of theoretical and experimental studies on static liquid bridge and dynamic liquid bridge, and direct numerical simulation of dynamic liquid bridge. The third part of this chapter mentioned the main objective and contents of this thesis.

Chapter II is devoted to numerical modeling of particle-particle adhesion by the dynamic liquid bridge and verification and validation of the simulation model. In this study, motions of the gas and liquid were solved by using a computational fluid dynamics (CFD) approach combined with a constrained interpolation profile (CIP) method, while the particle motion was simulated by a Lagrangian approach considering liquid bridge force that is unsteadily changed due to deformation of the liquid bridge. To verify the simulation results, the following verifications and validations were conducted; (i) wetting behavior of a droplet on a curved surface, (ii) static liquid bridge force between particles, and (iii) rupture behavior of a liquid bridge with free falling of a particle. The particle-particle adhesion though a droplet on a particle surface was then simulated. In particular, effect of a collision velocity ($v$ shown in Fig. 1-7) on a wet restitution coefficient was investigated. A critical velocity for particle adhesion (i.e., adhesiveness of the two colliding particles) was determined from the simulation results and compared with that estimated from a previous analytical model.
Chapter III is concerned with an investigation of effect of particle wettability on the particle-particle adhesion by the dynamic liquid bridge. The particle wettability is a critical factor affecting quality of the granules. Therefore, understanding of effect of the particle wettability is an important issue for rational design of the wet granulation. The particle wettability was evaluated by the contact angle between the particle and liquid ($\theta$ shown in Fig. 1-7). In particular, effect of the contact angle on a critical velocity for the particle adhesion was investigated. To elucidate the role of the particle wettability, the liquid bridge force acting on a particle and deformation of the liquid bridge were analyzed.

Chapter IV studies the effect of binder droplet size ($d_{\text{drop}}$ shown in Fig. 1-7) on the particle-particle adhesion by the dynamic liquid bridge. It has been recognized that the nuclei size of granules are determined by size of the binder droplets. Therefore, understanding of effect of the droplet size is an important issue for precise control of the wet granulation. In this simulation, droplet size was varied under constant liquid volume. In particular, effect of the droplet size on a critical velocity for the particle adhesion was investigated and the role of the binder droplet size in the particle adhesion by the dynamic liquid bridge was discussed.

Chapter V investigates the effect of collision angle ($\alpha$ shown in Fig. 1-7) on the particle-particle adhesion by the dynamic liquid bridge. In an actual wet granulation process, the particle-particle collision mediated by a droplet can occur at various collision angles, because the particles randomly collide with each other. Therefore, understanding of effect of the collision angle against the droplet on a particle surface is important. In particular, difference between the direct numerical simulation and a conventional static liquid bridge force model at an oblique particle collision was focused.

Chapter VI summarizes the conclusions of this thesis.
Fig. 1-7. Schematic of parameters analyzed in this thesis.

\( v \) : collision velocity (Chapter II)
\( \theta \) : contact angle (Chapter III)
\( d_{\text{drop}} \) : droplet size (Chapter IV)
\( \alpha \) : collision angle (Chapter V)
References


Chapter II

Numerical modeling of particle-particle adhesion
by dynamic liquid bridge
1. Introduction

In many powder handling processes, particles containing a small amount of liquid are frequently processed, and liquid bridges formed between particles can induce a particle-particle adhesion, leading to determine quality of the final products. Therefore, understanding of the particle-particle adhesion by a liquid bridge has been an important issue. However, a theoretical and experimental approach cannot directly analyze the particle-particle adhesion due to their complexities, because the liquid bridge is not static but dynamic. In such a case, a direct numerical simulation can be a powerful tool.

To simulate a microscopic particle-particle adhesion by a dynamic liquid bridge, a solid-liquid-gas three-phase flow is needed to be solved. Especially, deformation of gas-liquid interface should be exactly tracked. In this chapter, a numerical simulation model of particle-particle adhesion by a dynamic liquid bridge was developed. A computational fluid dynamics approach combined with a constrained interpolation profile method was applied to simulate gas-liquid two-phase flow. A Lagrangian approach considering the dynamic liquid bridge was used to calculate motion of a solid particle. First of all, verifications and validations were conducted. The particle-particle adhesion by the dynamic liquid bridge was then simulated. In particular, the effect of collision velocity on a wet restitution coefficient between particles was investigated.
2. Numerical simulation model

2.1. Governing equations of gas-liquid two-phase flow

The governing equations of gas-liquid two-phase flow were given by equations of continuity and motion, and advection equation for color function as follows:

**Equation of continuity**
\[ \nabla \cdot \mathbf{u} = 0 \]  
(2-1)

**Equation of motion**
\[ \frac{\partial \mathbf{u}}{\partial t} + (\mathbf{u} \cdot \nabla) \mathbf{u} = -\frac{1}{\rho_f} \nabla p + \frac{1}{\rho_f} \nabla \cdot \mathbf{u} [\nabla \mathbf{u} + (\nabla \mathbf{u})^T] + f_{sf} + f_p + \mathbf{g} \]  
(2-2)

**Advection equation for color function**
\[ \frac{\partial \phi}{\partial t} + (\mathbf{u} \cdot \nabla) \phi = 0 \]  
(2-3)

where \( \mathbf{u}, t, \rho_f, p, \mu_f, f_{sf}, f_p \) and \( \mathbf{g} \) are the fluid velocity, time, fluid density, pressure, fluid viscosity, surface tension force, interaction force from particle to fluid, and gravity, respectively. \( \phi \) is a color function, which is a parameter to distinguish phase of the fluid; i.e., \( \phi = 1 \) means the liquid phase, while \( \phi = 0 \) means the gas phase. The fluid density and viscosity in a fluid cell were given as follows:

\[ \rho_f = \phi \rho_l + (1 - \phi) \rho_g \]  
(2-4)

\[ \mu_f = \phi \mu_l + (1 - \phi) \mu_g \]  
(2-5)

where \( \rho_l, \rho_g, \mu_l \) and \( \mu_g \) are the liquid density, gas density, liquid viscosity, and gas viscosity, respectively. In this simulation model, the fluid flow was obtained by solving these equations in the three-dimensional region which is divided into a fixed Cartesian grid.
2.1.1 Calculation of deformation of gas-liquid interface

Unsteady motion of a gas-liquid interface was tracked by solving Eq. (2-3). To numerically solve Eq. (2-3), a CIP method [1-3] was adapted. A CIP method will be explained as follows. A one-dimensional advection equation was used as an example. Fig. 2-1 shows an advection of arbitrary quantity $f$. The vertical axis means the quantity $f$, while the horizontal axis means a spatial coordinate $x$. When $f$ is advected at a velocity $u$, advection equation is given as follows:

$$\frac{\partial f}{\partial t} + u \frac{\partial f}{\partial x} = 0$$

(2-6)

As shown in Fig. 2-1, a profile of quantity $f$ after a time interval $\Delta t$ (dashed line) is obtained by advecting a profile of quantity $f$ at time $t$ (solid line) by $u\Delta t$. Therefore, following equation is given:

$$f(x,t+\Delta t) = f(x-u\Delta t,t)$$

(2-7)

To accurately solve Eq. (2-6), an interpolation method for the quantity $f$ between each spatially discretized point is important. In the CIP method, the profile between neighboring points (e.g. $[i_{up}, i]$ shown in Fig. 2-1) is interpolated by a cubic function as follows:

$$f(x) = c_3(x-x_i)^3 + c_2(x-x_i)^2 + c_1(x-x_i) + c_0$$

(2-8)

$$\frac{\partial f(x)}{\partial x} = g(x) = 3c_3(x-x_i)^2 + 2c_2(x-x_i) + c_1$$

(2-9)

where $g(x)$ is the derivative value of $f(x)$. $c_3$, $c_2$, $c_1$ and $c_0$ are the coefficients given as follows:
Fig. 2-1. Advection of arbitrary quantity $f$. 
\[
c_3 = \frac{g'_i + g'_{i_{up}}}{D^2} + \frac{2(f'_i - f'_{i_{up}})}{D^3}
\]

(2-10)

\[
c_2 = \frac{3(f'_i - f'_{i_{up}})}{D^2} - \frac{2g'_i + g'_{i_{up}}}{D}
\]

(2-11)

\[
c_1 = g'_i
\]

(2-12)

\[
c_0 = f'_i
\]

(2-13)

where \( D = -\Delta x \) when \( u \) is positive, while \( D = \Delta x \) when \( u \) is negative. Superscript \( t \) means time, and subscripts \( i \) and \( i_{up} \) mean position of points. At \( t + \Delta t \), \( f \) and \( g \) are given as following equations:

\[
f'_{i}^{t+\Delta t} = f(x_i - u \Delta t, t) = c_3(-u \Delta t)^3 + c_2(-u \Delta t)^2 + c_1(-u \Delta t) + c_0
\]

(2-14)

\[
g'_{i}^{t+\Delta t} = g(x_i - u \Delta t, t) = 3c_3(-u \Delta t)^2 + 2c_2(-u \Delta t) + c_1
\]

(2-15)

In the CIP method, not only quantity \( f \) at each spatial point but differential value \( g \) (i.e., gradient of \( f \)) are used as constraint conditions to interpolate quantity \( f \) between each point. In a common interpolation scheme such as first order upwind difference scheme, exact solution is discretized and is interpolated using a linear interpolation. As a result, there is a large difference between exact solution and numerical solution after advection (Fig. 2-2 (a)). On the other hand, in the CIP method, the waveform after advection is similar with exact solution, because the differential value \( g \) is used as a constraint condition (Fig. 2-2 (b)). Therefore, the CIP method is useful for solving an advection equation for color function with high accuracy. A solution of the three-dimensional advection equation for the color function (Eq. (2-3)) is shown in Appendix A.
(a) Common scheme (e.g. first order upwind difference scheme)

(b) CIP method

Fig. 2-2. Comparison between common scheme and CIP method.
2.1.2. Calculation of surface tension force

The surface tension force \( f_{sf} \) in Eq. (2-2) was calculated by a continuous surface force (CSF) model [4] as follows:

\[
f_{sf} = \frac{\sigma \kappa \nabla \phi}{\rho_i}
\]

(2-16)

where \( \sigma \) is the surface tension coefficient and \( \kappa \) is the local curvature of a gas-liquid interface. The curvature was calculated by the following equation:

\[
\kappa = - (\nabla \cdot \mathbf{n}_{lg}) = - \left( \nabla \cdot \frac{\nabla \phi_{sm}}{\left| \nabla \phi_{sm} \right|} \right)
\]

(2-17)

where \( \mathbf{n}_{lg} \) is the unit normal vector at the interfaces, and \( \phi_{sm} \) is the smoothed color function. \( \phi_{sm} \) is given as following equations [5]:

\[
\phi_{sm,i,j,k} = \frac{1}{2} \phi_{i,j,k} + \frac{1}{2(1 + 6 C_1 + 12 C_2 + 8 C_3)} \left\{ \phi_{i,j,k} + C_1 (\phi_{i-1,j,k} + \phi_{i+1,j,k} + \phi_{i,j-1,k} + \phi_{i,j+1,k}) \\
+ \phi_{i,j,k+1} + \phi_{i,j,k-1} + \phi_{i+1,j,k+1} + \phi_{i+1,j,k-1} + \phi_{i-1,j,k+1} + \phi_{i-1,j,k-1} + \phi_{i,j+1,k+1} + \phi_{i,j+1,k-1} + \phi_{i,j-1,k+1} + \phi_{i,j-1,k-1} + \phi_{i-1,j+1,k+1} + \phi_{i-1,j+1,k-1} + \phi_{i+1,j-1,k+1} + \phi_{i+1,j-1,k-1} \right\}
\]

(2-18)

\[
C_1 = \frac{1}{6 + 12 / \sqrt{2} + 8 / \sqrt{3}}
\]

(2-19)

\[
C_2 = \frac{C_1}{\sqrt{2}}
\]

(2-20)
Using the CSF model, the surface tension force at the solid-liquid-gas interface on a solid particle (i.e., contact line) was also calculated. When a droplet is in equilibrium with the solid surface at a contact angle $\theta$ (Fig. 2-3), the unit normal vector ($n_{lg}$) at the solid-liquid-gas interface is defined as follows:

$$n_{lg} = n_s \cos \theta + t_s \sin \theta$$

(2-22)

where $n_s$ and $t_s$ are the unit normal and tangential vectors at the solid surface, defined as follows:

$$n_s = \frac{\nabla \phi_s}{|\nabla \phi_s|}$$

(2-23)

$$t_s = \frac{\nabla \phi - (n_s \cdot \nabla \phi)n_s}{|\nabla \phi - (n_s \cdot \nabla \phi)n_s|}$$

(2-24)

where $\phi_s$ is a volume fraction of the solid in a fluid cell. In this simulation model, the wetting on a solid surface was taken into account through the contact angle ($\theta$), which was preliminary set as a calculation condition, and $n_{lg}$ defined by Eq. (2-22) was used to calculate the curvature ($\kappa$) at the solid-liquid-gas interface.
Fig. 2-3. Schematic of unit vectors at three-phase interface.
2.1.3. Calculation of interaction force from particle to fluid

The interaction force from particle to fluid \((f_p)\) in Eq. (2-2) was calculated by an immersed boundary (IB) method proposed by Kajishima et al. [6]. In the IB method, the interaction force from particle to fluid \((f_p)\) was given as follows:

\[
f_p = \frac{\phi_s (u_p - u^C)}{\Delta t} \tag{2-25}
\]

\[
u_p = v_p + r \times \omega_p \tag{2-26}
\]

where \(u^C\), \(v_p\), \(r\) and \(\omega_p\) are fluid velocity, translational particle velocity, relative position from the center of gravity of the particle to the fluid cell, and rotational particle velocity.
2.2. Governing equations of particle motion

The governing equations of particle motion (Newton’s second law) were given as follows:

*Translational motion*

\[
m_p \frac{dv_p}{dt} = F_d + F_{sf} + F_{cp} + m_p g
\]  
(2-27)

*Rotational motion*

\[
I_p \frac{d\omega_p}{dt} = M_d + M_{sf}
\]  
(2-28)

where \( m_p, F_{cp}, F_d, F_{sf}, I_p, M_d \) and \( M_{sf} \) are the particle mass, capillary pressure force, viscous drag force, surface tension force acting on the particle, particle inertia, torque associated with the viscous drag force, and torque associated with the surface tension force, respectively.

2.2.1. Calculations of viscous drag force and torque

The viscous drag force \( (F_d) \) and torque associated with the viscous drag force \( (M_d) \) were calculated by integrating the counteracting force from particle to fluid over the particle volume as follows [6, 7]:

\[
F_d = -\int_V \rho_f f_p dV
\]  
(2-29)

\[
M_d = -\int_V \rho_f r \times f_p dV
\]  
(2-30)

where \( V_p \) is the particle volume.
2.2.2. Calculation of surface tension force and torque acting on particle

The surface tension force ($F_{sf}$) and torque associated with the surface tension force ($M_{sf}$) were calculated by a continuous capillary force (CCF) model [8]. In the CCF model, $F_{sf}$ and $M_{sf}$ were given as follows:

$$F_{sf} = \int_V \sigma t_c (\nabla \phi \cdot t_s)(\nabla \phi \cdot n_s) dV$$

$$(2-31)$$

$$M_{sf} = \int_V r \times \{ \sigma t_c (\nabla \phi \cdot t_s)(\nabla \phi \cdot n_s) \} dV$$

$$(2-32)$$

where $t_c$ is the unit tangential vector along a gas-liquid interface at a contact line (shown in Fig. 2-4), calculated as follows:

$$t_c = \frac{n_s - (n_s \cdot n_c)n_c}{|n_s - (n_s \cdot n_c)n_c|}$$

$$(2-33)$$

where $n_c$ is the unit normal vector along a gas-liquid interface at a contact line (shown in Fig. 2-4), calculated as follows:

$$n_c = \frac{\nabla \phi}{|\nabla \phi|}$$

$$(2-34)$$
Fig. 2-4. Schematic of unit vectors used in CCF model.
2.2.3. Calculation of capillary pressure force

The capillary pressure force ($F_{cp}$) was calculated from the area enclosed by the solid-liquid-gas interface and the pressure difference between liquid phase and gas phase as follows [9]:

$$F_{cp} = t_c \pi (R_p \sin \alpha_{hf})^2 \Delta p$$

(2-35)

where $R_p$, $\alpha_{hf}$ and $\Delta p$ are the particle radius, half-filling angle, and pressure difference between liquid phase and gas phase, respectively (Fig. 2-5). $R_p \sin \alpha_{hf}$, which is radius of the solid-liquid-gas interface, was calculated from perimeter of the solid-liquid-gas interface:

$$R_p \sin \alpha_{hf} = \frac{1}{2\pi} \int_{\gamma_s} (\nabla \phi \cdot \mathbf{t_s})(\nabla \phi_s \cdot \mathbf{n_s}) dV$$

(2-36)
Fig. 2-5. Schematic of liquid bridge between particles.
2.2.4. Calculation of particle-particle collision

When the two particles collided, translational and rotational rebound velocities of the two particles just after the particle-particle collision ($v_{1,rc}$, $v_{2,rc}$, $\omega_{1,rc}$, and $\omega_{2,rc}$) were calculated using a hard-sphere model [10] as follows:

**Translational rebound velocity**

$$v_{1,rc} = v_1 - \left\{ (1 + e)(n \cdot G)n + \frac{2}{7} |G_1||t| \right\} \frac{m_{p2}}{m_{p1} + m_{p2}}$$  \hspace{1cm} (2-37)

$$v_{2,rc} = v_2 + \left\{ (1 + e)(n \cdot G)n + \frac{2}{7} |G_1||t| \right\} \frac{m_{p1}}{m_{p1} + m_{p2}}$$  \hspace{1cm} (2-38)

**Rotational rebound velocity**

$$\omega_{1,rc} = \omega_1 - \frac{5}{7 R_{p1}} |G_1| (n \times t) \frac{m_{p2}}{m_{p1} + m_{p2}}$$  \hspace{1cm} (2-39)

$$\omega_{2,rc} = \omega_2 - \frac{5}{7 R_{p2}} |G_1| (n \times t) \frac{m_{p1}}{m_{p1} + m_{p2}}$$  \hspace{1cm} (2-40)

where $v_1$ and $v_2$ are the translational velocities of the particle-1 and particle-2 just before collision, $\omega_1$ and $\omega_2$ are the rotational velocities of the particle-1 and particle-2 just before collision. $e$, $G$, $G_0$, $n$ and $t$ are the restitution coefficient between solid particles, relative translational velocity between two particles, tangential relative velocity, unit normal vector from the particle-1 to particle-2, and unit vector of the tangential relative velocity, respectively. $G$, $G_0$, and $t$ are given as follows:

$$G = v_1 - v_2$$  \hspace{1cm} (2-41)

$$G_t = G - (G \cdot n)n + R_{p1} \omega_1 \times n + R_{p2} \omega_2 \times n$$  \hspace{1cm} (2-42)

$$t = \frac{G_t}{|G_t|}$$  \hspace{1cm} (2-43)
2.3. Computational algorithm

In a direct numerical simulation, the all spatial domain is dived into cells which are smaller than liquid bridge and particle size. The governing equations were solved in a three-dimensional fixed Cartesian coordinate. Fig. 2-6 shows the flow diagram of calculation. The calculation was performed by a following procedure:

1) Eqs. (2-2) and (2-3) were solved by a CIP combined unified procedure (CCUP) method [11], in which the advection and non-advection phases were separately solved. In Fig. 2-6, the fluid velocity and color function before solving the advection are expressed as \( u^n \) and \( \phi^n \), respectively. The advection terms of Eqs. (2-2) and (2-3) (Eqs. (2-44) and (4-45)) were solved by the CIP method.

\[
\frac{\partial u}{\partial t} + (u \cdot \nabla) u = 0 \quad (2-44)
\]

\[
\frac{\partial \phi}{\partial t} + (u \cdot \nabla) \phi = 0 \quad (2-45)
\]

After solving the advection, the fluid velocity \( u^A \) and color function \( \phi^{n+1} \) were obtained. \( u^A \) was used in the next step as an input valuable.

2) The non-advection terms of Eq. (2-2), excepting for the pressure gradient and interaction force from particle to fluid \( f_p \) (Eq. (2-46)), were discretized by a central difference scheme.

\[
\frac{\partial u}{\partial t} = \frac{1}{\rho_f} \nabla \cdot \mu_f [\nabla u + (\nabla u)^T] + f_{sf} + g \quad (2-46)
\]

After solving Eq. (2-46), fluid velocity \( u^B \) was obtained. \( u^B \) was used in the next step as an input valuable.

3) To consider the pressure gradient, the fluid pressure at next time step \( p^{n+1} \) was needed. To obtain \( p^{n+1} \), the pressure Poisson equation (Eq. (2-47)) was solved by a biconjugate gradient stabilized (Bi-CGSTAB) method [12].

\[
\frac{\nabla \cdot u}{\Delta t} = \nabla \left( \frac{1}{\rho_f} \nabla p \right) \quad (2-47)
\]
The non-advection term of Eq. (2-2) including pressure gradient (Eq. (2-48)) was solved using the fluid pressure $p^{n+1}$.

$$\frac{\partial u}{\partial t} = -\frac{1}{\rho_f} \nabla p \quad (2-48)$$

After solving Eq. (2-48), the fluid velocity $u^C$ was obtained. $u^C$ was used in the next step as an input valuable.

4) The non-advection term of Eq. (2-2) including the interaction force from particle to fluid $f_p$ (Eq. (2-49)) was solved.

$$\frac{\partial u}{\partial t} = f_p \quad (2-49)$$

After solving Eq. (2-49), the fluid velocity $u^{n+1}$ was obtained.

5) After solving the fluid flow, particle motion was calculated considering effect of the fluid flow. External forces of Eq. (2-27) and torques of Eq. (2-28) were calculated. Translational velocity, position, and rotational velocity were obtained by solving Eqs. (2-27) and (2-28). The time-derivative term of Eq. (2-27) was solved by a symplectic Euler scheme.

6) A subdivision volume counting (SVC) method [13] was used to calculate the volume fraction of solid ($\phi_s$). In the SVC method, each interfacial cell for a solid particle is subdivided into small elements as shown in Fig. 2-7. If the distance from center of the particle to center of the element ($L_{sub}$) is shorter than particle radius ($R_p$), the volume of the subdivided element counted as a part of the particle. The volume fraction of solid ($\phi_s$) is then calculated as follows:

$$\phi_s = \frac{\text{Number of subdivided cells which satisfy } L_{sub} < R_p}{\text{Number of all subdivided elements in a cell}} \quad (2-50)$$

In this study, staggered layout was used for definition of the physical and vector values (shown in Appendix B). The discretized equations are also shown in Appendix C.
Start
Initialize

$u^n, \phi^n, \phi_s^n$

Solve advection terms
(Eqs. (2-44) and (2-45))

$u^A, \phi^{n+1}$

Compute Eq.(2-46)

$u^B$

Compute Eq.(2-48)

$u^C$

Compute Eq.(2-49)

$u^{n+1}$

Solve equations of particle motion
(Eqs. (2-27) and (2-28))

Compute SVC method

$\phi_s^{n+1}$

Save data

End

$n \Rightarrow n+1$

Fig. 2-6. Flow diagram of calculation.
Fig. 2-7. Schematic of subdivision volume counting (SVC) method.
3. Verification and validation of simulation results

To verify the simulation model, three verifications and validations were conducted. Initially, in order to verify the accuracy of calculated surface tension force acting on a droplet on a spherical particle surface, wetting behavior of a droplet on a spherical particle surface was simulated at various contact angles. Subsequently, in order to verify the accuracy of calculated static liquid bridge force between particles, a static liquid bridge force between two fixed particles was simulated and compared to that estimated by available analytical models. Finally, in order to validate the accuracy of calculated unsteady motions of liquid bridge and particle, rupture behavior of a liquid bridge with free falling of a particle was simulated and compared to experimental results.

3.1. Wetting behavior of a droplet on a spherical particle surface

Table 2-1 shows calculation conditions. The simulation was conducted at various contact angles, i.e., set value of the contact angle \( \theta \), shown in Eq. (2-22), was changed from 10 to 120 deg. As the initial condition, a spherical droplet was placed just above a surface of a fixed spherical particle, i.e., surface distance between the particle and droplet was set as zero (Fig. 2-8 \( t = 0 \) \( \mu s \)). Initial velocity and pressure of the droplet were set as zero. Unsteady motion of the droplet under gravity was then simulated. In this calculation condition, Bond number, which is the ratio of gravity to surface tension, was \( 1.94 \times 10^{-4} \ll 1 \), confirming that the droplet shape in equilibrium state is not affected by gravity [14].

Fig. 2-8 indicates simulated wetting behaviors of droplets. At \( \theta = 30 \) deg, the droplet largely spread on the particle surface due to relatively hydrophilic surface, while at \( \theta = 120 \) deg, the droplet slightly spread due to relatively hydrophobic surface. Fig. 2-9 shows comparison between set value (\( \theta \)) and calculated contact angle (\( \theta_{\text{cal}} \)). The calculated contact angle was defined as a contact angle obtained from simulated droplet shape after reaching steady state. As shown in Fig. 2-9, calculated contact angle showed good agreement with set value at higher than \( \theta = 30 \) deg, while at \( \theta = 10 \) deg calculated contact angle did not show good agreement with the set value. At \( \theta = 10 \) deg, thickness of the droplet on the particle surface was small. Thus, to calculate wetting
behavior with high accuracy at the smaller contact angle, it was considered that higher spatial resolution, i.e., smaller size of fluid cell, was required. From the result, in this simulation, the surface tension force acting on liquid on a spherical particle surface was accurately calculated at higher than $\theta = 30$ deg. This range covers solid-liquid contact angles normally used in wet granulation [15, 16].
Table 2-1 Calculation conditions for wetting behavior of droplet on spherical particle surface

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Particle diameter</td>
<td>50.0 µm</td>
</tr>
<tr>
<td>Static contact angle</td>
<td>10.0 – 120.0 deg</td>
</tr>
<tr>
<td>Liquid density</td>
<td>1023.0 kg/m³</td>
</tr>
<tr>
<td>Liquid viscosity</td>
<td>0.014 Pa·s</td>
</tr>
<tr>
<td>Surface tension coefficient</td>
<td>0.0322 N/m</td>
</tr>
<tr>
<td>Droplet diameter</td>
<td>15.6 µm</td>
</tr>
<tr>
<td>Gas density</td>
<td>1.184 kg/m³</td>
</tr>
<tr>
<td>Gas viscosity</td>
<td>1.82×10⁻⁵ Pa·s</td>
</tr>
<tr>
<td>Time step</td>
<td>5.0×10⁻⁹ s</td>
</tr>
<tr>
<td>Fluid cell size</td>
<td>1.0 µm</td>
</tr>
</tbody>
</table>
Fig. 2-8. Wetting behavior of droplets on spherical particle surface at different contact angles.
Fig. 2-9. Comparison between $\theta$ and $\theta_{\text{cal}}$. 
3.2. Static liquid bridge force between particles

For comparison between the simulation result of the static liquid bridge force and analytical solutions, two analytical models proposed by Rabinovich et al. [17] and Mikami et al. [18] were selected. The analytical model proposed by Rabinovich et al. [17] was given as follows:

\[
F_{LB} = \frac{2\pi R_p \sigma \cos \theta}{1 + h/2d} + 2\pi R_p \sigma \sin \alpha_{hf} \sin(\theta + \alpha_{hf})
\]  
(2-51)

\[
d = \frac{h}{2} \left( 1 + \sqrt{1 + \frac{2V_{LB}}{\pi R_p h^2}} \right)
\]  
(2-52)

\[
\cos \alpha_{hf} = 1 - \frac{R_p}{d}
\]  
(2-53)

where \( F_{LB} \), \( h \), \( d \) and \( V_{LB} \) are the static liquid bridge force, surface distance between particles, distance shown in Fig. 2-5, and liquid bridge volume, respectively. The analytical model proposed by Mikami et al. [18] was given as follows:

\[
\hat{F}_{LB} = \exp(A \hat{h} + B) + C
\]  
(2-54)

\[
A = -1.1V_{LB}^{-0.53}
\]  
(2-55)

\[
B = -(0.34 \ln \hat{V}_{LB} - 0.96) \hat{\theta}^2 - 0.019 \ln \hat{V}_{LB} + 0.48
\]  
(2-56)

\[
C = 0.0042 \ln \hat{V}_{LB} + 0.078
\]  
(2-57)

where \( \hat{F}_{LB} \), \( \hat{V}_{LB} \) and \( \hat{h} \) are the dimensionless liquid bridge force (= \( F_{LB}/\pi R_p \sigma \)), dimensionless liquid bridge volume (= \( V_{LB}/R_p^3 \)), and dimensionless distance between
particles ($= h/R_p$), respectively. As shown in Fig. 2-10, through comparisons with experimental results, which were conducted using sapphire spheres as a particle and silicone oil as a liquid [19], it was preliminarily confirmed that these two analytical models show high accuracy for predicting the static liquid bridge force.

Table 2-2 shows calculation conditions. The physical properties of dibutyl phthalate were set as calculation conditions of the liquid. In this simulation, the particle diameter, liquid bridge volume and contact angle were constant, while the distance between particles was changed. The static liquid bridge force was defined as the sum of the surface tension force ($F_{sf}$) and capillary pressure force ($F_{cp}$) in Eq. (2-27).

Fig. 2-11 indicates dimensionless liquid bridge forces calculated from the present simulation and estimation results by the analytical models as a function of dimensionless separation distance between particles. The dimensionless surface tension force ($= F_{sf}/\pi R_p \sigma$) and dimensionless capillary pressure force ($= F_{cp}/\pi R_p \sigma$) are also shown. The simulated liquid bridge force showed good agreement with the estimation results, confirming that the static liquid bridge force between particles was accurately calculated by the present simulation. The simulation result indicates that at the longer distance between particles, the static liquid bridge force was dominated by the surface tension force, while the capillary pressure force had little contribution to the static liquid bridge force. However, at the smaller distance between particles, contribution of the capillary pressure force became larger and cannot be ignored. This result means that the capillary pressure force was required for accurate calculation of the static liquid bridge force.
Theoretical and experimental values of static liquid bridge force.

Table 2-2 Calculation conditions for static liquid bridge force

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Particle diameter</td>
<td>3.97</td>
<td>mm</td>
</tr>
<tr>
<td>Static contact angle</td>
<td>30.0</td>
<td>deg</td>
</tr>
<tr>
<td>Liquid density</td>
<td>1023.0</td>
<td>kg/m³</td>
</tr>
<tr>
<td>Liquid viscosity</td>
<td>0.014</td>
<td>Pa·s</td>
</tr>
<tr>
<td>Surface tension coefficient</td>
<td>0.0322</td>
<td>N/m</td>
</tr>
<tr>
<td>Dimensionless liquid volume</td>
<td>0.02</td>
<td>–</td>
</tr>
<tr>
<td>Gas density</td>
<td>1.184</td>
<td>kg/m³</td>
</tr>
<tr>
<td>Gas viscosity</td>
<td>1.82×10⁻⁵</td>
<td>Pa·s</td>
</tr>
<tr>
<td>Fluid cell size</td>
<td>0.0428</td>
<td>mm</td>
</tr>
</tbody>
</table>
Fig. 2-11. Comparison between simulated and estimated values of static liquid bridge force.
3.3. Rupture behavior of liquid bridge with free falling of a particle

As an initial condition, two particles were aligned vertically and a liquid bridge was formed between two particles (Fig. 2-12). The lower particle was fallen by gravity, while the upper particle was fixed. Table 2-3 shows calculation conditions. These calculation conditions were identical with the experimental conditions. Fig. 2-13 illustrates schematic diagram of the experimental set-up. A stainless steel ball made of SUS440C (G-28, Azuma Zikuuke Co., Ltd.) was used as a particle. The upper particle was fixed with an electromagnetic (TMN-105SA, Gigateco Co., Ltd.) through an adhesive tape. As an initial state, the lower particle was adhered to the upper particle by the electromagnetic force, and the liquid bridge was formed between the particles by injecting a liquid. A silicon oil (KF-96L-1cs, Shinetsu Chemical Co., Ltd.) was used as the liquid. The lower particle was then fallen by gravity by turning off a DC voltage for the electromagnetic. The rupture behavior of the liquid bridge and free falling behavior of the lower particle were observed by means of a high speed video camera (FASTCAM-MAX 120K, Photron). The frame rate was set as 3000 fps, while the shutter speed was set as 1/3000 s. As a light source, a metal halide lamp (HVC-SL250, Kyowa Co. Ltd.) was used. The experiment was repeated 10 times, and the reproducibility was confirmed.

To confirm the accuracy of the experiment, free falling behavior without liquid bridge was observed and compared to a theoretical result. Theoretical value was calculated by solving an equation of motion which includes gravitational force, buoyancy force, and fluid drag force as external forces. The experimental result of the temporal change in surface distance between particles showed good agreement with the theoretical value, confirming that the accuracy of the experiment.

Fig. 2-14 indicates snapshots of the rupture behavior of the liquid bridge with the free falling particle. In the experiment, as the lower particle was fallen by gravity, the liquid bridge was gradually elongated (Fig. 2-14 \( t = 10 \) ms and \( t = 20 \) ms), and eventually ruptured (Fig. 2-14 \( t = 26 \) ms). The simulation result exhibited quite similar behavior with experimental one. Fig. 2-15 shows the temporal change in the distance between particles. The rupture distance of the liquid bridge was also shown by the arrows. The
simulation result showed good agreement with the experimental result. Therefore, validity of the simulation result was confirmed.
Fig. 2-12. Initial conditions for rupture behavior of liquid bridge with free falling particle.

Table 2-3 Calculation conditions for rupture behavior of liquid bridge with free falling particle

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Particle diameter</td>
<td>6.35</td>
<td>mm</td>
</tr>
<tr>
<td>Particle density</td>
<td>7800.0</td>
<td>kg/m³</td>
</tr>
<tr>
<td>Static contact angle</td>
<td>3.0</td>
<td>deg</td>
</tr>
<tr>
<td>Liquid density</td>
<td>818.0</td>
<td>kg/m³</td>
</tr>
<tr>
<td>Liquid viscosity</td>
<td>8.18×10⁻⁴</td>
<td>Pa·s</td>
</tr>
<tr>
<td>Surface tension coefficient</td>
<td>0.0169</td>
<td>N/m</td>
</tr>
<tr>
<td>Liquid volume</td>
<td>2.6</td>
<td>mm³</td>
</tr>
<tr>
<td>Gas density</td>
<td>1.184</td>
<td>kg/m³</td>
</tr>
<tr>
<td>Gas viscosity</td>
<td>1.82×10⁻⁵</td>
<td>Pa·s</td>
</tr>
<tr>
<td>Time step</td>
<td>1.0×10⁻⁶</td>
<td>s</td>
</tr>
<tr>
<td>Fluid cell size</td>
<td>0.109</td>
<td>mm</td>
</tr>
</tbody>
</table>
Fig. 2-13. Schematic diagram of experimental set-up for observation of rupture of liquid bridge.
Fig. 2-14. Rupture behavior of liquid bridge between particles.
Fig. 2-15. Temporal change in surface distance between particles.
4. Adhesion of two colliding particles through droplet

4.1. Simulation set-up

Adhesion of two colliding particles through a droplet on a particle surface was simulated. This corresponds to a micro level agglomeration phenomenon in a fluidized bed spray granulation. Fig. 2-16 shows the initial condition. Two particles, on which a droplet was adhered, collided with each other in the normal direction. Therefore, rotational motion of particles was not considered in this simulation. The simulations were conducted at various relative collision velocities $v (v = |v_{1,\text{init}} - v_{2,\text{init}}|)$, while magnitude of the velocity of each particle was constant ($|v_{1,\text{init}}| = |v_{2,\text{init}}|$). Table 2-4 lists calculation conditions. The physical properties of water were used for the calculation conditions of the liquid. A model particle with spherical shape was used. The size and density of a lactose particle, which is a common pharmaceutical excipient, was used for the simulation. The droplet volume was set based on a mist diameter sprayed in a fluidized bed granulation [20]. The contact angle between water and lactose was determined from literature data [21]. Under these conditions, Bond number was $8.47 \times 10^{-5} \ll 1$, meaning that the influence of gravity can be ignored [14]. Therefore, in this simulation, the gravity was not taken into account.
Fig. 2-16. Initial condition for calculation of two colliding particles through a droplet on a particle surface.

<table>
<thead>
<tr>
<th>Calculation conditions for adhesion of two colliding particles</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Particle diameter</strong></td>
</tr>
<tr>
<td><strong>Particle density</strong></td>
</tr>
<tr>
<td><strong>Static contact angle</strong></td>
</tr>
<tr>
<td><strong>Restitution coefficient between solid particles</strong></td>
</tr>
<tr>
<td><strong>Liquid density</strong></td>
</tr>
<tr>
<td><strong>Liquid viscosity</strong></td>
</tr>
<tr>
<td><strong>Surface tension coefficient</strong></td>
</tr>
<tr>
<td><strong>Droplet volume</strong></td>
</tr>
<tr>
<td><strong>Gas density</strong></td>
</tr>
<tr>
<td><strong>Gas viscosity</strong></td>
</tr>
<tr>
<td><strong>Time step</strong></td>
</tr>
<tr>
<td><strong>Fluid cell size</strong></td>
</tr>
</tbody>
</table>
4.2. Particle and liquid bridge behaviors

Fig. 2-17 shows simulation results of colliding particles and liquid bridge behaviors at different relative collision velocities. With an approach of two particles, a liquid bridge was initially formed between particles (Fig. 2-17 (a) $t = 3 \mu s$, (b) $t = 2 \mu s$). The particles then collided with each other (Fig. 2-17 (a) $t = 10 \mu s$, (b) $t = 3 \mu s$). After the particle-particle collision, the particles rebounded and moved away from each other. At the lower collision velocity, the liquid bridge was not ruptured (Fig. 2-17 (a) $t = 23 \mu s$), and the particles were eventually adhered (Fig. 2-17 (a) $t = 80 \mu s$). On the other hand, at the higher relative collision velocity, the liquid bridge was largely elongated (Fig. 2-17 (b) $t = 10 \mu s$). Eventually, the liquid bridge was ruptured and the particles were not adhered but separated (Fig. 2-17 (b) $t = 15 \mu s$).

Fig. 2-18 indicates temporal change in velocity of the particle-1 (left particle) at different relative collision velocities. The positive and negative velocities mean the rightward and leftward velocities, respectively. At the lower relative collision velocity (Fig. 2-18 (a)), the particle velocity increased during the approach stage due to the attractive surface tension force. At the moment of the particle-particle collision, the particle velocity changed from positive to negative, indicating that the particles rebounded. During the subsequent separation stage magnitude of the particle velocity gradually decreased and reached to zero. At this moment, the particle velocity changed from negative to positive, i.e., the particles re-approached. The particles then collided with each other again. Magnitude of the particle velocity gradually decreased with repeating this approach-separation stage, and the particles were finally adhered. On the other hand, at the higher relative collision velocity (Fig. 2-18 (b)), the liquid bridge ruptured before the particle velocity reached to zero during the separation stage, resulting in the separation of the two particles.
Fig. 2-17. Simulation results of particles and liquid bridge behaviors at different relative collision velocities.
Fig. 2-18. Temporal change in velocity of particle-1 at different relative collision velocities. (a) $v = 1.0 \text{ m/s}$ and (b) $v = 5.0 \text{ m/s}$. 
4.3. Liquid bridge force acting on particle

Fig. 2-19 shows all components of the liquid bridge forces (capillary pressure force $F_{cp}$, drag force $F_d$ and surface tension force $F_{st}$) acted on the particle-1 as a function of separation distance during the approach and separation of the particle at different relative collision velocities. The positive and negative forces mean the repulsive and attractive forces for two particles, respectively. At the lower relative collision velocity (Fig. 2-19 (a)), the capillary pressure force acted on the particle as a repulsive force in the approach stage. Magnitude of the capillary pressure force increased with the approach. The drag force initially acted as an attractive force in the approach stage at the larger separation distance. However, the drag force increased with the approach and acted as a repulsive force before the particle-particle collision. The surface tension force acted as an attractive force and its magnitude slightly increased with the approach. Just after the particle-particle collision, the capillary pressure force and the drag force rapidly shifted to negative, i.e., these two forces acted as attractive forces. With separation of the two particles, magnitude of the capillary pressure force and the drag force steeply decreased, followed by a constant. The capillary pressure force showed almost zero, while the drag force acted as an attractive force. Magnitude of the surface tension force slightly decreased with separation of the two particles. At the higher relative collision velocity (Fig. 2-19 (b)), change in each liquid bridge force showed similar tendency with that at the lower relative collision velocity. However, magnitude of the drag force just before the particle-particle collision showed much larger than that at the lower relative collision velocity. Magnitude of the capillary pressure force and the drag force just after the particle-particle collision also showed much larger than that at the lower relative collision velocity. The separation distance where the capillary pressure force and the drag force reached to almost constant in the separation stage became longer than that at the lower relative collision velocity.

Fig. 2-20 compares magnitude of the liquid bridge forces calculated from the present simulation and an analytical model proposed by Mikami et al. [17]. In each relative collision velocity, magnitude of the liquid bridge force calculated from the present simulation showed much higher than that obtained by an analytical model at the smaller
separation distance. This results from the large capillary pressure force as indicated in Fig. 2-19. These results revealed that the large capillary pressure force acts on particles colliding through the dynamic liquid bridge. This cannot be modeled by any analytical models for the static liquid bridge force.
Fig. 2-19. Liquid bridge forces acted on particle-1 as a function of separation distance at different relative collision velocities. (a) $v = 1.0 \text{ m/s}$ (6.7 µs to 15.5 µs), (b) $v = 5.0 \text{ m/s}$ (1.7 µs to 4.3 µs). Insets show liquid bridge forces at smaller separation distance.

Fig. 2-20. Comparison of liquid bridge forces acted on particle-1 calculated from the present simulation and an analytical model as a function of separation distance during the separation at different relative collision velocities. (a) $v = 1.0 \text{ m/s}$ (10.2 µs to 15.5 µs), (b) $v = 5.0 \text{ m/s}$ (2.9 µs to 4.3 µs). Insets show liquid bridge forces at smaller separation distance.
4.4. Wet restitution coefficient and critical velocity for particle adhesion

Fig. 2-21 shows a wet restitution coefficient \( (e_{\text{wet}}) \) as a function of the relative collision velocity \( (v) \). The wet restitution coefficient \( (e_{\text{wet}}) \) was defined as follows:

\[
e_{\text{wet}} = \frac{v_{1,\text{rup}} - v_{2,\text{rup}}}{v_{1,\text{init}} - v_{2,\text{init}}} \tag{2-58}
\]

where \( v_{1,\text{rup}} \) and \( v_{2,\text{rup}} \) are the particle velocities when the liquid bridge was ruptured. \( e_{\text{wet}} = 0 \) means adhesion of two particles, while \( e_{\text{wet}} > 0 \) means separation of two particles. As indicated in Fig. 2-21, at the lower relative collision velocities less than 2.5 m/s, \( e_{\text{wet}} \) was zero. However, with an increase in the relative collision velocity, \( e_{\text{wet}} \) became higher than zero at a specific relative collision velocity. With further increase in the relative collision velocity, \( e_{\text{wet}} \) showed a steep increase, followed by a gradual increase. This result is qualitatively consistent with an experimental result of collision between a particle and a liquid film, reported in Sutkar et al. [22]. From these simulation results, the critical velocity for the particle adhesion \( v_c \) was determined as 2.9 m/s.

This result was compared to a value estimated by an analytical model proposed by Ennis et al. [23]. They proposed a model based on a lubrication theory, in which the particle-particle adhesion by the dynamic pendular liquid bridge was simplified as an adhesion phenomenon between particles coated with a thin liquid film (Fig. 2-22). This model was expressed as follows:

\[
v_c = \frac{3 \pi \mu R_p}{2 m_p} \left( 1 + \frac{1}{e} \right) \ln \left( \frac{z}{z_a} \right) \tag{2-59}
\]

where \( e \), \( z \) and \( z_a \) are the restitution coefficient of solid particle, thickness of the liquid film, and particle surface roughness, respectively. Here, the ratio \( z/z_a \) was assumed as \( z/z_a = 20 \), which is used in literatures [23, 24], while the other parameters were taken from Table 2-4. As a result, the \( v_c \) calculated from Eq. (2-59) was 0.16 m/s. This \( v_c \) is
much smaller than the $v_c$ determined from the simulation results (2.9 m/s). Therefore, it was demonstrated that there is a large difference between the result obtained by the present simulation and that obtained from the analytical model.

Although the analytical model proposed by Ennis et al. [23] qualitatively explains the particle adhesion with the viscous decay, this model contains an uncertain parameter $z_a$, which strongly influences the value of $v_c$. Thus, it is hard to use the analytical model proposed by Ennis et al. [23] for a quantitative analysis. On the other hand, no uncertain parameters are contained in the present simulation model. Consequently, the present simulation model can be used for quantitative analysis of the particle-particle adhesion by the dynamic liquid bridge.
Fig. 2-21. Wet restitution coefficient as a function of relative collision velocity.

Fig. 2-22. Schematic of two colliding granules proposed by Ennis et al.
5. Conclusions

This chapter presents a numerical modeling of particle-particle adhesion by a dynamic liquid bridge. To verify the simulation results, three verifications and validations were conducted. It was confirmed that the simulation model can accurately calculate (i) surface tension force acting on liquid, (ii) static liquid bridge force between particles, and (iii) rupture behavior of liquid bridge with free falling of a particle. The adhesion of two colliding particles through a droplet on a particle surface was then simulated. From the simulation results, the critical velocity for the particle adhesion $v_c$ was determined. This critical velocity was compared to that estimated from analytical model proposed by Ennis et al. [23]. As a result, it was demonstrated that there is a big difference between the critical velocity obtained by the present simulation and that obtained by the previous analytical model.

In this chapter, the validity of the simulation model for analyzing the particle-particle adhesion by the dynamic liquid bridge was confirmed. From next chapter, effects of critical parameters on the particle-particle adhesion by the dynamic liquid bridge were investigated using this simulation model.

Nomenclature

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>$A$</td>
<td>parameter shown in Eq. (2-54)</td>
<td>[-]</td>
</tr>
<tr>
<td>$B$</td>
<td>parameter shown in Eq. (2-54)</td>
<td>[-]</td>
</tr>
<tr>
<td>$C$</td>
<td>parameter shown in Eq. (2-54)</td>
<td>[-]</td>
</tr>
<tr>
<td>$d$</td>
<td>distance shown in Fig. 2-5</td>
<td>[m]</td>
</tr>
<tr>
<td>$e$</td>
<td>restitution coefficient between solid particles</td>
<td>[-]</td>
</tr>
<tr>
<td>$e_{\text{wet}}$</td>
<td>wet restitution coefficient</td>
<td>[-]</td>
</tr>
<tr>
<td>$F_{cp}$</td>
<td>capillary pressure force</td>
<td>[N]</td>
</tr>
<tr>
<td>$F_d$</td>
<td>viscous drag force</td>
<td>[N]</td>
</tr>
<tr>
<td>$F_{LB}$</td>
<td>static liquid bridge force</td>
<td>[N]</td>
</tr>
<tr>
<td>$\hat{F}_{LB}$</td>
<td>dimensionless static liquid bridge force</td>
<td>[-]</td>
</tr>
<tr>
<td>$F_{sf}$</td>
<td>surface tension force at solid-liquid-gas interface</td>
<td>[N]</td>
</tr>
</tbody>
</table>
\( f_p \): interaction force from particle to fluid \[ \text{[m/s}^2] \]
\( f_{sf} \): surface tension force \[ \text{[m/s}^2] \]
\( g \): gravity acceleration \[ \text{[m/s}^2] \]
\( h \): surface distance between particles \[ \text{[m]} \]
\( \hat{h} \): dimensionless surface distance between particles \[ \text{[–]} \]
\( I_p \): particle inertia \[ \text{[kg} \cdot \text{m}^2] \]
\( M_d \): torque associated with viscous drag force \[ \text{[N} \cdot \text{m]} \]
\( M_{sf} \): torque associated with surface tension force \[ \text{[N} \cdot \text{m]} \]
\( m_p \): particle mass \[ \text{[kg]} \]
\( n_c \): unit normal vector along gas-liquid interface at contact line \[ \text{[–]} \]
\( n_{lg} \): unit normal vector at solid-liquid-gas interface \[ \text{[–]} \]
\( n_s \): unit normal vector at solid surface \[ \text{[–]} \]
\( p \): fluid pressure \[ \text{[Pa]} \]
\( R_p \): particle radius \[ \text{[m]} \]
\( r \): relative position from center of gravity of particle to fluid cell \[ \text{[–]} \]
\( t \): time \[ \text{[s]} \]
\( t_c \): unit tangential vector along gas-liquid interface at contact line \[ \text{[–]} \]
\( t_s \): unit tangential vector at solid surface \[ \text{[–]} \]
\( u \): fluid velocity \[ \text{[m/s]} \]
\( u^C \): fluid velocity in Eq. (2-25) \[ \text{[m/s]} \]
\( u_p \): particle velocity in Eq. (2-25) \[ \text{[m/s]} \]
\( V_{LB} \): liquid bridge volume \[ \text{[m}^3] \]
\( \dot{V}_{LB} \): dimensionless liquid bridge volume \[ \text{[–]} \]
\( V_p \): particle volume \[ \text{[m}^3] \]
\( v_c \): critical velocity for particle adhesion \[ \text{[m/s]} \]
\( v_p \): translational velocity of particle \[ \text{[m/s]} \]
\( v_i \): translational velocity of particle just before collision \[ \text{[m/s]} \]
\( v_{i,init} \): initial translational velocity of particle \[ \text{[m/s]} \]
\( v_{i,re} \): translational velocity of particle just after rebound \[ \text{[m/s]} \]
\( v_{i,rup} \): translational velocity of particle when liquid bridge is ruptured \[ \text{[m/s]} \]
Greek letters

\( \alpha \) : half-filling angle [rad]
\( \phi \) : color function [–]
\( \phi_s \) : volume fraction of solid [–]
\( \kappa \) : local curvature of liquid-gas interface [1/m]
\( \mu_f \) : fluid viscosity [Pa·s]
\( \mu_g \) : gas viscosity [Pa·s]
\( \mu_l \) : liquid viscosity [Pa·s]
\( \rho_f \) : fluid density [kg/m³]
\( \rho_g \) : gas density [kg/m³]
\( \rho_l \) : liquid density [kg/m³]
\( \sigma \) : surface tension coefficient [N/m]
\( \theta \) : static contact angle [deg]
\( \theta_{cal} \) : calculated contact angle [deg]
\( \omega_p \) : rotational velocity of particle [rad/s]
\( \omega_i \) : rotational velocity of particle just before collision [rad/s]
\( \omega_{i.re} \) : rotational velocity of particle just after rebound [rad/s]

References


Chapter III

Effect of particle wettability on particle-particle adhesion of colliding particles through droplet
1. Introduction

Particle wettability is a critical factor affecting quality of granules [1, 2]. The particle wettability is generally evaluated by the contact angle between particle and liquid. The effect of contact angle on the wet granulation characteristics have been investigated by some experiments [2-5]. Saleh and Guigon [2] investigated the effect of contact angle on the particle growth rate of powders in a fluidized bed granulation. However, the effect of particle adhesion phenomenon by the dynamic liquid bridge at the primary particles scale cannot be quantitatively analyzed by the experimental approaches.

Some theoretical studies on the effects of contact angle on the static liquid bridge force and rupture distance of the liquid bridge have been reported [6-8]. Mikami et al. [6] proposed a mathematical model for the static liquid bridge acting on a particle and rupture distance of the liquid bridge as a function of liquid volume, contact angle, and separation distance. In accordance with their model, with an increase in the contact angle, the static liquid bridge force monotonically decreases and the rupture distance increases. However, the effect of contact angle on the particle adhesion phenomenon by the dynamic liquid bridge cannot be explained by the conventional models for the static liquid bridge.

In this chapter, the effect of particle wettability on the particle adhesion phenomenon by a dynamic liquid bridge was numerically analyzed by means of the direct numerical simulation proposed in Chapter II. In particular, the effect of contact angle on a critical velocity for the particle adhesion was analyzed and role of the wettability in the particle adhesion phenomenon by the dynamic liquid bridge was discussed.
2. Simulation set-up

In this chapter, to simulate the behaviors of the liquid bridge and particles, the direct numerical simulation model described in Chapter II was used.

Fig. 3-1 shows the initial configurations at various contact angles. A single droplet was adhered on a particle surface. \( v_{1,\text{init}} \) and \( v_{2,\text{init}} \) are the initial velocities of the particle-1 and particle-2, respectively. \( \theta \) and \( h_{\text{drop}} \) are the contact angle and distance between surfaces of the droplets, respectively. In each simulation, the two particles were collided with each other in the normal direction at various collision velocities \( v \) (\( v = |v_{1,\text{init}} - v_{2,\text{init}}| \)), while magnitude of the initial velocity of each particle was set as constant (\( |v_{1,\text{init}}| = |v_{2,\text{init}}| \)). Under this condition, the particles did not rotate. Therefore, rotational motion of the particle (expressed by Eq. (2-28) in Chapter II) was not considered. Table 3-1 lists calculation conditions. The physical properties of water and lactose were used for the calculation conditions of the liquid and spherical particle, respectively. The droplet volume was set based on a typical spray mist volume used in a fluidized bed spray granulation [9]. The particle-particle adhesion was simulated at various contact angles (\( \theta \)). In the present study, to simply analyze the effect of contact angle on the particle adhesion phenomenon, a static contact angle was adapted. Distance between surfaces of the droplets (\( h_{\text{drop}} \) in Fig. 3-1) at the initial condition was set as 1.6 \( \mu \text{m} \) regardless of the contact angle. Under these conditions, Bond number, which is the ratio of gravity to surface tension, was \( 8.47 \times 10^{-5} \ll 1 \). The influence of gravity is negligible at such much smaller Bond number [10]. Therefore, the gravity was not considered.
Table 3-1 Calculation conditions

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Particle diameter</td>
<td>50.0 µm</td>
</tr>
<tr>
<td>Particle density</td>
<td>1500.0 kg/m³</td>
</tr>
<tr>
<td>Restitution coefficient between solid particles</td>
<td>1.0</td>
</tr>
<tr>
<td>Liquid density</td>
<td>997.0 kg/m³</td>
</tr>
<tr>
<td>Liquid viscosity</td>
<td>8.94×10⁻⁴ Pa·s</td>
</tr>
<tr>
<td>Surface tension coefficient</td>
<td>0.072 N/m</td>
</tr>
<tr>
<td>Droplet volume</td>
<td>1150.3 µm³</td>
</tr>
<tr>
<td>Contact angle</td>
<td>15.0 – 120.0 deg</td>
</tr>
<tr>
<td>Gas density</td>
<td>1.184 kg/m³</td>
</tr>
<tr>
<td>Gas viscosity</td>
<td>1.82×10⁻⁵ Pa·s</td>
</tr>
<tr>
<td>Time step</td>
<td>2.0×10⁻⁹ s</td>
</tr>
<tr>
<td>Fluid cell size</td>
<td>0.813 µm</td>
</tr>
</tbody>
</table>

Fig. 3-1. Initial configurations at various contact angles.
3. Results and discussion

3.1. Wet restitution coefficient and critical velocity for particle adhesion

Fig. 3-2 shows behaviors of particles and liquid bridge at different contact angles. At $\theta = 30$ deg, concave-shaped liquid bridge was formed both in approach stage (Fig. 3-2 (a) $t = 1.9 \, \mu s$) and in separation stage (Fig. 3-2 (a) $t = 15.5 \, \mu s$). The liquid bridge was elongated but then shrunk, and the particles were eventually adhered (Fig. 3-2 (a) $t = 39.5 \, \mu s$). On the other hand, at $\theta = 120$ deg, convex-shaped liquid bridge was formed (Fig. 3-2 (b) $t = 2.2 \, \mu s$ and $t = 19.5 \, \mu s$). The liquid was repelled from the particle surface, and the particles were eventually separated (Fig. 3-2 (b) $t = 25.5 \, \mu s$). Thus, it can be seen that behaviors of the particles and liquid was largely different depending on the contact angle even at the same relative collision velocity.

To quantitatively analyze the particle behaviors at different contact angles, a wet restitution coefficient ($e_{\text{wet}}$) was calculated:

$$
e_{\text{wet}} = -\frac{v_{1,\text{rup}} - v_{2,\text{rup}}}{v_{1,\text{init}} - v_{2,\text{init}}} \tag{3-1}$$

where $v_{1,\text{rup}}$ and $v_{2,\text{rup}}$ are the particle velocities when the liquid bridge was ruptured. Fig. 3-3 indicates a wet restitution coefficient as a function of the relative collision velocities at different contact angles. At a specific relative collision velocity ($v_c$ in Fig. 3-3), $e_{\text{wet}}$ became higher than zero. This velocity was defined as the critical velocity for the particle adhesion. Over the critical velocity, $e_{\text{wet}}$ increased with an increase in the relative collision velocity. At the much higher relative collision velocity than 12.0 m/s, $e_{\text{wet}}$ showed almost same value regardless of the contact angle.

From these results, the critical velocity for the particle adhesion ($v_c$) was determined at various contact angles. This corresponds to adhesiveness of the two colliding particles. Fig. 3-4 describes the critical velocity for the particle adhesion as a function of the contact angle. With an increase in the contact angle, the critical velocity increased and reached a local maximum value at $\theta = 60$ deg, followed by a decrease. Thus, the critical velocity non-monotonically changed with the contact angle. As explained by an
analytical model proposed by Mikami et al. [6], a static liquid bridge force monotonically decreases with an increase in the contact angle. Therefore, it was found that non-monotonic dependence of the critical velocity on the contact angle, which cannot be explained by the static liquid bridge force models.
Fig. 3-2. Simulation results of particles and liquid bridge behaviors at different contact angles ($v = 2.0$ m/s).
Fig. 3-3. Wet restitution coefficient as a function of relative collision velocity at different contact angles.

Fig. 3-4. Critical velocity for particle adhesion as a function of contact angle.
3.2. Dissipation energy of particle motion

To understand the non-monotonic dependence of the critical velocity on the contact angle, a dissipation energy of particle motion, expressing how much the liquid bridge dissipated the particle motion, was calculated. This dissipation energy \( E \) was defined as following equations:

\[
E = \int_{0}^{x_0} \beta(x) |F(x)| dx + \int_{0}^{x_{\text{max}}} \beta(x) |F(x)| dx
\]  

(3-2)

\[
\beta(x) = \begin{cases} 
1 & \text{if } \text{sgn}(v_p \cdot n) \neq \text{sgn}(F(x) \cdot n) \\
-1 & \text{if } \text{sgn}(v_p \cdot n) = \text{sgn}(F(x) \cdot n)
\end{cases}
\]  

(3-3)

where \( x \) is the separation distance between particles, \( F(x) \) is the total liquid bridge force acting on a particle. \( x_0 \) means the separation distance when the liquid bridge was formed in the approach stage, while \( x_{\text{max}} \) means the separation distance at the maximum elongation length of the liquid bridge in the separation stage. In Eq. (3-2), the first and second terms of the right-hand side mean the dissipation energy in the approach and separation stages, respectively. \( \beta(x) \) is the sign function defined as Eq. (3-3), \( v_p \) is the translational velocity of the particle, and \( n \) is the unit normal vector from objective particle to another particle. According to this sign function, the energy dissipating the particle motion was defined as positive, while the energy accelerating the particle motion was defined as negative. Fig. 3-5 shows the dissipation energy \( E \) as a function of the contact angle at \( v = 3.0 \) m/s. Dissipation energy in the approach and separation stages are also shown. With an increase in the contact angle, the total dissipation energy increased to \( \theta = 60 \) deg, followed by a decrease. This non-monotonic dependence on the contact angle was consistent with the critical velocity for the particle adhesion (Fig. 3-4). Fig. 3-6 indicates the critical velocity as a function of the dissipation energy. It was confirmed that the critical velocity was well correlated with the dissipation energy. Therefore, the dissipation energy was focused in the following section. For a further understanding, the dissipation energy was decomposed into contributions from the approach and separation stages and discussed separately.
Fig. 3-5. Dissipation energy of particle motion as a function of contact angle ($v = 3.0 \text{ m/s}$).

Fig. 3-6. Critical velocity for particle adhesion as a function of dissipation energy of particle motion ($v = 3.0 \text{ m/s}$).
3.2.1. Approach stage

As shown in Fig. 3-5, with an increase in the contact angle, the dissipation energy in the approach stage monotonically increased. The dissipation energy was shifted from negative to positive between $\theta = 60$ deg and 75 deg. This means that the particles approaching were accelerated at lower than $\theta = 60$ deg, while the particles approaching were dissipated at higher than $\theta = 75$ deg. To understand this result, the total liquid bridge force ($F(x)$) and the separation distance when the liquid bridge was formed ($x_0$) were separately analyzed.

Fig. 3-7 indicates the total liquid bridge force acting on the particle-1 in the approach stage as a function of the separation distance at different contact angles. In Fig. 3-7, positive value means the force acted as a dissipative force, while negative value means the force acted as an acceleration force. With an increase in the contact angle, the total liquid bridge force increased, indicating that the force acts as higher dissipative force for the particle motion at higher contact angle. For further understanding, all components of the liquid bridge forces (capillary pressure force $F_{cp}$, drag force $F_d$, and surface tension force $F_{sf}$) were analyzed. Fig. 3-8 describes all components of the liquid bridge forces acting on the particle-1 as a function of the separation distance. At the relatively lower contact angles ($\theta \leq 60$ deg), the surface tension force ($F_{sf}$) acted as an acceleration force regardless of the separation distance. The drag force ($F_d$) acted as an acceleration force for larger separation distance, while the drag force increased and acted as a dissipative force with approaching the particles. The capillary pressure force ($F_{cp}$) acted as a dissipative force, because the liquid bridge between the approaching particles was largely compressed and the pressure inside the liquid bridge increased. From these results, the surface tension force ($F_{sf}$) and the drag force ($F_d$) largely contributed to the acceleration of the particle motion at the lower contact angles. At the relatively higher contact angles ($\theta \geq 90$ deg), the surface tension force ($F_{sf}$) acted as an acceleration force, while the drag force ($F_d$) and capillary pressure force ($F_{cp}$) acted as dissipative forces regardless of the separation distance. At the higher contact angles, convex-shaped liquid bridge was formed between colliding particles as shown in Fig. 3-9 (b). In this case, the pressure inside the liquid bridge became highly positive, and the capillary pressure force
(\(F_{cp}\)) acts as a large dissipative force. From these results, it was found that the drag force (\(F_d\)) and capillary pressure force (\(F_{cp}\)) largely contributed to the dissipation of the particle motion in the approach stage. In addition, as shown in Fig. 3-10, the separation distance when the liquid bridge was formed (\(x_0\)), i.e., the initial length of the liquid bridge, increased with an increase in the contact angle, because the initial height of the droplet on the particle surface increased (Fig. 3-1). This results in higher dissipation energy at the higher contact angle due to increase in the integration interval of the first term in Eq. (3-2). In summary, it was found that the increase in the dissipation energy in the approach stage at higher contact angle is derived from higher drag force (\(F_d\)) and capillary pressure force (\(F_{cp}\)) as well as higher initial length of the liquid bridge. However, the non-monotonic dependence of the total dissipation energy cannot still be explained.
Fig. 3-7. Total liquid bridge force acting on particle-1 in approach stage as a function of separation distance at different contact angles ($v = 3.0 \text{ m/s}$).
Fig. 3-8. Liquid bridge forces acting on particle-1 in approach stage as a function of separation distance at different contact angles ($\theta = 3.0$ m/s).
Fig. 3-9. Snapshots of liquid bridge in approach stage at different contact angles ($v = 3.0$ m/s, $x = 5.0$ µm).

Fig. 3-10. Separation distance when liquid bridge was formed as a function of contact angle ($v = 3.0$ m/s).
3.2.2. Separation stage

As shown in Fig. 3-5, the dissipation energy of the particle motion in the separation stage was positive regardless of the contact angle. At $\theta = 15$ deg to 60 deg, the dissipation energy showed almost the same value, while at higher contact angles than $\theta = 60$ deg, the dissipation energy decreased with an increase in the contact angle. To understand why the dissipation energy in the separation stage showed such non-monotonic relationship, the total liquid bridge force ($F(x)$) and the maximum elongation length of the liquid bridge ($x_{\text{max}}$) were analyzed.

Fig. 3-11 indicates the total liquid bridge force acting on the particle-1 in the separation stage as a function of the separation distance at different contact angles. In Fig. 3-11, positive value means the force acted as a dissipative force, while negative value means the force acted as an acceleration force. With a decrease in the contact angle, the total liquid bridge force increased, indicating that the force acts as higher dissipative force for the particle motion at the lower contact angle. Fig. 3-12 describes all components of the liquid bridge forces acting on the particle-1 as a function of the separation distance. At the relatively lower contact angles ($\theta \leq 60$ deg), the surface tension force ($F_{\text{sf}}$) and drag force ($F_d$) acted as a dissipative force regardless of the separation distance. Although the capillary pressure force ($F_{\text{cp}}$) acted as a dissipative force after the particle-particle collision, the capillary pressure force steeply decreased and reached almost zero with separating the particles. From these results, the surface tension force ($F_{\text{sf}}$) and drag force ($F_d$) largely contributed to the dissipation of the particle motion at the lower contact angles. At the relatively higher contact angles ($\theta \geq 90$ deg), the surface tension force ($F_{\text{sf}}$) also acted as a dissipative force regardless of the separation distance. The drag force ($F_d$) and capillary pressure force ($F_{\text{cp}}$) acted as high dissipative forces just after particle-particle collision, followed by a steeply decrease with separating the particles. The drag force ($F_d$) converted to zero, while the capillary pressure force ($F_{\text{cp}}$) highly acted as an acceleration force. This is due to the highly positive pressure inside the convex-shaped liquid bridge at the higher contact angle. From these results, at the higher contact angles, the capillary pressure force ($F_{\text{cp}}$) largely contributed to the acceleration of the particle motion. However, the
Fig. 3-11. Total liquid bridge force acting on particle-1 in separation stage as a function of separation distance at different contact angles ($v = 3.0$ m/s).
Fig. 3-12. Liquid bridge forces acting on particle-1 in separation stage as a function of separation distance at different contact angles ($v = 3.0$ m/s).
non-monotonic dependence of the dissipation energy (Fig. 3-5) on the contact angle cannot be explained only by the liquid bridge force acting on a particle.

The maximum elongation length of the liquid bridge ($x_{max}$ in Eq. (3-2)) was then focused. Fig. 3-13 shows the maximum elongation length of the liquid bridge in the separation stage as a function of the contact angle. With an increase in the contact angle, the maximum elongation length increased and showed local maximum value at $\theta = 60$ deg, followed by a decrease. This non-monotonic dependence of the maximum elongation length on the contact angle is considered to determine the non-monotonic dependence of the dissipation energy. To understand this result, deformation behavior of the liquid bridge was observed. Fig. 3-14 indicates deformation behaviors of the liquid bridge in the separation stage at different contact angles. At the lower contact angle (Fig. 3-14 (a)), with separating the particles, the neck radius of the liquid bridge shrank and the liquid bridge was ruptured at center of the bridge. This is due to relatively hydrophilic surface of the particle. On the other hand, at the higher contact angle (Fig. 3-14 (d)), with separating the particles, the neck radius of the liquid bridge increased and the liquid was repelled from the particle surface. This is due to relatively hydrophobic surface of the particle. At $\theta = 60$ deg and 75 deg, (Fig. 3-14 (b) and (c)), the liquid bridge formed a cylinder-like shape. This results in the longer duration and elongation of the liquid bridge. Consequently, the dissipation energy in the separation stage was determined not only by the liquid bridge force but the elongation behavior of the liquid bridge. In summary, it was revealed that a combined effect of the instantaneous liquid bridge force and dynamics of the liquid bridge deformation results in the non-monotonic dependence of the critical velocity for the particle adhesion on the contact angle (Fig. 3-4).
Fig. 3-13. Maximum elongation length of liquid bridge as a function of contact angle ($v = 3.0$ m/s).
Fig. 3-14. Snapshots of liquid bridge in separation stage at different contact angles ($v = 3.0$ m/s).
4. Conclusions

In this chapter, the effect of particle wettability on the particle-particle adhesion of two colliding particles through a droplet was investigated. In particular, the effect of contact angle on a critical velocity for the particle adhesion phenomenon was analyzed. It was found that the critical velocity for the particle adhesion non-monotonically changed with the contact angle. With an increase in the contact angle, the critical velocity increased and reached a local maximum value at \( \theta = 60 \) deg, followed by a decrease. To understand this result, the dissipation energy of the particle motion was analyzed. In the approach stage, the dissipation energy monotonically increased with an increase in the contact angle. However, in the separation stage, the dissipation energy showed non-monotonic dependence on the contact angle. This was derived from non-monotonic dependence of the maximum elongation length of the liquid bridge on the contact angle. From these findings, it was revealed that a combined effect of the instantaneous liquid bridge force and dynamics of the liquid bridge deformation results in the non-monotonic dependence of the critical velocity for the particle adhesion on the particle wettability.
Nomenclature

*E*: dissipation energy of particle motion  
*\(\varepsilon\)_{wet}*: wet restitution coefficient  
*F*: total liquid bridge force  
*\(\mathbf{F}_{cp}\)*: capillary pressure force  
*\(\mathbf{F}_d\)*: viscous drag force  
*\(\mathbf{F}_{sf}\)*: surface tension force at solid-liquid-gas interface  
*\(h_{\text{drop}}\)*: surface distance between droplets at initial condition  
*n*: unit normal vector from objective particle to another particle  
*u_p*: particle velocity  
*\(v\)*: relative collision velocity  
*\(v_c\)*: critical velocity for particle adhesion  
*\(v_{\text{init}}\)*: initial translational velocity of particle  
*\(v_{\text{rup}}\)*: particle velocity when liquid bridge is ruptured  
*x*: separation distance between particles  
*\(x_0\)*: separation distance when the liquid bridge is formed  
*\(x_{\text{max}}\)*: separation distance at maximum elongation length of liquid bridge

Greek letters

*\(\beta\)*: sign function  
*\(\theta\)*: static contact angle
References


Chapter IV

Effect of droplet size on particle-particle adhesion of colliding particles through droplet
1. Introduction

Droplet size of a binder liquid is a critical factor affecting quality of the granules. It has been recognized that the nuclei size of granules is determined by size distribution of the droplets [1]. Some experimental studies investigating the effect of sprayed binder liquid size on the performance of wet granulation have been reported [2-6]. Tan et al. [5] investigated the particle growth rate and the final granule size at different droplet sizes under constant amount of sprayed binder liquid in the fluidized bed granulation. They found that the overall growth rate and final granule size increased with an increase in the droplet size. However, the effect of droplet size on the particle-particle adhesion by the dynamic liquid bridge cannot be quantitatively analyzed by the experimental approaches. In this chapter, the particle-particle adhesion under a constant total liquid volume, where number of droplets on a particle surface is varied depending on the droplet size, was focused. This resulted in the formation of multiple pendular liquid bridges between particles. The particle-particle adhesion by multiple liquid bridges cannot be analyzed by experiments and previous theoretical results due to their complexities.

In this chapter, the effect of droplet size of binder liquid under constant liquid volume on the particle-particle adhesion by the dynamic liquid bridge was analyzed. In particular, the effect of droplet size on a critical velocity for particle adhesion was investigated and microscopic mechanism of the particle adhesion was discussed.
2. Simulation set-up

In this chapter, to simulate the behaviors of the liquid bridge and particles, the direct numerical simulation model described in Chapter II was used.

In this investigation, equivalent volume diameter of a droplet \(d_{\text{drop}}\) was varied under constant liquid volume, i.e., the number of droplets \(N_{\text{drop}}\) on a particle surface was changed in each droplet size. As illustrated in Fig. 4-1, a condition where the moisture content is relatively high and the particle surface is covered several droplets was focused. To solely investigate the effect of droplet size, a condition where droplets are located symmetrically at the contact area between two colliding particles was also focused. Fig. 4-2 shows droplets arrangement on a particle surface at various droplet diameters. The number of droplets \(N_{\text{drop}}\) was changed from 1 to 5, corresponding that the range of droplet diameter \(d_{\text{drop}}\) from 10.0 to 5.85 μm. Fig. 4-3 indicates initial configurations. Droplets were symmetrically adhered on the two particle surfaces so that multiple liquid bridges can be formed simultaneously when the two particles approach. In each simulation, the two particles were collided with each other in the normal direction at various collision velocities \(v = \|v_{1,\text{init}} - v_{2,\text{init}}\|\), while magnitude of the initial velocity of each particle was set as constant \(\|v_{1,\text{init}}\| = \|v_{2,\text{init}}\|\). Rotational motion of the particle (expressed by Eq. (2-28) in Chapter II) was not considered. Table 4-1 lists calculation conditions. The physical properties of water and lactose were set as calculation conditions of the liquid and spherical particle, respectively. The droplet volume was set based on a typical spray mist volume used in a fluidized bed spray granulation [7]. Under these conditions, Bond number was \(8.47 \times 10^{-5} << 1\), i.e., the influence of gravity can be ignored [8].
\[ d_{\text{drop}} = 10.0 \, \mu m \quad (N_{\text{drop}} = 1) \]
\[ d_{\text{drop}} = 7.94 \, \mu m \quad (N_{\text{drop}} = 2) \]
\[ d_{\text{drop}} = 6.93 \, \mu m \quad (N_{\text{drop}} = 3) \]
\[ d_{\text{drop}} = 6.30 \, \mu m \quad (N_{\text{drop}} = 4) \]
\[ d_{\text{drop}} = 5.85 \, \mu m \quad (N_{\text{drop}} = 5) \]

**Fig. 4-1.** Schematic of droplets and particles.

**Fig. 4-2.** Initial arrangement of droplets on particle surface at various droplet diameters.
**Table 4-1 Calculation conditions**

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
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<tr>
<td>Particle diameter</td>
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<tr>
<td>Particle density</td>
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<tr>
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<td>Liquid viscosity</td>
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</tr>
<tr>
<td>Surface tension coefficient</td>
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<tr>
<td>Contact angle</td>
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<tr>
<td>Gas density</td>
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</tr>
<tr>
<td>Gas viscosity</td>
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<td>Time step</td>
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</tr>
<tr>
<td>Fluid cell size</td>
<td>0.685 µm</td>
</tr>
</tbody>
</table>

**Fig. 4-3.** Initial configurations at various droplet diameters.
3. Results and discussion

3.1. Wet restitution coefficient and critical velocity for particle adhesion

Fig. 4-4 shows behaviors of the particles and multiple liquid bridges at different droplet diameters. At \( d_{\text{drop}} = 10.0 \, \mu m \) \((N_{\text{drop}} = 1)\), with approaching the particles, the liquid bridge was formed \((\text{Fig. 4-4} \ (a) \ t = 1.6 \, \mu s)\) and the particles then collided with each other \((\text{Fig. 4-4} \ (a) \ t = 4.1 \, \mu s)\). After the collision, the liquid bridge was largely elongated \((\text{Fig. 4-4} \ (a) \ t = 20.0 \, \mu s)\), and eventually the liquid bridge was ruptured \((\text{Fig. 4-4} \ (a) \ t = 27.0 \, \mu s)\). At \( d_{\text{drop}} = 5.85 \, \mu m \) \((N_{\text{drop}} = 5)\), with approaching the particles, five liquid bridges were initially formed \((\text{Fig. 4-4} \ (b) \ t = 1.1 \, \mu s)\). The particles then collided with each other, and the liquid bridges partially coalesced \((\text{Fig. 4-4} \ (b) \ t = 1.9 \, \mu s)\). After the collision, the liquid bridge completely coalesced into a single liquid bridge and was elongated \((\text{Fig. 4-4} \ (b) \ t = 17.0 \, \mu s)\). Finally, the liquid bridge was ruptured \((\text{Fig. 4-4} \ (b) \ t = 20.2 \, \mu s)\).

To quantitatively analyze the particle behavior at different droplet diameters, a wet restitution coefficient \( e_{\text{wet}} \) (expressed by Eq. (3-1) in Chapter III) was calculated. Fig. 4-5 indicates a wet restitution coefficient as a function of the relative collision velocity at different droplet diameters. At the lower relative collision velocity, \( e_{\text{wet}} \) was zero, i.e., the two particles were adhered. However, \( e_{\text{wet}} \) became larger than zero at a specific relative collision velocity, i.e., the two particles were separated. At the much higher relative collision velocities, \( e_{\text{wet}} \) showed almost same value regardless of the droplet diameter.

From these simulation results, the critical velocity for the particle adhesion \((v_c)\) was determined at various droplet diameters. Fig. 4-6 shows the critical velocity for the particle adhesion as a function of the droplet diameter. The critical velocity increased with a decrease in the droplet diameter. Generally, in a wet granulation, granule growth rate and final granule size have been reported to increase with an increase in the droplet size. This tendency seems to be different from the present simulation results. However, when a granule growth at the initial stage was focused, tendency of the present simulation results was consistent with that of an experimental result. Tan et al. [5] experimentally analyzed growth process of granules in a fluidized bed spray.
Fig. 4-4. Simulations results of particles and droplets behaviors at different droplet diameters under constant liquid volume ($v = 3.25$ m/s).
Fig. 4-5. Wet restitution coefficient as a function of relative collision velocity at different droplet diameters under constant liquid volume.

Fig. 4-6. Critical velocity for particle adhesion as a function of droplet diameter under constant liquid volume.
granulation. They investigated the effect of binder droplet size on the granule growth under constant total binder liquid amount. Moreover, in their experiment, it can be considered that a pendular liquid bridge was formed because of much smaller droplet size than the particle size. These experimental conditions were very similar with the present simulation conditions. Therefore, this experimental study is suitable for comparison with the present simulation result. Fig. 4-7 shows granule growth as a function of total binder liquid at different atomizing pressures. $B$ means binder mass (time $\times$ spray rate) and $P$ means particle mass. $l_{50}$ means median size of granules. With an increase in the atomizing pressure, droplet size decreased. The simulation result was compared with the initial stage of the experimental result, because the simulation set-up corresponds to the initial stage of wet granulations. As indicated in Fig. 4-7, at the initial stage, higher granule growth rate was shown at smaller droplet size (i.e., higher atomizing pressure). This tendency was consistent with the simulation result. However, when overall growth rate and final granule size were focused, higher granule growth rate and larger granule size were obtained at larger droplet size. This tendency was opposite to the present simulation result. This result indicated that not only primary scale particle-particle adhesion but other important rate process such as drying, solidification, growth, and breakage may play a critical role for overall particle growth in a wet granulation.
Fig. 4-7. Granule growth at different atomizing pressures. Reprinted with permission from [5]. Copyright: (2005) Elsevier Ltd.
3.2. Dissipation energy of particle motion.

To understand the simulation result of critical velocity shown in Fig. 4-6, a dissipation energy of the particle motion $E$ (expressed by Eqs. (3-2) and (3-3) in Chapter III) was calculated. Fig. 4-8 shows the dissipation energy of the particle motion as a function of the droplet diameter at $v = 3.0$ m/s. Dissipation energies in the approach and separation stages are also shown. The total dissipation energy increased with a decrease in the droplet diameter. This tendency was consistent with the critical velocity for the particle adhesion (Fig. 4-6). For further understanding of this result, the dissipation energies in the approach and separation stages were separately analyzed in the following section.

3.2.1. Approach stage

As shown in Fig. 4-8, the dissipation energy in the approach stage decreased with an increase in the droplet diameter. To understand this result, the liquid bridge force acting on a particle and separation distance between particles when the liquid bridge was formed ($x_0$ in Eq. (3-2)) were separately analyzed.

Fig. 4-9 indicates the liquid bridge forces (drag force $F_d$, capillary force $F_{cp}$, and surface tension force $F_{st}$) acting on the particle-1 in the approach stage as a function of the separation distance at different droplet diameters. In Fig. 4-9, positive value means the force acted as a dissipative force, while negative value means the force acted as an acceleration force. The capillary pressure force ($F_{cp}$) acted as a dissipative force at each droplet diameter and increased at smaller droplet diameter. The pressure of the droplet on a particle surface was positive before the liquid bridge was formed. If the static liquid bridge is formed, the equilibrium pressure inside the liquid bridge becomes negative. However, in this simulation, with approaching the particles, the liquid bridge was largely compressed and the pressure inside the liquid bridge was kept as positive. Thus, the capillary pressure force ($F_{cp}$) acted as a dissipative force. In addition, magnitude of the internal pressure of a droplet increases with a decrease in the droplet diameter. This is also confirmed by the Young-Laplace equation. Therefore, in case of the smaller droplet diameter, the internal pressure and the number of droplets was
Fig. 4-8. Dissipation energy for particle motion as a function of droplet diameter ($v = 3.0 \text{ m/s}$).
Fig. 4-9. Liquid bridge force acting on particle-1 in approach stage as a function of separation distance at different droplet diameters ($v = 3.0 \text{ m/s}$).
higher, resulting in higher capillary pressure force ($F_{cp}$). This result cannot be derived from conventional static liquid bridge force models. The drag force ($F_d$) acted as an acceleration force at larger separation distance, while the drag force ($F_d$) acted as a dissipative force before particle-particle collision. The surface tension force ($F_d$) acted as an acceleration force regardless of the separation distance and the droplet diameter. From these results, the capillary pressure force ($F_{cp}$) largely contributed as a dissipative force in the approach stage. On the other hand, the separation distance when the liquid bridge was formed ($x_0$) decreased with a decrease in the droplet diameter as shown in Fig. 4-10, because the initial height of droplet on particle surface decreased with a decrease in the droplet diameter. Therefore, at the smaller droplet diameter, the dissipative force was significantly higher, although distance that the dissipative force acted was smaller. Consequently, this results in higher dissipation energy at the smaller droplet diameter (Fig. 4-8).
Fig. 4-10. Separation distance when liquid bridge was formed as a function of droplet diameter ($v = 3.0 \text{ m/s}$).
3.2.2. Separation stage

As shown in Fig. 4-8, the dissipation energy in the separation stage decreased with a decrease in the droplet diameter. This tendency was different from the result in the approach stage. To understand this result, the liquid bridge force acting on a particle and the maximum elongation length \( (x_{\text{max}} \text{ in Eq. (3-2)}) \) were separately analyzed.

Fig. 4-11 indicates the liquid bridge forces acting on the particle-1 in the separation stage as a function of the separation distance at different droplet diameters. The drag force \( (F_d) \) and the surface tension force \( (F_{st}) \) acted as dissipative forces regardless of the separation distance. The capillary pressure force \( (F_{cp}) \) acted as a dissipative force just after the particle-particle collision, followed by a decrease and reached almost zero. The liquid bridge forces acting on a particle in the separation stage were almost same regardless of the droplet diameter, i.e., tendency of the dissipation energy in the separation stage shown in Fig. 4-8 cannot be explained from the liquid bridge forces. The maximum elongation length of the liquid bridge was then analyzed. Fig. 4-12 shows the maximum elongation length of the liquid bridge in the separation stage. The maximum elongation length decreased with a decrease in the droplet diameter. At the larger droplet diameter, the distance that the dissipative force acted was longer, while the liquid bridge force acting on a particle was almost same. This results in larger dissipation energy at larger droplet diameter (Fig. 4-8).

To further understand this result, shape of the liquid bridge when the particles collided was focused. Fig. 4-13 shows shapes of the liquid bridge at the moment of the particle-particle collision. The shapes of the liquid bridge were significantly different depending on the droplet diameter even at the constant total liquid volume. At \( d_{\text{drop}} = 10.0 \, \mu\text{m} \) \( (N_{\text{drop}} = 1) \), the liquid bridge showed isotropic circular cylindrical shape (Fig. 4-13 (a)). At \( d_{\text{drop}} = 7.94 \, \mu\text{m} \) \( (N_{\text{drop}} = 2) \), the two liquid bridges coalesced and formed anisotropic cylindrical shape (Fig. 4-13 (b)). At \( d_{\text{drop}} = 6.93 \, \mu\text{m} \) to \( 5.85 \, \mu\text{m} \) \( (N_{\text{drop}} = 3 \text{ to } 5) \), the liquid bridges partially coalesced and formed an annulus-like shape (Fig. 4-13 (c), (d), (e)). From these results, it was hypothesized that the dependency of the maximum elongation length of the liquid bridge on the droplet diameter can be correlated with shapes of the liquid bridge at the particle-particle collision. To confirm
Fig. 4-11. Liquid bridge force acting on particle-1 in separation stage as a function of separation distance at different droplet diameters ($v = 3.0$ m/s).
Fig. 4-12. Maximum elongation length of liquid bridge as a function of droplet diameter ($v = 3.0$ m/s).
this consideration, the maximum elongation length of the liquid bridge against rebound velocity of the particle was analyzed. This rebound velocity means the velocity just after the particle-particle collision. In the present results, the maximum elongation length was different in accordance with the droplet diameter even at the same initial collision velocity (Fig. 4-12), because the dissipation energy in the approach stage was different (Fig. 4-8). This means that the rebound velocity of a particle-particle collision was different in accordance with the droplet diameter even at the same initial velocity. Therefore, the rebound velocity of a particle was focused. Fig. 4-14 shows the maximum elongation length of the liquid bridge as a function of the rebound velocity of the particle-1 at different droplet diameters. At $d_{\text{drop}} = 10.0$ to $6.93$ µm ($N_{\text{drop}} = 1$ to 3), where the shapes of the liquid bridges at the collision were not similar, the maximum elongation length of the liquid bridge decreased with a decrease in the droplet diameter. However, at $d_{\text{drop}} = 6.93$ to $5.85$ µm ($N_{\text{drop}} = 3$ to 5), where the shapes of the liquid bridges were almost similar, the maximum elongation length was linearly correlated with the rebound velocity regardless of the droplet diameter. From these results, it was found that the shapes of the liquid bridge at the particle-particle collision largely contributed to the maximum elongation length of the liquid bridge. In the present simulation, the rebound velocity decreased with a decrease in the droplet diameter at the same initial collision velocity, because the dissipation energy of the approach stage increased as shown in Fig. 4-8. Thus, at $d_{\text{drop}} = 6.93$ to $5.85$ µm ($N_{\text{drop}} = 3$ to 5), the maximum elongation length of the liquid bridge at the same initial collision velocity decreased at smaller droplet diameter (Fig. 4-12), although the maximum elongation length of the liquid bridge at the same rebound velocity showed same value (Fig. 4-14). On the other hand, at $d_{\text{drop}} = 10.0$ to $6.93$ µm ($N_{\text{drop}} = 1$ to 3), the maximum elongation length increased at the larger droplet diameter at the same initial collision velocity as well as the same rebound velocity (Figs. 4-12 and 4-14).

In summary, the capillary pressure force in the approach stage and the shape of the liquid bridge at the particle-particle collision largely contributed to the dissipation energy, and the tendency of the total dissipation energy was determined by the balance between dissipation energies in the approach and separation stages.
Fig. 4-13. Shapes of liquid bridge at particle-particle collision at various droplet diameters ($v = 3.0 \text{ m/s}$).

Fig. 4-14. Maximum elongation length of liquid bridge as a function of rebound velocity of particle-1 at different droplet diameters.
4. Conclusions

In this chapter, the effect of droplets size on the particle-particle adhesion of two colliding particles was investigated. In particular, the effect of droplet diameter under constant liquid volume on the critical velocity for the particle adhesion was analyzed. It was found that the critical velocity increased with a decrease in the droplet diameter. In an initial stage of the granule growth in a fluidized bed spray granulation, the tendency of the calculation result was consistent with an experimental result. To understand this result, the dissipation energy of the particle motion was analyzed. In the approach stage, the dissipation energy increased with a decrease in the droplet diameter. This was due to higher capillary pressure force which is acted as a dissipative force. On the other hand, in the separation stage, the dissipation energy decreased with a decrease in the droplet diameter. This was derived from difference in shapes of the liquid bridge at the particle-particle collision. Consequently, the dissipation energy of the particle motion was determined by the balance between dissipation energies in the approach and separation stages. In summary, it can be concluded that magnitude of the capillary pressure force and shapes of the liquid bridge are key factors for the particle-particle adhesion by the dynamic liquid bridge through the different sizes of binder droplets under constant total liquid volume.
Nomenclature

\( d_{\text{drop}} \): droplet diameter \[ \text{[m]} \]

\( E \): dissipation energy of particle motion \[ \text{[J]} \]

\( e_{\text{wet}} \): wet restitution coefficient \[ \text{[–]} \]

\( F_{\text{cp}} \): capillary pressure force \[ \text{[N]} \]

\( F_{\text{d}} \): viscous drag force \[ \text{[N]} \]

\( F_{\text{sf}} \): surface tension force at solid-liquid-gas interface \[ \text{[N]} \]

\( N_{\text{drop}} \): number of droplet \[ \text{[–]} \]

\( v \): relative collision velocity \[ \text{[m/s]} \]

\( v_{\text{c}} \): critical velocity for particle adhesion \[ \text{[m/s]} \]

\( v_{\text{i,init}} \): initial translational velocity of particle \[ \text{[m/s]} \]

\( v_{\text{i,re}} \): rebound velocity of particle \[ \text{[m/s]} \]

\( x \): separation distance between particles \[ \text{[m]} \]

\( x_0 \): separation distance when the liquid bridge is formed \[ \text{[m]} \]

\( x_{\text{max}} \): separation distance at maximum elongation length of liquid bridge \[ \text{[m]} \]
References


Chapter V

Effect of collision angle on particle-particle adhesion of colliding particles through droplet
1. Introduction

In an actual wet granulation process, particles randomly collide with each other, resulting in an oblique particle collision. This means that the particle-particle collision via a binder droplet occurs at various collision angles. Therefore, collision angle against a binder droplet on a particle surface is a critical factor for the particle-particle adhesion phenomenon.

When a particle collides with another particle through a binder droplet in an oblique direction, the liquid bridge force acts not only in the normal direction but in the tangential direction. However, previous studies on the static liquid bridge force only calculate a liquid bridge force in the normal direction [1-3]. Although the normal and tangential liquid bridge force models were proposed using a lubrication theory [4, 5], a capillary force was not considered in these force models, but a viscous force was only considered. Moreover, deformation of the liquid bridge and influence on the collision angle of the particle were not taken into account in these models. Therefore, the effect of collision angle on the dynamic liquid bridge force cannot be explained by the previous models.

As a simple experimental study, collision between a dry particle and a thin liquid film at different collision angles has been reported [6-9]. Crüger et al. [9] investigated effect of critical parameters (e.g. liquid layer thickness, viscosity, and surface tension) on the normal and tangential restitution coefficient at different collision angles. They showed that the tangential wet restitution coefficient and liquid bridge behavior was varied depending on the collision angle. However, the effect of collision angle on the microscopic particle-particle adhesion by the dynamic liquid bridge has not been experimentally analyzed due to their complexities.

In this chapter, the effect of collision angle on the particle-particle adhesion by the dynamic liquid bridge was analyzed. In particular, translational and rotational motions of the particle at different collision angles were investigated through comparison with a conventional static liquid bridge force model.
2. Simulation set-up

In this chapter, to simulate the behaviors of the liquid bridge and particles, the direct numerical simulation model described in Chapter II was used.

**Fig. 5-1** illustrates a collision angle $\alpha$ used in this study. $v_{1,\text{init}}$ and $v_{2,\text{init}}$ are the initial translational velocities of the particle-1 and particle-2, respectively. The particle-2 collided with the stationary particle-1 ($|v_{1,\text{init}}| = 0$) at various collision velocities ($|v_{2,\text{init}}|$). The point O is the center of mass of particle-2 when two particles contact. Based on this point O, collision angle $\alpha$ was defined. The collision angle was set from 0 deg (collision in the normal direction) to 90 deg (collision in the tangential direction). **Fig. 5-2** shows the initial configurations. In this study, a droplet was only set on the particle-1 to simply define the collision angle. The x-component of the initial velocity of the particle-2 was set as zero. Therefore, the two particles can only move in the y- and z-direction during simulation. The rotational velocities of the two particles were set as zero. **Table 5-1** lists calculation conditions. The physical properties of water were set as calculation conditions of the liquid. The droplet volume was determined based on a typical mist volume used in a fluidized bed spray granulation [10]. The physical properties of a lactose were set as calculation conditions of the particle. In the present study, restitution coefficient between solid particles was set as 1.0 to clearly investigate the acceleration and dissipation of the particle motion derived from the liquid bridge force. Under these conditions, Bond number was $8.47 \times 10^{-5} \ll 1$, i.e., the influence of gravity can be ignored [11].
Fig. 5-1. Definition of collision angle $\alpha$.

Fig. 5-2. Initial configurations at $\alpha = 30$ deg and $\alpha = 90$ deg.
### Table 5-1 Calculation conditions

<table>
<thead>
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<th>Parameter</th>
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<td>Particle diameter</td>
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<td>Particle density</td>
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<td>Restitution coefficient between solid particles</td>
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<td>Collision velocity</td>
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</table>
3. Results and discussion

3.1. Translational motion of particle

Fig. 5-3 shows simulation results of the particles and liquid bridge behaviors at the different collision angles. At the collision angle $\alpha = 30$ deg, the liquid bridge was formed with approaching the particle-2 (Fig. 5-3 (a) $t = 1.0$ $\mu$s), and the particle-2 collided with the particle-1 (Fig. 5-3 (a) $t = 2.8$ $\mu$s). The particle-1 was the pushed and the liquid bridge was largely elongated (Fig. 5-3 (a) $t = 10.0$ $\mu$s). Finally, the liquid bridge was ruptured (Fig. 5-3 (a) $t = 14.6$ $\mu$s). At the collision angle $\alpha = 90$ deg, the liquid bridge was formed with approaching the particle-2 (Fig. 5-3 (b) $t = 2.0$ $\mu$s). In this case, the two particles do not collide if there is no liquid droplet. However, surfaces of the two particles were collided with each other due to attractive force derived from the liquid bridge (Fig. 5-3 (b) $t = 6.2$ $\mu$s). The liquid bridge was then largely elongated (Fig. 5-3 (b) $t = 35.0$ $\mu$s) and finally ruptured (Fig. 5-3 (b) $t = 40.0$ $\mu$s). From these results, it was found that the behaviors of the particles and liquid bridge were largely different depending on the collision angle.

To further understand the particle behavior, trajectory of each particle was analyzed. The direct numerical simulation result was compared with a result calculated by a conventional static liquid bridge force model. The static liquid bridge force and rupture distance of the liquid bridge were calculated by using following equations [11]:

$$\hat{F}_{LB} = \exp(A \hat{h} + B) + C$$

(5-1)

$$A = -1.1 \hat{V}_{LB}^{-0.53}$$

(5-2)

$$B = -(0.34 \ln \hat{V}_{LB} - 0.96)\theta^2 - 0.019 \ln \hat{V}_{LB} + 0.48$$

(5-3)

$$C = 0.0042 \ln \hat{V}_{LB} + 0.078$$

(5-4)

$$\hat{h}_{\text{rap}} = (0.62 \theta + 0.99) \hat{V}_{LB}^{0.34}$$

(5-5)
Fig. 5-3. Simulation results of particles and liquid bridge behaviors at different collision angles $\alpha$ ($v = 3.0$ m/s).
where $F_{LB}, \ V_{LB}, \ h,$ and $h_{rup}$ are the dimensionless liquid bridge force ($= F_{LB}/\pi R_p \sigma$), dimensionless liquid bridge volume ($= V_{LB}/R_p^3$), dimensionless distance between particles ($= h/R_p$), and dimensionless rupture distance of the liquid bridge, respectively. In this previous model, the static liquid bridge force only acts in the normal direction, which is the direction connecting the center of gravities of two particles. In this simulation, it was assumed that the liquid bridge was formed when the separation distance between the particles’ solid surfaces became smaller than initial droplet height (5.0 µm). It was also assumed that the liquid bridge was ruptured when the separation distance between particles became larger than the critical rupture distance expressed by Eq. (5-5). Fig. 5-4 shows the particle trajectories at different collision angles. The solid lines show the direct numerical simulation results (i.e., DNS), while the dashed lines show the simulation results of the static liquid bridge force model (i.e., SLB). The circle plots mean the initial position of the particles, while the triangle plots mean particles’ positions when the liquid bridge was ruptured. In Fig. 5-4, initial position of the particle-1 was set at $(z, y) = (0, 0)$ regardless of the collision angle. At $\alpha = 30$ deg, the particle trajectories were similar between the DNS and SLB, although the moving distance of the DNS was larger than that of the SLB. At $\alpha = 90$ deg, there is a large difference between the DNS and SLB. In the SLB, the particle-2 moved to the positive z-direction after the particle-particle collision. On the other hand, in the DNS, the particle-2 moved to the negative z-direction. The moving distance of the DNS was much larger than that of the SLB. Moreover, the particle-2 in the DNS moved like drawing a circular arc.

Fig. 5-5 indicates the translational kinetic energy of the particles when the liquid bridge was ruptured. With an increase in the collision angle, the energy of the particle-1 decreased, while the energy of the particle-2 increased. This is because that the momentum of the particles in the normal direction was exchanged between the two particles at the particle-particle collision. Dependence of the translational kinetic energy on the collision angle was relatively similar between the DNS and SLB, although magnitude of the energy was slightly different because the liquid bridge force model was different. From these results, it was found that the particle trajectories of the DNS
were largely different from that of the SLB especially at the higher collision angle, although tendency of the translational kinetic energy of the particle when the liquid bridge was ruptured was relatively similar.
**Fig. 5-4.** Particle trajectories at different collision angles ($v = 3.0$ m/s). DNS means results of direct numerical simulation. SLB means results of static liquid bridge force model.

**Fig. 5-5.** Translational kinetic energy of particles when liquid bridge was ruptured as a function of collision angles ($v = 3.0$ m/s).
3.2. Rotational motion of particle

In the present investigation, the rotational motion of the particle is also important behavior. Fig. 5-6 shows the temporal change in the rotational velocity of the particle-1 at different collision angles. In the SLB, the rotational velocity steeply increased at a specific time, followed by a constant. This step-like increase was derived from the collision between the particles’ solid surfaces. With an increase in the collision angle, the rotational velocity after the collision increased. In the DNS, the rotational velocity steeply increased at the collision between the particles’ solid surfaces, followed by a monotonical increase. This monotonical increase in the rotational velocity was induced by the dynamic liquid bridge.

Fig. 5-7 indicates the rotational kinetic energy of the particle-1 when the liquid bridge was ruptured. The energy monotonically increased with an increase in the collision angle. As shown in Fig. 5-6, the rotational velocity induced by the collision between particles’ solid surfaces was almost same between the DNS and SLB. Therefore, difference in the energy between the DNS and SLB ($E_{LB}$ shown in Fig. 5-7) corresponds to the rotational kinetic energy generated by the dynamic liquid bridge. This energy difference increased with an increase in the collision angle. These results mean that the influence of the dynamic liquid bridge on the rotational motion of the particle was higher at the higher collision angle.

This rotational motion was derived from the tangential liquid bridge force associated with the tangential deformation of the liquid on the particle surface. Fig. 5-8 shows the total liquid bridge force (sum of drag force $F_d$, capillary pressure force $F_{cp}$, and surface tension force $F_{sf}$) in the tangential direction at different collision angles when the two particles were being separated. Snapshots of the liquid bridge at different collision angles are also shown in Fig. 5-9. As shown in Fig. 5-8, at the smaller collision angle, the tangential liquid bridge force was almost zero regardless of the separation distance. This is because that the liquid bridge was formed along the normal direction between the two particles as shown in Fig. 5-9 (a) and (b). On the other hand, at the higher collision angle, the tangential liquid bridge force was acting on the particles significantly (Fig. 5-8). In this case, the liquid bridge was formed along a direction
Fig. 5-6. Temporal change in rotational velocity of particle-1 at different collision angles ($v = 3.0 \text{ m/s}$).

Fig. 5-7. Rotational kinetic energy of particle-1 when liquid bridge was ruptured as a function of collision angle ($v = 3.0 \text{ m/s}$).
Fig. 5-8. Tangential liquid bridge forces acting on particles in separation stage as a function of separation distance at different collision angles (v = 3.0 m/s).

Fig. 5-9. Location of liquid bridge in separation stage at different collision angles (v = 3.0 m/s, x = 10.0 µm).
deviated from the normal direction (Fig. 5-9 (c) and (d)). This tangential deformation of the liquid bridge largely contributed to generation of the tangential liquid bridge force at the higher collision angles. In Fig. 5-8 (b), the tangential liquid bridge force at $\alpha = 90$ deg steeply decreased around $x = 6.0 \, \mu$m. This is because the shape of the liquid bridge was largely deformed around $x = 6.0 \, \mu$m with an increase in the separation distance. From these results, it was found that the tangential deformation of the dynamic liquid bridge on the particle surface plays an important role for the rotational motion of the particle especially at the higher collision angle.

In the conventional static liquid bridge force model, the rotational motion of the particle induced by the liquid bridge cannot be calculated. To further understand the difference between the DNS and SLB, rotating behavior of the particle and deformation of the liquid bridge were focused. Fig. 5-10 shows the rotating behavior of the particle-1 at $\alpha = 90$deg. The rotational motion was shown as the red line. In case of the SLB (Fig. 5-10 (b)), the liquid bridge was virtually depicted for clarity. In the DNS, the particle-1 was not rotated between the liquid bridge formation (Fig. 5-10 (a) $t = 2.0 \, \mu$s) and the collision between the particles’ solid surfaces (Fig. 5-10 (a) $t = 6.2 \, \mu$s). After the collision, the particle was rotated without the slip of the liquid bridge on the particle surface (Fig. 5-10 (a) $t = 10.0 \, \mu$s and $t = 15.0 \, \mu$s), i.e., the liquid bridge was not slipped on the particle surface in the DNS. On the other hand, in the SLB, the liquid bridge was always treated to be formed along the normal direction between the two particles, i.e., along the dashed line shown in Fig. 5-10 (b). In addition, the particles did not rotate before the collision between the particles’ solid surfaces. Therefore, the simulated location of the liquid bridge at the collision (dashed line in Fig. 5-10 (b) $t = 5.2 \, \mu$s) significantly deviated from the initial location of the liquid bridge (red line in Fig. 5-10 (b) $t = 0.4 \, \mu$s). After the collision, the particle was rotated and the angle between red line and dashed line was varied depending on the time (Fig. 5-10 (b) $t = 10.0 \, \mu$s and $t = 15.0 \, \mu$s). This means that the liquid bridge was treated to be slipped on the particle surface in the conventional static liquid bridge force model. In an experimental study on an oblique collision between a particle and thin liquid film, it was observed that the particle was rotated without slip of the liquid bridge on the particle surface [9].
Therefore, it was considered that the DNS results can be reasonable.

From these results, it was found that the slip and non-slip behaviors are the large difference between the DNS and SLB. The non-slip behavior, that leads to the tangential deformation of the liquid bridge and liquid bridge force in the tangential direction, cannot be considered in the conventional static liquid bridge force model. Therefore, the non-slip liquid bridge behavior is a key phenomenon for modeling the particle-particle adhesion by the liquid bridge especially at the oblique particle collision. In the conventional static liquid bridge force model, the liquid bridge force is modeled to act on the center of mass of the particle. However, the direct numerical simulation suggests that the liquid bridge force should be modeled as a surface force at the oblique particle collision. This conclusion provides an important insight into modeling a particulate flow with liquid bridge adhesion.
Fig. 5-10. Rotating behavior of particle-1 at $\alpha = 90$ deg ($v = 3.0$ m/s).
4. Conclusions

In this chapter, the effect of collision angle on the particle-particle adhesion of two colliding particles through a droplet on a particle surface was investigated. In particular, difference in the simulation results between the direct numerical simulation and a conventional static liquid bridge force model was focused. The translational motion of the particles was largely different between these two cases especially at the higher collision angle. The rotational motion of the particle was then analyzed. In the conventional static liquid bridge force model, the rotational motion was induced only by the collision between the particles’ solid surfaces. On the other hand, in the direct numerical simulation, the rotational motion was accelerated by the liquid bridge at the higher collision angle after the collision. This was derived from the tangential deformation of the liquid bridge. To further understand the difference in how to treat the liquid bridge between the direct numerical simulation and static liquid bridge force model, rotating behavior of the particle and deformation of the liquid bridge were focused. In the direct numerical simulation, the liquid bridge did not slip and the particle was rotated due to the non-slip liquid bridge. By contrast, in the conventional static liquid bridge force model, the liquid bridge was slipped because the liquid bridge was treated to be formed along the normal direction between particles. From these results, it was concluded that the non-slip behavior is a key phenomenon for adequately modeling the particle-particle adhesion by the dynamic liquid bridge at the oblique particle collision.
Nomenclature

\( A \) : parameter shown in Eq. (5-1) \([-\) \]
\( B \) : parameter shown in Eq. (5-1) \([-\) \]
\( C \) : parameter shown in Eq. (5-1) \([-\) \]
\( E_{LB} \) : rotational kinetic energy induced by liquid bridge \([J]\)
\( E_R \) : rotational kinetic energy when liquid bridge was ruptured \([J]\)
\( E_T \) : translational kinetic energy when liquid bridge was ruptured \([J]\)
\( F_{LB} \) : static liquid bridge force \([N]\)
\( \hat{F}_{LB} \) : dimensionless static liquid bridge force \([-\) \]
\( F_{cp} \) : capillary pressure force \([N]\)
\( F_d \) : viscous drag force \([N]\)
\( F_{sf} \) : surface tension force at solid-liquid-gas interface \([N]\)
\( h \) : surface distance between particles \([m]\)
\( \hat{h} \) : dimensionless surface distance between particles \([-\) \]
\( R_p \) : particle radius \([m]\)
\( V_{LB} \) : liquid bridge volume \([m^3]\)
\( \hat{V}_{LB} \) : dimensionless liquid bridge volume \([m^3]\)
\( v \) : collision velocity \([m/s]\)
\( v_{init} \) : initial translational velocity of particle \([m/s]\)
\( v_{re} \) : rebound velocity of particle \([m/s]\)
\( x \) : separation distance between particles \([m]\)

Greek letters

\( \alpha \) : collision angle \([\text{deg}]\)
\( \sigma \) : surface tension coefficient \([\text{N/m}]\)
\( \theta \) : static contact angle \([\text{deg}]\)
\( \omega_p \) : rotational velocity of particle \([\text{rad/s}]\)
References


Chapter VI

Overall conclusions
This thesis has been devoted to a numerical modeling of a microscopic particle-particle adhesion by dynamic liquid bridge and elucidation of mechanism of the particle adhesion phenomenon. In this thesis, the pendular liquid bridge between two particles was focused. The main objective of this thesis were (i) development of a direct numerical simulation model for a particle-particle adhesion by a dynamic liquid bridge, (ii) elucidation of effect of particle wettability on the particle adhesion phenomenon and its physical mechanism, (iii) elucidation of effect of liquid droplet size under constant liquid volume on the particle adhesion phenomenon, and (iv) elucidation of effect of collision angle on the particle adhesion phenomenon through comparison with a conventional static liquid bridge force model. The main results and obtained findings are summarized in below:

In Chapter II, a direct numerical simulation model for a particle-particle adhesion by a dynamic liquid bridge was developed. Motions of gas and liquid were solved by using a computational fluid dynamics (CFD) approach with a constrained interpolation profile (CIP) method. Particle motion was simulated by a Lagrangian approach considering liquid bridge force that is unsteadily changed due to deformation of the liquid bridge. To verify the simulation results, verifications and validations were conducted. It was confirmed that the simulation model accurately calculated (i) surface tension force acting on a liquid, (ii) static liquid bridge force between spherical particles, and (iii) rupture behavior of liquid bridge with free falling of a particle. From these results, validity of the simulation model was confirmed. The adhesion of two colliding particles through a droplet was then simulated under various relative collision velocities. The dynamic liquid bridge force acting on a particle and wet restitution coefficient were analyzed. From the simulation results, the critical velocity for the particle adhesion, i.e., adhesiveness of the colliding particles, was determined. This critical velocity was compared to the one estimated from an analytical model. It was found that there is a big difference between the critical velocity obtained by the present simulation and the one obtained by the previous analytical model.
In Chapter III, the effect of particle wettability on the particle-particle adhesion of two colliding particles through a liquid droplet was analyzed using the direct numerical simulation developed in Chapter II. In particular, the effect of contact angle on a critical velocity for particle adhesion was investigated. As a result, it was found that the critical velocity non-monotonically changed with the contact angle. With an increase in the contact angle, the critical velocity increased and reached a local maximum value at a specific contact angle, followed by a decrease. To understand this result, the dissipation energies of the particle motion in the approach and separation stages were analyzed. The dissipation energy in the approach stage monotonically increased with an increase in the contact angle. However, the dissipation energy in the separation stage showed non-monotonic dependence on the contact angle. This was derived from the non-monotonic dependence of the maximum elongation length of the liquid bridge on the contact angle. The maximum elongation length was determined by the shape of the liquid bridge depending on the contact angle. It was concluded that a combined effect of the instantaneous liquid bridge force and dynamics of the liquid bridge deformation results in the non-monotonic dependence of the critical velocity for the particle adhesion on the particle wettability.

In Chapter IV, the effect of droplet size on the particle-particle adhesion of two colliding particles through a liquid droplet was analyzed using the direct numerical simulation developed in Chapter II. In particular, the effect of droplet size under constant liquid volume on a critical velocity for the particle adhesion was investigated. In this case, multiple liquid bridges were formed between particles, and these liquid bridges were coalesced during the approach and separation of the particles. It was found that the critical velocity increased with a decrease in the droplet size. In an initial stage of the granule growth in a fluidized bed spray granulation, the tendency of the simulation result was consistent with the experimental result. To understand this result, the dissipation energy of the particle motion was analyzed. The dissipation energy in the approach stage increased with a decrease in the droplet size. This was due to the higher capillary pressure force. On the other hand, the dissipation energy in the separation
stage decreased with a decrease in the droplet size. This was derived from difference in shapes of the liquid bridge at the particle-particle collision. From these findings, the dissipation energy of the particle motion was determined by the balance between dissipation energies in the approach and separation stages. It was concluded that magnitude of the capillary pressure force and shapes of the liquid bridge are key factors for the particle-particle adhesion by the dynamic liquid bridge with the different sizes of liquid droplets.

In Chapter V, the effect of collision angle on the particle-particle adhesion of two colliding particles through a liquid droplet was analyzed using the direct numerical simulation developed in Chapter II. In particular, difference in the simulation results between the direct numerical simulation and a conventional static liquid bridge force model was investigated. Translational and rotational motions of the particles were analyzed. As a result, it was found that both translational and rotational motions were largely different between simulation results of the direct numerical simulation and static liquid bridge force model, especially at the oblique particle collision. To further understand the difference between the direct numerical simulation and static liquid bridge force model, rotating behavior of the particle and deformation of the liquid bridge were investigated. In the direct numerical simulation, the liquid bridge did not slip and the particle was rotated due to the non-slip liquid bridge. On the other hand, in the conventional static liquid bridge force model, the liquid bridge was slipped because the liquid bridge was treated to be formed along the normal direction between particles. From these results, it was concluded that the non-slip behavior is a key phenomenon for adequately modeling the particle-particle adhesion by the dynamic liquid bridge at the oblique particle collision.

In this thesis, a particle-particle adhesion by the dynamic liquid bridge at a primary particle scale was focused, and the effects of critical parameters on the particle-particle adhesion were individually analyzed to understand the role of each parameter. As an application of the simulation model developed in this thesis, a simulation of wet
granulation including multiple particles (e.g. several thousands to several millions of particles) is conceivable. However, the calculation cost of the direct numerical simulation including unsteady deformation of the liquid bridge is too high to directly expand the direct numerical simulation model developed in this thesis to the multiple particles simulation. Therefore, alternative approach will be required. Development of a mathematical formulation of the critical velocity for the particle adhesion can be a potential approach. By using a formulated critical velocity as a boundary condition at the particle-particle collision, it is considered that adhesion/separation phenomenon by the dynamic liquid bridge can be taken into account in a simulation of wet granulation including multiple particles. This will make a progress in developing a new modeling of granulation that can directly predict physical properties of granules (e.g. granule size distribution and structure) without empirical models and parameters.

The author hopes that this thesis can contribute to the better understanding for the mechanism of particle adhesion by the dynamic liquid bridge and new directions in a next-generation wet granulation technology.
Appendix
A: Constrained interpolation profile (CIP) method

Solution of three-dimensional advection equation for color function shown in Eq. (2-3) is illustrated. Eq. (2-3) is rewritten using $u = (u, v, w)$ as follows:

$$\frac{\partial \phi}{\partial t} + u \frac{\partial \phi}{\partial x} + v \frac{\partial \phi}{\partial y} + w \frac{\partial \phi}{\partial z} = 0$$  \hspace{1cm} (A-1)

The color function at three-dimensional grid is interpolated as following equation [A1]:

$$\phi(x, y, z) = [(B_{1i,j,k} X + B_{2i,j,k} Y + B_{3i,j,k} Z + B_{4i,j,k} ) X + B_{5i,j,k} Y + \partial_x \phi_{i,j,k} ] X$$
$$+ [(B_{6i,j,k} Y + B_{7i,j,k} Z + B_{8i,j,k} X + B_{9i,j,k} ) Y + B_{10i,j,k} Z + \partial_y \phi_{i,j,k} ] Y$$
$$+ [(B_{11i,j,k} Z + B_{12i,j,k} X + B_{13i,j,k} Y + B_{14i,j,k} ) Z + B_{15i,j,k} X + \partial_z \phi_{i,j,k} ] Z$$
$$+ B_{16i,j,k} XYZ + \phi_{i,j,k}$$  \hspace{1cm} (A-2)

where $X = x - x_{i,j,k}$, $Y = y - y_{i,j,k}$, $Z = z - z_{i,j,k}$. Coefficients $B_1$ to $B_{16}$ are given as follows:

$$B_{1i,j,k} = [-2 D_i + \partial_x (\phi_{i+1,j,k} + \phi_{i,j,k})] \Delta x / \Delta x^3$$  \hspace{1cm} (A-3)

$$B_{2i,j,k} = -[B_{17i,j,k} + \partial_x D_j \Delta x] / \Delta x^2 \Delta y$$  \hspace{1cm} (A-4)

$$B_{3i,j,k} = -[B_{18i,j,k} + \partial_x D_k \Delta x] / \Delta x^2 \Delta z$$  \hspace{1cm} (A-5)

$$B_{4i,j,k} = [3 D_i - \partial_x (\phi_{i+1,j,k} + 2\phi_{i,j,k})] \Delta x / \Delta x^2$$  \hspace{1cm} (A-6)

$$B_{5i,j,k} = [B_{17i,j,k} + \partial_x D_j \Delta x + \partial_y D_i \Delta y] / \Delta x \Delta y$$  \hspace{1cm} (A-7)

$$B_{6i,j,k} = [-2 D_j + \partial_y (\phi_{i,j+1,k} + \phi_{i,j,k})] \Delta y / \Delta y^3$$  \hspace{1cm} (A-8)

$$B_{7i,j,k} = -[B_{19i,j,k} + \partial_y D_k \Delta y] / \Delta y^2 \Delta z$$  \hspace{1cm} (A-9)
\[ B_{8, i,j,k} = - [B_{17, i,j,k} + \partial_y D_i \Delta y] / \Delta x \Delta y^2 \] (A-10)

\[ B_{9, i,j,k} = [3 D_j - \partial_y (\phi_{i,j+1,k} + 2\phi_{i,j,k}) \Delta y] / \Delta y^2 \] (A-11)

\[ B_{10, i,j,k} = [B_{19, i,j,k} + \partial_y D_k \Delta y + \partial_z D_j \Delta z] / \Delta y \Delta z \] (A-12)

\[ B_{11, i,j,k} = [-2 D_k + \partial_z (\phi_{i,j,k+1} + \phi_{i,j,k}) \Delta z] / \Delta z^3 \] (A-13)

\[ B_{12, i,j,k} = - [B_{18, i,j,k} + \partial_z D_i \Delta z] / \Delta x \Delta z^2 \] (A-14)

\[ B_{13, i,j,k} = - [B_{19, i,j,k} + \partial_z D_j \Delta z] / \Delta y \Delta z^2 \] (A-15)

\[ B_{14, i,j,k} = [3 D_k - \partial_z (\phi_{i,j,k+1} + 2\phi_{i,j,k}) \Delta z] / \Delta z^2 \] (A-16)

\[ B_{15, i,j,k} = [B_{18, i,j,k} + \partial_z D_i \Delta z + \partial_x D_k \Delta x] / \Delta x \Delta z \] (A-17)

\[ B_{16, i,j,k} = [B_{17, i,j,k} + \phi_{i,j,k+1} - (\phi_{i,j+1,k} + \phi_{i+1,j,k+1}) + \phi_{i+1,j+1,k+1}] / \Delta x \Delta y \Delta z \] (A-18)

where \( D_i = \phi_{i+1,j,k} - \phi_{i,j,k} \), \( D_j = \phi_{i,j+1,k} - \phi_{i,j,k} \), \( D_k = \phi_{i,j,k+1} - \phi_{i,j,k} \). Coefficients \( B_{17} \) to \( B_{19} \) are given as follows:

\[ B_{17, i,j,k} = - \phi_{i,j,k} + \phi_{i+1,j,k} + \phi_{i,j+1,k} - \phi_{i+1,j+1,k} \] (A-19)

\[ B_{18, i,j,k} = - \phi_{i,j,k} + \phi_{i+1,j,k} + \phi_{i,j,k+1} - \phi_{i+1,j,k+1} \] (A-20)

\[ B_{19, i,j,k} = - \phi_{i,j,k} + \phi_{i,j+1,k} + \phi_{i,j,k+1} - \phi_{i,j+1,k+1} \] (A-21)

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Reference

B: Definition point of physical quantity in computational fluid dynamics (CFD)

A fixed Cartesian grid is used with staggered layout as shown in Fig. B-1 (for $x$-$y$ direction). In the staggered layout, scalar quantity is defined at the cell center, while vector quantity is defined at the cell face.

![Fig. B-1. Schematic of staggered layout.](image-url)
C: Calculation procedure and discretization of governing equations

1) Advection equations (Eqs. (C-1) and (C-2)) were solved by a CIP method, and fluid velocity \( u^A = (u^A, v^A, w^A) \) and \( \phi^{n+1} \) were obtained.

\[
\frac{\partial u}{\partial t} + (u \cdot \nabla) u = 0 \quad \text{(C-1)}
\]

\[
\frac{\partial \phi}{\partial t} + (u \cdot \nabla) \phi = 0 \quad \text{(C-2)}
\]

2) Navier-Stokes equation (Eq. (C-3)), excepting for pressure and interaction force from particle to fluid terms, was discretized by a central differential scheme and fluid velocity \( u^B = (u^B, v^B, w^B) \) was obtained as follows:

\[
\frac{\partial u}{\partial t} = \frac{1}{\rho_t} \nabla \cdot \mu_t \left[ \nabla u + (\nabla u)^T \right] + f_{sf} + g \quad \text{(C-3)}
\]

\[
u^B_{i,j,k} = \nu^A_{i,j,k} + \left[ 0.5(\mu_{t,i,j,k+1} + \mu_{t,i,j,k}) \right] \times \left\{ \frac{u^{A}_{i+1,j,k} - u^{A}_{i,j,k} - u^{A}_{i,j,k+1} + u^{A}_{i,j,k-1}}{(\Delta x)^2} \right. \\
+ \left. \frac{u^{A}_{i,j+1,k} - u^{A}_{i,j,k} - u^{A}_{i,j,k+1} + u^{A}_{i,j,k-1}}{(\Delta y)^2} \right. \\
+ \left. \frac{u^{A}_{i,j,k+1} - u^{A}_{i,j,k} - u^{A}_{i,j,k+1} + u^{A}_{i,j,k-1}}{(\Delta z)^2} \right\} + f_{sf,x,i,j,k} + g_x] \Delta t \quad \text{(C-4)}
\]

\[
u^B_{i,j,k} = \nu^A_{i,j,k} + \left[ 0.5(\mu_{t,i,j,k+1} + \mu_{t,i,j,k}) \right] \times \left\{ \frac{v^{A}_{i+1,j,k} - v^{A}_{i,j,k} - v^{A}_{i,j,k+1} + v^{A}_{i,j,k-1}}{(\Delta x)^2} \right. \\
+ \left. \frac{v^{A}_{i,j+1,k} - v^{A}_{i,j,k} - v^{A}_{i,j,k+1} + v^{A}_{i,j,k-1}}{(\Delta y)^2} \right. \\
+ \left. \frac{v^{A}_{i,j,k+1} - v^{A}_{i,j,k} - v^{A}_{i,j,k+1} + v^{A}_{i,j,k-1}}{(\Delta z)^2} \right\} + f_{sf,y,i,j,k} + g_y] \Delta t \quad \text{(C-5)}
\]
The surface tension force \( f_{sf} \) was given as following equations:

\[
\begin{align*}
    f_{sf, x,i,j,k} &= \frac{2 \sigma}{\rho_{t,i+1,j,k} + \rho_{t,i,j,k}} \frac{\kappa_{i+1,j,k} + \kappa_{i,j,k}}{2} \phi_{n+1,i+1,j,k} - \phi_{n+1,i,j,k} \Delta x \\
    f_{sf, y,i,j,k} &= \frac{2 \sigma}{\rho_{t,i,j+1,k} + \rho_{t,i,j,k}} \frac{\kappa_{i,j+1,k} + \kappa_{i,j,k}}{2} \phi_{n+1,i,j+1,k} - \phi_{n+1,i,j,k} \Delta y \\
    f_{sf, z,i,j,k} &= \frac{2 \sigma}{\rho_{t,i,j,k+1} + \rho_{t,i,j,k}} \frac{\kappa_{i,j,k+1} + \kappa_{i,j,k}}{2} \phi_{n+1,i,j,k+1} - \phi_{n+1,i,j,k} \Delta z \\
    \kappa_{i,j,k} &= \left( \frac{n_{lgx, x,i,j,k} - n_{lgx, x,i-1,j,k}}{\Delta x} + \frac{n_{lgx, y,i,j,k} - n_{lgx, y,i,j-1,k}}{\Delta y} + \frac{n_{lgx, z,i,j,k} - n_{lgx, z,i,j,k-1}}{\Delta z} \right) \\
    n_{lgx, x,i,j,k} &= \frac{n_{lgx, x,i,j,k}^{*}}{n_{lgx, x,i,j,k}} \\
    n_{lgx, y,i,j,k} &= \frac{n_{lgx, y,i,j,k}^{*}}{n_{lgx, y,i,j,k}} \\
    n_{lgx, z,i,j,k} &= \frac{n_{lgx, z,i,j,k}^{*}}{n_{lgx, z,i,j,k}}
\end{align*}
\]
\[ n_{lgx_{x,i,j,k}}^* = \frac{\phi_{sm,i+1,j,k} - \phi_{sm,i,j,k}}{\Delta x} \]  \hspace{1cm} (C-14)

\[ n_{lgx_{y,i,j,k}}^* = \frac{\phi_{sm,i,j+1,k} - \phi_{sm,i,j,k}}{\Delta y} \]  \hspace{1cm} (C-15)

\[ n_{lgx_{z,i,j,k}}^* = \frac{\phi_{sm,i,j,k+1} - \phi_{sm,i,j,k}}{\Delta z} \]  \hspace{1cm} (C-16)

\[ n_{lgx_{y,i,j,k}}^* = \frac{\frac{n_{lgx_{y,i,j,k}}^* + n_{lgx_{y,i+1,j,k}}^* + n_{lgx_{y,i,j-1,k}}^* + n_{lgx_{y,i+1,j-1,k}}^*}{4}}{4} \]  \hspace{1cm} (C-17)

\[ n_{lgx_{z,i,j,k}}^* = \frac{\frac{n_{lgx_{z,i,j,k}}^* + n_{lgx_{z,i+1,j,k}}^* + n_{lgx_{z,i,j-1,k}}^* + n_{lgx_{z,i+1,j-1,k}}^*}{4}}{4} \]  \hspace{1cm} (C-18)

\[ n_{lgx_{x,i,j,k}}^* = \frac{\frac{n_{lgx_{x,i,j,k}}^* + n_{lgx_{x,i+1,j,k}}^* + n_{lgx_{x,i,j-1,k}}^* + n_{lgx_{x,i+1,j-1,k}}^*}{4}}{4} \]  \hspace{1cm} (C-19)

\[ n_{lgx_{z,i,j,k}}^* = \frac{\frac{n_{lgx_{z,i,j,k}}^* + n_{lgx_{z,i+1,j,k}}^* + n_{lgx_{z,i,j-1,k}}^* + n_{lgx_{z,i+1,j-1,k}}^*}{4}}{4} \]  \hspace{1cm} (C-20)

\[ n_{lgx_{x,i,j,k}}^* = \frac{\frac{n_{lgx_{x,i,j,k}}^* + n_{lgx_{x,i,j+1,k}}^* + n_{lgx_{x,i,j-1,k}}^* + n_{lgx_{x,i+1,j-1,k}}^*}{4}}{4} \]  \hspace{1cm} (C-21)

\[ n_{lgx_{y,i,j,k}}^* = \frac{\frac{n_{lgx_{y,i,j,k}}^* + n_{lgx_{y,i,j+1,k}}^* + n_{lgx_{y,i,j-1,k}}^* + n_{lgx_{y,i+1,j-1,k}}^*}{4}}{4} \]  \hspace{1cm} (C-22)
3) The pressure Poisson equation (Eq. (C-23)) was solved by a bi-conjugate gradient stabilized (Bi-CGSTAB) method and fluid pressure at new time ($p^{n+1}$) was obtained.

$$\frac{\nabla \cdot \mathbf{u}}{\Delta t} = \nabla \left( \frac{1}{\rho_f} \nabla p \right)$$  \hspace{1cm} (C-23)

Calculation procedure of Bi-CGSTAB method was described below:

i. Initial vector $x_0$ was set and $r_0$, $p_0$, and $s_0$ was obtained as following equations:

$$r_0 = b - Ax_0$$ \hspace{1cm} (C-24)

$$p_0 = r_0$$ \hspace{1cm} (C-25)

$$s_0 = r_0$$ \hspace{1cm} (C-26)

where $A$ is non-singular matrix.

ii. Following calculations were repeated for $k = 0, 1, 2, \cdots$:

$$\alpha_k = \frac{(s \cdot r_k)}{(s \cdot A p_k)}$$ \hspace{1cm} (C-27)

$$t_k = r_k - \alpha_k A p_k$$ \hspace{1cm} (C-28)

$$\xi_k = \frac{(A t_k \cdot t_k)}{(A t_k \cdot A t_k)}$$ \hspace{1cm} (C-29)

$$x_{k+1} = x_k - \alpha_k p_k + \xi_k t_k$$ \hspace{1cm} (C-30)

$$r_{k+1} = t_k - \xi_k A t_k$$ \hspace{1cm} (C-31)
If a convergence condition was satisfied, the calculation was end. If not, the calculation was continued using $\beta_k$ and $p_{k+1}$.

$$
\beta_k = \frac{\alpha_k (s \cdot r_{k+1})}{\xi_k (s \cdot r_k)} \quad (C-32)
$$

$$
p_{k+1} = r_{k+1} + \beta_k (p_k - \xi_k A p_k) \quad (C-33)
$$

In this study, $x_{k+1}$ corresponds to $p^{n+1}_{i,j,k}$, and $x_k$ corresponds to $p^n_{i,j,k}$. $b$ was obtained as follows:

$$
b_{i,j,k} = \frac{1}{\Delta t} \left( \frac{u^B_{i,j,k} - u^B_{i-1,j,k}}{\Delta x} + \frac{v^B_{i,j,k} - v^B_{i,j-1,k}}{\Delta y} + \frac{w^B_{i,j,k} - w^B_{i,j-1,k}}{\Delta z} \right) \quad (C-34)
$$

Eq. (C-35) was solved using fluid pressure ($p^{n+1}$) and fluid velocity ($u^C = (u^C, v^C, w^C)$) was obtained as follows:

$$
\frac{\partial u}{\partial t} = -\frac{1}{\rho_f} \nabla p \quad (C-35)
$$

$$
u^C_{i,j,k} = u^B_{i,j,k} - \frac{2\Delta t}{\rho_{l,i,j,k+1} + \rho_{t,i,j,k}} \left( \frac{p^{n+1}_{i+1,j,k} - p^{n+1}_{i,j,k}}{\Delta x} \right) \quad (C-36)
$$

$$
v^C_{i,j,k} = v^B_{i,j,k} - \frac{2\Delta t}{\rho_{l,i,j,k+1} + \rho_{t,i,j,k}} \left( \frac{p^{n+1}_{i,j+1,k} - p^{n+1}_{i,j,k}}{\Delta y} \right) \quad (C-37)
$$

$$
w^C_{i,j,k} = w^B_{i,j,k} - \frac{2\Delta t}{\rho_{l,i,j,k+1} + \rho_{t,i,j,k}} \left( \frac{p^{n+1}_{i,j,k+1} - p^{n+1}_{i,j,k}}{\Delta z} \right) \quad (C-38)
$$
4) Fluid velocity at a next time step \( \mathbf{u}^{n+1} = (u^{n+1}, v^{n+1}, w^{n+1}) \) was obtained by solving Eq. (C-39) as follows:

\[
\frac{\partial \mathbf{u}}{\partial t} = f_p
\]  

\( (C-39) \)

\[
u^{n+1}_{i,j,k} = \nu^C_{i,j,k} + f_{p_x,i,j,k} \Delta t
\]  

\( (C-40) \)

\[
u^{n+1}_{i,j,k} = \nu^C_{i,j,k} + f_{p_y,i,j,k} \Delta t
\]  

\( (C-41) \)

\[
u^{n+1}_{i,j,k} = \nu^C_{i,j,k} + f_{p_z,i,j,k} \Delta t
\]  

\( (C-42) \)

The interaction force from particle to fluid \( (f_p) \) was given as following equations:

\[
f_{p_x,i,j,k} = \frac{\phi_{s,i+1,j,k} + \phi_{s,i,j,k}}{2} \left( u^p_{i,j,k} - u^C_{i,j,k} \right)
\]  

\( (C-43) \)

\[
f_{p_y,i,j,k} = \frac{\phi_{s,i+1,k} + \phi_{s,i,j,k}}{2} \left( v^p_{i,j,k} - v^C_{i,j,k} \right)
\]  

\( (C-44) \)

\[
f_{p_z,i,j,k} = \frac{\phi_{s,i,j,k+1} + \phi_{s,i,j,k}}{2} \left( w^p_{i,j,k} - w^C_{i,j,k} \right)
\]  

\( (C-45) \)
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