Adhesional Contact between Rigid Sphere and Elastic Plane Covered with Thin Liquid Film Considering Contact-Angle Hysteresis

Yoji Iguchi1), Dooyoung Baek2), Satoshi Momozono3), Pasomphone Hemthavy4), Shigeki Saito5) and Kunio Takahashi4)

1) Department of International Development Engineering, Tokyo Institute of Technology, 2-12-1 I4-11, O-okayama, Meguro-ku, Tokyo 152-8552, Japan
2) Lab of Adhesion and Bio-Composites, Research Institute of Agriculture and Life Sciences, Seoul National University, 200-1046A, Seoul National University, I Gwanak-ro, Gwanak-ku, Seoul 08826, Korea
3) School of Engineering, Tokyo Institute of Technology, 2-12-1 I6-14, O-okayama, Meguro-ku, Tokyo 152-8552, Japan
4) School of Environment and Society, Tokyo Institute of Technology, 2-12-1 I4-11, O-okayama, Meguro-ku, Tokyo 152-8552, Japan
5) School of Environment and Society, Tokyo Institute of Technology, 2-12-1 I1-47, O-okayama, Meguro-ku, Tokyo 152-8552, Japan

*Corresponding author: Yoji Iguchi (iguchi.y.ab@m.titech.ac.jp)

Abstract

This paper proposes a contact model between a rigid sphere and an elastic plane covered with a thin liquid film. The elastic contact is determined by Johnson–Kendall–Roberts theory, which deals with the energy equilibrium of elastic and interfacial energies. In elastic contact under presence of thin liquid film, the energy needed to separate the two bodies is defined as the summation of the interfacial energy among the sphere, liquid, and plane. Capillary force affects the two bodies because a liquid bridge is formed between them. Shape of the liquid bridge, assumed by the Clark’s toroidal approximation, determines the capillary force. The hysteresis of the liquid contact angles and that of the liquid volume between loading and unloading processes are considered. In loading process, the liquid film is squeezed out from the contact area and accumulates at the contact edge with increasing the liquid contact angles. In unloading process, the accumulated liquid is dragged to the contact edge, and the contact angles decrease. An irreversible force curve is obtained from these two hystereses, and the effect of liquid bridge is discussed based on the calculated results. In addition, the adhesion-hysteresis mechanisms caused by the capillary force are discussed.

Keywords

adhesion contact, analytical model, capillary force, liquid contact angle, sphere, elastic half space

1 Introduction

The elastic-contact problems between two spherical bodies were first studied by Hertz (Hertz theory) [1]. In addition to the Hertz theory, Johnson, Kendall, and Roberts considered the adhesion force between the two bodies from the viewpoint of energy equilibrium of the elastic and interfacial energy [Johnson–Kendall–Roberts (JKR theory)] [2, 3]. According to the JKR theory, the amount of energy required to separate a unit area of two contact bodies is defined as an adhesion process. Derjaguin, Muller, and Toporov considered the elastic deformation calculated by the Hertz theory and the long-range force generated by intermolecular potential [Derjaguin–Muller–Toporov (DMT theory)] [4]. Both the JKR and DMT theories have different availability conditions, which are consolidated as an “adhesion map” by Johnson and Greenwood [5]. Maugis assumed the gap between the two bodies as a crack and explained the transition from the JKR to the DMT theory [6]. However, the Maugis model cannot describe the adhesion hysteresis, which represents the difference of the force curve between the loading and unloading processes in an actual contact [7–10]. The origin of this hysteresis was explained as a viscoelastic effect in some reports [7, 11, 12], as the effect of surface roughness [13, 14], as the hysteresis of the work of adhesion between the loading and unloading processes [15, 16], and also as Van der Waals interactions [17, 18].

The presence of a thin liquid film between two bodies is said to form a liquid bridge and significantly affects the adhesion. The effect of liquid bridge, i.e., capillary force, on two rigid axisymmetric bodies have been studied using experimental and theoretical approaches [19–21]. Orr and Scriven solved the Young–Laplace equation of the liquid bridge between two bodies using a numerical solution [22]. McFarlane and Tabor studied the capillary force between a rigid sphere and a rigid
plane with a liquid film [23]. They obtained a very simple and significant approximation, described as:

\[ F_{MT} = 4\pi\gamma_{\text{liquid}} R \theta, \]

where \( \gamma_{\text{liquid}} \) is the surface tension of the liquid, \( R \) is the sphere radius, and \( \theta \) is the contact angle of the liquid. The contact angles were often assumed to be \( \theta = 0 \) (perfect wetting), and the capillary force was approximated as [24, 25]

\[ F_{MT} = 4\pi\gamma_{\text{liquid}} R. \]  (1)

Fogden and White studied an elastic contact with a liquid film considering the Hertz theory and the effect of the liquid bridge on the elastic contact [26]. However, the adhesion force was neglected in this model, and the adhesion process and the gap geometry between the two bodies were not considered.

Some papers suggest that the adhesion force is determined not only by the JKR or DMT theory but also by the theory of the liquid-bridge effect [24, 27–32]. A liquid bridge is categorized into two patterns: one is the capillary condensation, and the other is the liquid-film distribution on the surface. When the liquid film on the surface is infinitesimally thin such as a few layers, it does not act similar to a fluid because of the surface forces and structural disjoining pressure [33–35]. If the liquid film can move during the contact process, it can be squeezed out of the contact area to form a liquid bridge at the contact edge. The liquid bridge moves with the contact edge. As the edge of the liquid bridge advances or recedes, its contact angles become respectively larger or smaller than a stable angle, i.e., the contact angle hysteresis [36–42]. When the contact area increases, the squeezed liquid film enlarges the liquid bridge. Thus, the capillary force between the two bodies varies.

The present study deals with the elastic contact between a rigid sphere and an elastic plane with a liquid film. During contact, a liquid bridge is assumed to be formed at the contact edge, and the shape of the liquid bridge is determined by the elastic deformation. The elastic deformation and the gap between the two bodies obey the JKR theory, as described in Section 2. The shape and volume of the liquid bridge is calculated using the gap geometry, as described in Section 3. The equations of the deformation and the forces can be calculated using the gap geometry, as described in Section 3. The equations of the deformation and the forces can be normalized by the sphere radius and the approximated capillary force [Eq. (1)]. This scalable model is described in Section 4. The contact angles and volume of the liquid bridge experience hysteresis between the loading and unloading processes, which are explained in Section 5. The calculated results and the mechanism of the adhesion hysteresis are discussed in Section 6.

\[ \Delta \gamma_{\text{LT}} = \gamma_{\text{LT}} + \gamma_{\text{LF}} - \gamma_{\text{PV}}, \]  (2)

where \( \gamma_{\text{LT}}, \gamma_{\text{LF}}, \) and \( \gamma_{\text{PV}} \) are the interfacial free energy between a sphere and a liquid, a liquid and a plane, and a plane and a solid. The second, the elastic deformation is only caused by the elastic contact described by the JKR theory [2], and the elastic deformation caused by the Laplace pressure is assumed to be much smaller. In the JKR theory, the system satisfies the energy equilibrium of the total energy, which is described by the sum of stored elastic energy and differential interfacial energy. This relationship is satisfied under static condition. Therefore, our model can experimentally apply to the quasi-static condition because our model uses same assumptions as the JKR theory. The elastic deformation of the elastic plane was solved by Johnson [3, 7],

\[ z_{\text{plane}}(r) = \begin{cases} \frac{1}{6\Gamma} (2a^2 - r^2) \sin \frac{d}{r} + \frac{a}{r} \sqrt{r^2 - a^2} + \frac{a}{r} \sin^{-1} \frac{a}{r}, & \text{when } r < a \\ \frac{1}{12\Gamma} \left( \frac{8a^2 R}{\pi E^*} \sin \frac{d}{r} + \frac{a}{r} \sqrt{r^2 - a^2} \right) + \frac{a}{r} \sin^{-1} \frac{a}{r}, & \text{when } r \geq a \end{cases}. \]  (3)

where \( z_{\text{plane}}(r) \) is the geometric function of the rigid sphere, \( r \) is the horizontal displacement from the center of the contact area, \( R \) is the radius of the rigid sphere, \( d \) is the displacement of the bottom of the rigid sphere from the plane surface, and \( a \) is the contact radius. \( E^* \) is the effective elastic modulus, and \( \Gamma \) is the non-dimensional adhesion parameter defined by \( \Delta \gamma_{\text{LT}}, E^* \), and \( R \). \( z_{\text{plane}}(r), \Gamma, \) and \( \delta \) are defined as follows:

\[ z_{\text{plane}}(r) = \frac{r^2 - \delta}{2R} - \delta, \Gamma = \frac{8a^2}{\pi E^* R}, \text{ and } \delta = \frac{a^2}{R} - \frac{\pi \Gamma}{2} \sqrt{\frac{a}{\tau}}. \]  (4)

Here, when \( r \) is small enough by comparison to the sphere radius \( r \ll R \), \( z_{\text{plane}}(r) \) can be assumed as a parabolic surface [2]. Therefore, during the contact between the sphere and the plane, we need to consider only a small amount of deformation and a small displacement from the center of the contact area. This problem does not occur during the contact between a parabolic surface and a plane. In the range \( r \geq a \), the gap distribution between the rigid sphere and elastic plane can be obtained as the sum of two parts:

\[ d(t) = z_{\text{plane}}(r) - z_{\text{liquid}}(r) = \frac{2a^2 - r^2}{\pi R} \cos^{-1} \left( \frac{a}{r} \right) \times \frac{a}{r} \sqrt{r^2 - a^2} \]

\[ + \frac{1}{2} \sqrt{r^2 - a^2} \cos^{-1} \left( \frac{a}{r} \right). \]  (5)
The first term \( d_{\text{Hertz}}(r) \) is calculated by the Hertz theory, and the second term \( d_{\text{contact}}(r) \) is calculated by the Boussinesq theory [3, 7]. The derivative of \( d_{\text{contact}}(r) \) is obtained as

\[
\frac{\partial d_{\text{contact}}}{\partial r}(r) = \frac{2r}{\sqrt{\pi}} \cos\left(\frac{\sqrt{r^2 - a^2}}{\sqrt{\pi}}\right) + \frac{2a}{\sqrt{\pi}r} \sqrt{r^2 - a^2} \Rightarrow \frac{\partial d_{\text{contact}}}{\partial r}(a) = 0. \tag{6}
\]

Here, from Eq. (5), the gap distribution near the contact edge \((r = a)\) can be approximated as

\[
d(t) = d_{\text{contact}}(r) = F_{\text{H}} \sqrt{\pi} \cos\left(\frac{t}{\sqrt{\pi}}\right).
\tag{7}
\]

Its error is expressed as

\[
\text{(error)} = \frac{\partial d_{\text{contact}}(r)}{d(t)} \times 100 \text{ [%]}
\tag{8}
\]

Equation (7) shows good conformity in the range of small \( r \) and large \( \Gamma \).

Next, let's consider the elastic force between two bodies. Pressure distribution \( p(r) \) in the contact area is described by the sum of the Hertz and Boussinesq pressure distributions [3], which are similar to the gap distribution.

\[
p(r) = \frac{2Ea}{\pi R} \left(1 - \frac{r^2}{a^2}\right)^{1/2} + \frac{E}{2} \frac{\alpha}{N} \left(1 - \frac{r^2}{a^2}\right)^{1/2}
\tag{9}
\]

Elastic force \( F_{\text{elas}} \) between the two bodies can be calculated by integrating the pressure distribution over the contact area.

\[
F_{\text{elas}} = \int_0^\pi p(r) \cdot 2\pi dr = \frac{4Ea}{3R} - \frac{\pi E}{\Gamma} \sqrt{\pi} R.
\tag{10}
\]

According to the JKR theory, the elastic force varies with the loading and unloading processes in the range \( \delta < 0 \). The contact begins at \( \delta = 0 \), but the detachment does not begin at the same displacement. The detachment between the two bodies begins at the point of elastic force \( F_\delta \), displacement \( \delta_c \), and contact radius \( a\). \( F_c \), \( \delta_c \), and \( a \) are respectively expressed as follows:

\[
F_c = \frac{5}{48} \pi^2 E \Gamma R^2, \quad a_c = \left(\frac{3}{8} \frac{1}{\Gamma} \right) R \quad \text{and} \quad \delta_c = -\frac{3a^2}{R}.
\tag{11}
\]

In the contact between two elastic spheres with radii, elastic constants, and Poisson’s ratios of \( R_1 \) and \( R_2 \), \( E_1 \) and \( E_2 \), and \( \nu_1 \) and \( \nu_2 \), respectively, the contact between the two elastic spheres can replace the contact between a rigid sphere and an elastic plane using the following expressions:

\[
\frac{1}{E_1} - \frac{1-v_1^2}{E_1} + \frac{1-v_2^2}{E_2} \quad \text{and} \quad 1 - \frac{1}{R_1} = \frac{1}{R_2}.
\tag{12}
\]

### 3 Shape of the liquid bridge and capillary force

In an elastic contact with a thin liquid film, the liquid film is squeezed out from the contact area to the contact edge. Then, a liquid bridge is formed between the two bodies, as shown in Fig. 1. This liquid bridge holds pressure, namely, Laplace pressure and surface tension. The sum of the Laplace pressure and surface tension represents the capillary force. When the liquid-bridge volume is sufficiently small, the effect of gravity is negligible, and the shape of the liquid bridge can be obtained by the Young–Laplace equation [22]. Our model does not consider the kinetic effects, for example, the fluid effect of liquid and visco-elastic effect of elastic plane. Therefore, our model assumes that the pressure inside liquid is determined by the Young-Laplace equation, and not changed by the fluid effect of liquid. Our model satisfies the energy equilibrium of the system, and can apply to the quasi-static experiments or very low speed phenomena. When the meridional curvature radius of the liquid bridge \( r_1 \) is much smaller than the azimuthal curvature radius \( r_2 \), the meridional curvature radius can be assumed to have a constant value [43]. Here, the Young–Laplace equation can be simplified as follows [25, 27]:

\[
P_{\text{Lap}} = \frac{\gamma_{\text{liquid}}}{r_1},
\tag{13}
\]

where \( \gamma_{\text{liquid}} \) is the surface tension of the liquid. The Laplace pressure is applied to the wetted area. Let us assume that the two bodies form a perfect contact and no liquid remains in the contact area. Here, wetted area \( A \) is obtained as follows:

\[
A = \pi(r_1^2 - a^2).
\tag{14}
\]

When \( r_1 \) is much smaller than \( 2a \), the wetted area can be approximated as

\[
A = 2\pi a(r_1 - a).
\tag{15}
\]

Then, the following approximation can be derived from the geometric condition shown in Fig. 1.

\[
r_1 = r_2 = r_f
\tag{16}
\]

In this condition, we approximate the height of the liquid bridge as the gap between the two bodies.

\[
h = d(r_f).
\tag{17}
\]

Equations (8), (17) and the geometric conditions lead to the following two expressions with regard to the height of the liquid bridge:

\[
h = \left\{ \begin{array}{l}
\frac{\Gamma}{\sqrt{\pi}} \cos\left(\frac{t}{\sqrt{\pi}}\right) \\
n_f \left[ \cos(\theta_c + \phi_f) + \cos(\theta_c - \phi_f) \right]
\end{array} \right.
\tag{18}
\]

where \( \theta_c \) and \( \theta_f \) are the contact angles between the sphere and liquid and the liquid and plane, respectively. In addition, \( \phi_f \) and \( \phi_f \) are the gradient angles of the sphere and the deformed plane, respectively.

\[
\phi_f - \tan^{-1}\left(\frac{\partial z_{\text{wetted}}(r)}{\partial r}\right) \quad \text{and} \quad \phi_f - \tan^{-1}\left(\frac{\partial z_{\text{wetted}}(r)}{\partial r}\right)
\tag{19}
\]

Equation (17) can be assumed to be near the contact edge \((r = a)\). The volume of the liquid bridge can be considered to be sufficiently small. Equation (18) yields the following expression:

\[
r_f = \frac{\Gamma}{\sqrt{\pi}} \cos\left(\frac{t}{\sqrt{\pi}}\right).
\tag{20}
\]

The liquid-bridge volume, i.e., \( V_{\text{liquid}} \) can be derived from Eqs. (5), (16).

\[
V_{\text{liquid}} = \int_0^\pi d(r) \cdot 2\pi dr = 2\pi \frac{\Gamma}{\sqrt{\pi}} \left[r_f^2 \cos\left(\frac{t}{\sqrt{\pi}}\right) - a \sqrt{r_f^2 - a^2}\right]
\tag{21}
\]

We know that the capillary force is expressed by the components of the Laplace pressure and the surface tension [22], which are respectively obtained as follows:

\[
F_{Lap} = \frac{p_{\text{Lap}}}{r_1} \quad \text{and} \quad F_{\text{wat}} = -2\pi a \gamma_{\text{liquid}} \sin(\theta_f + \phi_f).
\tag{22}
\]

Therefore, the capillary force is obtained as follows:

\[
F_{\text{cap}} = F_{Lap} + F_{\text{wat}} = 2\pi a \gamma_{\text{liquid}} \left[ \frac{d(r = a)}{r_1} + r_f \sin(\theta_f + \phi_f) \right].
\tag{23}
\]
4 Scalable model

The equations presented in Sections 2 and 3 can be normalized by sphere radius \( R \), which are respectively expressed as

\[
\tilde{z}_{\text{plane}}(\tilde{r}) = \frac{z_{\text{plane}}}{R} = \frac{1}{\gamma} \left( \frac{\tilde{r}^2 - \tilde{\delta}}{} \right), \quad (\tilde{r} < 1)
\]

(24)

\[
\tilde{z}_{\text{sphere}}(\tilde{r}) = \frac{z_{\text{sphere}}}{R} = \frac{1}{\gamma} \left( \frac{\tilde{r}^2 - \tilde{\delta}}{} \right), \quad (\tilde{r} \geq 1)
\]

(25)

\[
\tilde{\delta} = \frac{\delta}{\gamma} = \frac{\tilde{a}^2}{\sqrt{s}} - \frac{\pi}{4} \sqrt{s} \tilde{\gamma},
\]

(26)

\[
\tilde{d}(\tilde{r}) = \frac{d(\tilde{r})}{R} = \Gamma \sqrt{s} \cos^{-1} \left( \frac{\tilde{a}}{s} \right),
\]

(27)

\[
\tilde{F}_\text{el} = \frac{F_{\text{el}}}{R} = \frac{\pi \Gamma \sqrt{s} \cos^{-1} \left( \frac{\tilde{a}}{s} \right)}{R},
\]

(28)

\[
\tilde{V}_{\text{liquid}} = \frac{V_{\text{liquid}}}{R^3} = -2\pi \Gamma \sqrt{s} \left\{ \tilde{r}^2 \cos^{-1} \left( \frac{\tilde{a}}{s} \right) - \sqrt{s} \tilde{\gamma} \tilde{a}^2 + \tilde{\delta} \right\},
\]

(29)

where \( \tilde{a}, \tilde{r}, \) and \( \tilde{\delta} \) are the normalized parameters, i.e.,

\[
\tilde{a} = \frac{a}{R}, \quad \tilde{r} = \frac{r}{R} \quad \text{and} \quad \tilde{\delta} = \frac{\delta}{R}.
\]

Next, the elastic and capillary forces can be normalized by the McFarlane and Tabor approximate capillary force [Eq. (1)] [23].

\[
\tilde{F}_{\text{elas}} = \frac{F_{\text{elas}}}{F_{\text{MT}}} = \frac{N_0}{4} \tilde{a}^4 - \frac{\Gamma N_0}{4} \tilde{a}^4,
\]

(30)

\[
\tilde{F}_{\text{cap}} = \frac{F_{\text{cap}}}{F_{\text{MT}}} = \frac{\tilde{d}(\tilde{r} - \tilde{\delta})}{2 \tilde{r}} - \frac{\tilde{r}}{2} \sin(\tilde{\theta} + \phi),
\]

(31)

\[
\tilde{F}_{\text{elas}} = \frac{F_{\text{elas}}}{F_{\text{MT}}} \quad \tilde{F}_{\text{cap}} = \frac{F_{\text{cap}}}{F_{\text{MT}}}
\]

(32)

where \( \tilde{F}_{\text{elas}} \) and \( \tilde{F}_{\text{cap}} \) are respectively the normalized components of Laplace pressure and surface tension, and \( N_0 \) represents the ratio of the elastic force to the surface tension, i.e.,

\[
N_0 = \frac{F_R}{F_{\text{MT}}},
\]

(33)

The total force is the sum of elastic force \( \tilde{F}_{\text{elas}} \) and capillary force \( \tilde{F}_{\text{cap}} \). Therefore, total normalized force \( \tilde{F}_\text{total} \) is obtained as follows:

\[
\tilde{F}_\text{total} = \tilde{F}_{\text{elas}} + \tilde{F}_{\text{cap}}.
\]

(34)

5 Effects of hysteresis on the force curve

The contact process can be divided into two: loading and unloading processes (Fig. 2). The two processes have two types of hysteresis: hysteretic forces of the liquid volume and contact areas. During the loading process, the rigid sphere starts making contact with the elastic plane at \( \delta = 0 \) and moves downward. Then, the contact area increases, and the liquid film over the contact area is squeezed out to the contact edge, resulting in the formation of a liquid bridge and increase in its volume. After loading, the rigid sphere moves upward; thus, the unloading process starts.
From Eqs. (35)-(37), \( \tilde{r}_c \) can be solved as a function of four parameters: \( \tilde{a}_s, \tilde{a}_{\text{max}}, \Gamma, \) and \( l_{\text{liquid}}. \) As expressed by Eq. (36), the liquid volume experiences hysteresis between the loading and unloading processes. Although these equations cannot be analytically solved, their numerical solutions can be obtained.

Second, during the loading process, the liquid is squeezed out, and its contact angles are larger than the stable angle. Meanwhile, during the unloading process, the liquid is dragged out, and its contact angles become smaller. The contact angles are considered to experience hysteresis between the loading and unloading processes. Obtaining these contact angles results in the solution of Eqs. (35), (36) of the force between the two bodies. The hysteresis of the contact angles affects the force in the solution of Eqs. (35), (36) of the force between the two bodies. The origin of this difference is the transition of the contact angles can be estimated by this model.

Finally, the behavior under \( \delta < 0 \) described by the JKR theory (as mentioned in Section 2) does not affect the adhesion hysteresis.

### 6 Results and discussion

#### 6.1 Effects of contact angles

To calculate the force curve, the following six parameters are necessary: \( \tilde{a}_{\text{max}} \) (or \( \tilde{a}_{\text{max}} \)), \( l_{\text{liquid}}, l_{\text{liquid}}, \theta_s, \) and \( \theta_p. \) First, let us consider the force curve calculated from several values of contact angles \( \theta_s \) and \( \theta_p. \) These force curves are shown in Fig. 3. This calculated result is comparable with the contact between a millimeter-scale rigid sphere and a plane made of a rubber-like material covered by a 100-nm-scale water film. When the contact angles are 90°, the force curve almost fits the JKR theory because the capillary force is relatively small under this condition [see Eqs. (28), (32)]. When the contact angles are smaller or larger than 90°, another characteristic is shown with respect to the JKR theory in which the force curves of the same contact angles have different paths between the loading and unloading processes. The origin of this difference is the variation in the liquid volume [see Eqs. (35), (36)]. By comparing the maximum adhesion forces between the JKR theory and the proposed model under the condition of \( \theta_s < 90° \), the calculated force is larger than that of the JKR theory, which suggests that even a 100-nm-scale liquid film can cause strong adhesion.

We know that an advancing or receding contact angle is different from a stable angle. The total force can be calculated by obtaining the transition of the contact angle. The transition from advancement to recession has not been clarified. For example, the contact angles are assumed to linearly vary, similar to the profile shown in Fig. 4 (b). Then, the force curve is calculated, as shown in Fig. 4 (a). Figure 4 shows that the contact-angle transition changes the force curve, and the maximum adhesion force is different from that in the JKR theory. When the force, contact radius, and \( z \)-axis displacement are experimentally measured, the transition of the contact angles can be estimated by this model.

#### 6.2 Effects of the maximum \( z \)-axis displacement

Maximum adhesion forces \( F_{\text{adh}} \) are changed by maximum \( z \)-axis displacement \( \tilde{\delta}_{\text{max}} \), as shown in Fig. 5 (a), which shows that \( F_{\text{adh}} \) increases with \( \tilde{\delta}_{\text{max}} \). Figure 5 (b) shows the ratio of the capillary force to the total force when \( F_{\text{total}} = F_{\text{adh}} \), which is described as follows:

\[
\tilde{F}_{\text{adh}} = \frac{F_{\text{cap}}}{F_{\text{adh}}}.
\]

Fig. 5 shows that the capillary force increases with \( \tilde{\delta}_{\text{max}} \). As \( \tilde{\delta}_{\text{max}} \) increases, the contact area becomes larger. Subsequently, the increase in the contact area results in the increase in two parameters, i.e., accumulated liquid volume and wetted area. These parameters change the capillary force and the force...
curves. In the same manner, the increase in thickness of the liquid film results in a similar effect [see Eq. (36)].

6.3 Effects of the sphere radius (scale effects)

In this model, radius $R$ of the rigid sphere affects four parameters [see Eqs. (4), (26), (33), (37)], namely, $\delta_{\text{max.}}$, $\Gamma$, $N_{\text{SLP}}$, and $E^t_{\text{liquid}}$. Because the effect of $\delta_{\text{max.}}$ was already discussed in the previous section, the effect of the other parameters for fixed $\delta_{\text{max.}}$ is discussed in this section. The calculated result is shown in Fig. 6. The parameters are supposed to be those of a 100-nm-thick film of water, and the material of the plane is rubber-like. From the calculation under $\theta_{s,p} < 90^\circ$, $F_{\text{adh.}}$ inversely varies with $R$, which decreases in the range of small $R$ and increases in the range of large $R$. The force ratios also show the same tendency. Although we know well that the capillary force has a strong effect on a micro-scale contact such as an atomic-force microscopy measurement [24, 27], this result suggests that the capillary force is strong in a large-scale contact. This phenomenon is simply explained by the gap between the two bodies. As $R$ increases, the gap decreases; therefore, the liquid bridge extends to a larger area (the wetted area increases). In addition, the meridional curvature of the liquid bridge becomes smaller, which enhances the capillary force relative to the elastic force.

As shown in Fig. 6, the normalized maximum adhesion force $F_{\text{adh.}}$ has the local minimum against the sphere radius $R$ under $\theta_{s,p} = 0$–$80^\circ$. $F_{\text{adh.}}$ is divided into the elastic force ($F_{\text{JKR}}$), and the capillary force ($F_{\text{cap.}}$ and $F_{\text{Lap.}}$), which are respectively plotted in Fig. 7. As shown in Fig. 7, the trend of the local minimum is caused by the variation of $F_{\text{Lap.}}$. From Eqs. (22), (32), $F_{\text{Lap.}}$ is constructed by the two terms; $a_0(r_s - a)$ and $1/2r$. From Eq. (15), $a(r_s - a)$ approximately means normalized the wetted area against the orthogonal projection area of the sphere $\pi R^2$, and
is decreasing function against the sphere radius. \(1/2\bar{r}\) means the normalized curvature of the liquid bridge. In contrast to \(\bar{\varepsilon}(\bar{r} - \bar{a})\), \(1/2\bar{r}\) is increasing function against the sphere radius because the normalized curvature radius \(\bar{r}\) decreases with the normalized gap against the sphere radius around the contact area between the two contact bodies. The gap around the contact area calculated by the JKR theory is weakly affected by the sphere size (the sphere radius) because of the adhesion elastic deformation. Then, the normalized gap decreases with the sphere radius.

6.4 Application to experimental data

As explained in Section 6.1, the transition of the contact angles can describe force curves, which can be used to fit the experimental data. To apply this model to the experimental data, some of the necessary parameters, shown in Section 6.1, must be determined, and the condition for the transition of contact angles are assumed as follows,

\[
\begin{align*}
\theta_p - \theta_{cap,stable} + \varepsilon, \\
\theta_p - \theta_{cap,stable} + \varepsilon
\end{align*}
\]

where \(\theta_{cap,stable}\) and \(\theta_{p,stable}\) are stable liquid contact angles of the two surfaces and \(\varepsilon\) is the difference from static contact angles.

The experimental results of Baek’s work[46] is used here as an example. Baek’s work deals with the elastic contact between a glass spherical lens (\(R = 207.6\) mm) and a PDMS rubber block. The contact was displacement controlled by a motorized stage, and its speed was very low (0.1 \(\mu\)m/s) for the assumption of quasi-static condition. The experimental environment was kept under 50% humidity and 20-22°C.

The experiment was carried out for three different maximum displacements. The thickness of the liquid film covered with PDMS block is assumed as 100-nm-scale water film; \(\tilde{t}_{liquid} = 10^{-6}\). The stable contact angles are roughly estimated by sessile water drop test; \(\theta_{cap,stable} = 30^\circ\) and \(\theta_{p,stable} = 90^\circ\). In our model, the whole energy dissipation is assumed due to the capillary force, which corresponds to the “gradient of total energy dissipation” in [46].

Figure 8 shows the experimental result of the normalized capillary force, the fitted force curves and contact angles obtained by this model. As shown in Fig. 8, during loading process, \(\tilde{F}_{cap}\) and \(\varepsilon(\theta_p, \theta_s)\) are almost constant, whereas they significantly vary at the beginning and at the end of the unloading process. At the end of the unloading process, the glass sphere detaches from the rubber block, where it is observed that \(\varepsilon\) significantly decreases and goes to the limit of \(\varepsilon\) (Eq. (40)). This means the collapse of the liquid bridge.

7 Conclusion

The proposed model assumes a contact between a rigid sphere and an elastic half-space covered with a thin liquid film and explains the adhesion hysteresis by considering the liquid film squeezed out from the contact area. This hysteresis is classified into two types, which are that of the contact angles and that of the accumulated liquid volume. These hystereses are influenced by six parameters, namely, \(\tilde{t}_{liquid}\), \(\Gamma\), \(N_{EL}\), \(\tilde{t}_{liquid}\), \(\theta_p\) and \(\theta_s\), which depend on not only the elastic bodies but also the liquid film.

Some of the calculated results show that the capillary force is comparable with the elastic force. By varying sphere radius \(R\) when the liquid-film thickness is fixed, a strong adhesion force due to the thin liquid film is obtained in not only the micro-scale level but also the macro-scale level. This result suggests that the force generated between two contacting bodies can be affected by the capillary force regardless of the size of the contacting bodies.

References


