Adhesion Phenomena of Small Particles and Fine Wires†

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Abstract
Elastic and elasto-plastic adhesion behavior of nano- and micro-particles and wires are discussed. Theoretical elastic contact area (width or radius) is examined, taking into account surface energy. Important equations for adhesion and separation of small solids are introduced. The equations which give a stress distribution on the bonded area are introduced. As the radius of solids decreases, the ratio of adhesion (fractional contact radius or width) goes up to 80%. The effect of surface roughness on the adhesion is also examined and it is suggested that fine roughness gives full contact without pressure. The bond-strength gradually increases after adhesion. This is due to the stress induced diffusion along the bonded interface which reduces the residual stress which occurs on the bonded interface. It is suggested that the relaxation time required to remove the residual stress is shortened to several seconds as the small solid is of nano order.

KEY WORDS: (Adhesion) (Micro-joining) (Nano) (Particle) (Wire) (Solid state) (Surface energy)

1. Introduction
As the size of solids becomes small, the surface area \( S \) increases, compared with the volume \( V \). If the solid is a sphere with a radius \( R \), as the radius \( R \) decreases, the ratio \( S/V \) increases in proportional to \( S/V = 3/R \). Also, the surface energy \( \gamma \) exists on solid surfaces. Therefore, the surface energy has to be taken into account, as the solid becomes small. That is, the surface energy has a striking influence on the adhesion behavior of micro- and nano-solids \(^1, 2\). Metal surfaces exposed to air are covered with an oxide film and contamination (or organic film) \(^3\). The oxide film and contamination affect the surface energy and prevent adhesion. If these preventing factors are removed, the solids have high surface energy which they would originally have in their native state. This means that the surface is activated. Two solids with activated surfaces are able to exhibit an adhesion without any compressive force \(^1, 2, 4-6\). For example, if a fine gold wire with a diameter of 100 \( \mu \)m is touched to gold foil, then an adhesion occurs \(^2\). Fine powder and small particles often adhere to surface of walls and desks. These phenomena can be affected by the surface energy. The contact area between them is also influenced by force or weight applied to particles or wires. In the present review, the fundamental equations with respect to the adhesion of small particles and fine wires are introduced. These equations are very useful to understand the adhesion behavior of minute solids. Also, the adhesion behavior under low and room temperature conditions are discussed.

2. Contact behavior taking into account surface energy
Adhesion between spheres (particles) were formulated by K. R Johnson et al. \(^7, 8\). Figure 1 illustrates the cross sections of two elastic solids (spheres or wires). If the Dupré energy of adhesion \( \Delta\gamma (= \gamma_1 + \gamma_2 - \gamma_i) \) is greater than zero, then two solids can a contact without any compressive force, where \( \gamma_1 \) is the surface energy of solid 1, \( \gamma_2 \) is the surface energy of solid 2 and \( \gamma_i \) is the interface energy of the bond interface formed by the elastic adhesion of two solids 1 and 2. Two solids adhere so that elastic strain energy taking the Dupré energy into consideration around the contact area can be a minimum. The theoretical equations for elastic adhesional contact are described in turn for small particles and fine wires.

2.1 Adhesion theory between spheres
In Fig. 1, \( F \) is the load (force) applied to solids 1 and 2 (for spheres, particles). Two spheres with the radii of \( R_1 \) and \( R_2 \) are contacted as illustrated in Fig. 1. The cylindrical coordinate \((r, \gamma)\) is used for the contact of spheres. The contact radius \( a_j \) due to elastic adhesion taking surface energy into account is expressed by

\[
a_j^3 = \frac{3\pi k}{2} \cdot R \left( F + 3\pi R \Delta\gamma \right) \left[ \left( 3\pi R \Delta\gamma \right)^2 + 6\pi R \Delta\gamma \cdot F \right] ,
\]

where \( R \) is given by

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\[ \frac{1}{R} = \frac{1}{R_1} + \frac{1}{R_2}, \quad (2) \]

The contact-radius \( a_j \) for \( F = 0 \) is expressed by

\[ a_j^3 \text{ for } F = 0 = 9 \cdot (\pi R)^2 k \cdot \Delta \gamma . \quad (5) \]

The force to separate two solids is given by

\[ F_{j \text{min}} = -\frac{3}{2} \pi R \Delta \gamma , \quad (6) \]

and the contact radius, which is the minimum in this stage is obtained by

\[ a_{j \text{ min}} = \left[ \frac{3^2 \pi^2}{2^2} k R^2 \Delta \gamma \right]^{1/3}. \quad (7) \]

Let us find the stress distribution \( \sigma_i(r) \) in the \( y \)-direction along the contact interface, where \( r \) is the radius distance as shown in Fig. 1. It is now assumed that the contact radius \( a_j \) is produced when \( F = F_j \) is applied to the two spheres under the condition of \( \Delta \gamma > 0 \). The Hertz solution \( a_h \) is obtained when \( F = F_e \) is applied to them, i.e., the \( a_h \) is obtained after \( \Delta \gamma = 0 \) is substituted for equation (1). In this stage, the \( a_j \) is equal to the \( a_h \) and \( F_e \) is greater than \( F_j \).

The relationship between the radius \( a_h \) and the effective force \( F_e \) is expressed by

\[ a_h^3 = \frac{3}{2} \pi \cdot k \cdot R \cdot F_e . \quad (8) \]

The stress distribution \( \sigma_i(r) \) of the Hertz solution for \( \Delta \gamma = 0 \) along the radius direction \( r \) is expressed by

\[ \sigma_h(r) = \frac{3F_e}{2\pi a_j^2} \left( 1 - \frac{r^2}{a_j^2} \right)^{1/2} , \quad (9) \]

where the \( \sigma_i(r) \) is the stress component perpendicular to the contact interface (\( x \) axis). When the force \( F \) is reduced from \( F_e \) to \( F_j \), keeping the contact area at \( a_j = a_h \), Boussinesq’s stress \( \sigma_m(r) \), which means the stress distribution lost by the stress reduction, is given by

\[ \sigma_m(r) = \frac{F_j - F_e}{2\pi a_j^2} \left( 1 - \frac{r^2}{a_j^2} \right)^{1/2} . \quad (10) \]

The stress distribution in the \( y \) direction along \( r \) axis is, therefore, obtained by

\[ \sigma_y(r) = \sigma_h(r) + \sigma_m(r) . \quad (11) \]

It is understood from eq. (11) that a tensile stress occurs in the periphery of the contact area when \( \Delta \gamma > 0 \). This is the traction which is the cause of stress induced diffusion, as stated later.

### 2.2 Adhesion theory between wire (cylinders).

When \( \Delta \gamma > 0 \), the adhesional contact is produced between wires as well as spheres. The formulation of adhesional elastic contact for wires was stated elsewhere \(^9\). The important equations will be shown in the present paper. It is
The separating force at that stage is given by
\[ f_j = -3 \left( \frac{R \cdot (\Delta y)^2}{2^5 k} \right)^{\frac{1}{3}} \]  

On the other hand, the stress distribution in the \( y \) direction along \( x \) axis is given by eq. (18) at the contact interface for Hertz contact under the condition of \( f = f_e \):
\[ \sigma_h = \frac{2f_e}{\pi a_j} \left( \frac{1 - \left( \frac{x}{a_j} \right)^2}{2} \right)^{\frac{1}{2}} \]

When the load is removed from \( f = f_e \) to \( f = f_j \), Boussinesq’s stress distribution \( \sigma_m \) is obtained by
\[ \sigma_m = \frac{f_j - f_e}{\pi a_j} \left( \frac{1 - \left( \frac{x}{a_j} \right)^2}{2} \right)^{\frac{1}{2}} \]

Boussinesq’s stress given by eq. (19) is the tensile stress distribution for the interface with a sharp notch. The stress distribution at the adhesion interface under \( f = f_j \) is, therefore, given by
\[ \sigma_y(x) = \sigma_h + \sigma_m \]

Figure 2 shows the stress distributions at the contact interface. The adhesional elastic contact has a tensile stress drive the vacancy diffusion along the adhesive interface. The equations (1) to (20) are very useful to understand the elastic adhesion of small solids. The stress distribution which has tensile and compressive stress areas drives the vacancy diffusion along the adhesive interface.

3. Elastic adhesion

Let us discuss the elastic adhesion of small solids for example a gold ball and wire, the material constants are shown in Table 1. At first, the plastic deformation is ignored. Figure 3 shows the elastic contact radius \( a_j \) of spheres depending on the applied force \( F \). The sign of \( F \) is taken to be positive when it is compressive. Fig. 3 (a) is for the sphere radius of \( R = 50 \mu m \) and Fig. 3 (b) is for \( R = 0.5 \mu m \). As seen in Fig. 3, the contact radius of a sphere/plane is larger than that of a sphere/sphere. As the radius \( R \) becomes small, the adhesion can be produced even if \( F \) is less than zero. The small particles cannot only easily adhere to each other but also to planes. The smaller they are, the more easily they adhere.
Table 1  Materials constant of pure gold.

<table>
<thead>
<tr>
<th>Name</th>
<th>Symbol</th>
<th>Value (Unit)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Surface energy</td>
<td>( \tau_s )</td>
<td>1.485 (J/m²)</td>
</tr>
<tr>
<td>Interface energy</td>
<td>( \tau_i )</td>
<td>0.26 (J/m²)</td>
</tr>
<tr>
<td>Energy difference</td>
<td>( \Delta \gamma = 2\gamma_s - \gamma_i )</td>
<td>2.61 (J/m²)</td>
</tr>
<tr>
<td>Shear modulus at 300K</td>
<td>( G(300) )</td>
<td>2.91 x 10¹⁰ (N/m²)</td>
</tr>
<tr>
<td>Poisson’s ratio</td>
<td>( \nu )</td>
<td>0.42</td>
</tr>
<tr>
<td>Young’s modulus</td>
<td>( E = 2(1+\nu)G )</td>
<td>8.27 x 10¹⁰ (N/m²)</td>
</tr>
<tr>
<td>Elastic constant</td>
<td>( k )</td>
<td>3.17 x 10⁻¹² (m²N⁻¹)</td>
</tr>
<tr>
<td>Yield stress at 300K</td>
<td>( \sigma_Y )</td>
<td>9.93 x 10⁷ (N/m²)</td>
</tr>
</tbody>
</table>

Fig. 3  Adhesional elastic contact, depending on compressive force \( F \) and radius \( R \) for gold sphere. 
(a) \( R = 50 \mu m \) and (b) \( R = 0.5 \mu m \).

Fig. 4  Half the contact width of elastic adhesional contact for gold wires.

Fig. 5  Dependence of elastic contact radius \( a_j \) and contact ratio \( a_j/R \) on sphere radius \( R \) for gold particle. 
(a) Sphere/sphere when spheres (particles) have the same radius \( (R = R_1 = R_2, F = 0) \), and (b) Sphere/plane \( (R_2 = \infty, F = 0) \).
Figure 4 shows half the elastic contact width $a_j$ of fine wires with radius of 50μm. It is actually difficult to adhere a wire to a wire in parallel to each other. Even if wire/wire contact can be made, the contact area is small as shown in Fig. 4. It is not so difficult to adhere a wire to a plane under room temperatures. The room temperature bonding of fine wires to metal substrates was actually carried out in the previous study 41.

As the radius $R$ decreases, the contact area decreases. This is a natural phenomenon but the adhesion (contact) ratio $a_j/R$ increases. Figures 5 and 6 show the dependences of the adhesion (elastic contact) radius (width) $a_j$ and the adhesion (elastic contact) ratio $a_j/R$ on $R$ for small particles and fine wires, respectively. These calculated results were obtained under applied force free. Fig. 5 (a) is for adhesion between small Au-particles and Fig. 5 (b) is between Au-sphere and Au-plane. Fig. 6 is the case when fine Au-wire is contacted with a flat Au substrate. As seen in Figs. 5 and 6, the adhesion ratio increases to be greater than 50% as the solids (particles or wire) become smaller, where $R$ is the initial radius of solids. Pressureless adhesive bonding is possible when the solid reaches the nano level.

4. Influence of plastic deformation on contact phenomena

As solids become small, down to the sub micron level, a number of mobile dislocations decrease in the crystalline solids and nano solids have no mobile dislocations because they go out through the solid-surface. So, plastic deformation due to dislocation slip can be ignored in sub micron and nano solids. However, as the radius $R$ of particles or wires increases up to a few tens of microns, the plastic deformation influences the adhesional contact, i.e., the elasto-plastic contact can occur with increasing the applied force $F$ or $f$.

Figure 7 shows the stress distribution (calculated results) along the adhesional contact interface (in the $x$ direction as shown in Fig. 1) when a fine Au-wire with $R = 50μm$ is contacted to a flat Au plane under $f = 500 \text{ N/m}$ and $T = 300 \text{ K}$, where $T$ is the absolute temperature. The stress $\sigma$ is a component in the $y$ direction. The dotted curve is for elastic deformation and the solid curve is for elasto-plastic deformation. Half the elastic contact width $a_j$ is 1.42μm and half the elasto-plastic contact width $a_{ep}$ is 1.67μm. The stress distribution is solved as the plane strain problem. The von Mises yield condition is expressed by

$$\sigma_{yield} = \frac{2}{\sqrt{3}} \left(1 + \frac{\pi}{2}\right) \sigma'$$

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where $\sigma_y$ is the yield stress for plastic deformation under a uniaxial stress condition. The plastic deformation occurs at the area where $\sigma$ is greater than $\sigma_{y} = 294.7$ MPa, increasing the contacted area. As seen in Fig. 7, the tensile stress occurs at the edge area bonded even if a compressive force $f = 500$ N/m is applied to the wire. The summation of tensile and compressive stress is equal to the compressive force $f$. The applied pressure $P$ is defined by $P = f/2R$. It is an average pressure and $P = 5$ MPa in Fig. 5. Plastic deformation can influence the contact behavior even under a low pressure such $P = 5$ MPa.

Figure 8 shows half the contact width $a$ (calculated results\(^4\)) depending on the applied force $f$. The applied pressure is calibrated in the horizontal axis at the upper side of Fig. 8. The solid curves are for elastic contact and dotted curves for elasto-plastic contact. When $P > 1$ MPa, the plastic deformation begins to contribute to the contact because the mobile dislocation can exist in the solids.

**5. Contact behaviors with fine surface roughness**

In this session, the surface roughness contact is argued shortly. The two dimensional surface roughness is approximated to a sinusoidal curve illustrated in Figure 9. $L$ is half the wave length and $h$ is the surface roughness height. The curvature radius $\rho$ at the top of the surface roughness is given by $2L^2/(h\pi^2)$. Half the local contact width $X$ was assumed to be obtained by the elastic contact between fine wires with radius of $\rho$. Figure 10 shows the percentage contact area $X/L$ depending on the local pressure $P_L$. When a fine wire adheres to the substrate, the contact interface is loaded by the stress greater than 300 MPa as shown in Fig. 7. It is, therefore, suggested that the contact area has no voids when the surface roughness has $h$ less than 50 nm, even if the small solid has such a fine surface roughness. Furthermore, as $h$ decreases to be less than 10 nm, the full contact can established under low pressure condition and even under pressureless condition\(^10\). As the solid (sphere) becomes smaller than sub-micron, the surface roughness does not affect the elastic contact so largely because of adhesion force. However, the surface roughness of the pad (substrate) has a very large influence on contact behavior. When $L$ and $h$ increase, it is natural that the influence becomes striking\(^10\).

**6. Experimental procedure**

It is necessary to activate the surface of solid to produce the effect of Dupré energy of adhesion. In the present study, the adhesion tests were carried out under ultra high vacuum condition of $< 1x10^{-8}$ Pa.

Figure 11 illustrates the adhesional bonding apparatus used in the present study. A gold wire with a diameter of 100 $\mu$m and a purity of 99.99 mass % and a gold foil with a thickness of 130 $\mu$m were used as the bonding specimens. After the bonding specimens were degreased by acetone and ethanol, they were placed one by one in the introduction chamber and moved into the Ar ion irradia-
time is less than 10min, the maximum surface roughness does not become so large. However, it is seen in Fig. 12 (c) for $I_e = 25$ mA, $V_a = 2$ kV and $t_i = 2$ hr that the surface is rouged by Ar ion irradiation ($R_{\text{max}} = 0.47 \mu$m).

Figure 13 shows the Vickers hardness $H_v$ of gold foils before and after Ar ion irradiation. It is expected that Ar ion irradiation increases vacancy concentration. Because vacancies are point defects, Ar ion irradiation increases the surface hardness. The Vickers hardness after irradiation

**Fig. 11** Apparatus of room temperature bonding.
The system consists of three chambers. Introduction chamber is to prepare the bonding specimens. Irradiation chamber is to activate the specimen’s surface by Ar ion irradiation. Main chamber is to bond the specimens (wire and foil).

Fig. 12 Surface profiles of gold foils. (a) before Ar ion irradiation, (b) after irradiation ($I_e = 1$ mA, $V_a = 2$ kV, $t_i = 2$ hr), (c) $I_e = 25$ mA, $V_a = 2$ kV, $t_i = 10$ min, and (d) $I_e = 25$ mA, $V_a = 2$ kV, $t_i = 2$ hr.

7. Experimental results and discussion

Figure 12 shows surface profiles of gold foils after Ar ion irradiation which are compared with the surface profile before Ar ion irradiation. The maximum surface roughness $R_{\text{max}}$ was 0.21 $\mu$m, before Ar ion irradiation. After Ar ion was irradiated on to gold foils, the surface was rough but when the emission current is less 1mA or the irradiation time is less than 10min, the maximum surface roughness does not become so large. However, it is seen in Fig. 12 (c) for $I_e = 25$mA, $V_a = 2$kV and $t_i = 2$hr that the surface is rouged by Ar ion irradiation ($R_{\text{max}} = 0.47 \mu$m).

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A compressive stress exist at the central area but a tensile stress remains at the peripheral area, i.e., vacancy concentration differs between the central area and peripheral area. Vacancy diffusion occurs to reduce the difference of elastic stress (residual stress) \( \sigma_{\text{max}} \). It was also found that the bonded area did not increase by holding the specimens after adhesion \( t_r \). Because of room temperature (low temperature), the vacancy diffusion flow is controlled predominantly by the interface self-diffusion. The maximum tensile stress \( \sigma_{\text{max}} \) is produced at the peripheral area by elastic deformation. The time \( t_r \) required for the interface self-diffusion to reduce \( \sigma_{\text{max}} \) by the same quantity must be proportional to the increase of bond-strength. The relationship between time \( t_r \) and the interface (boundary) self-diffusion \( D_b \) is expressed by

\[
D_b \cdot E \cdot t_r / T = \text{Const},
\]

where \( E \) is Young’s modulus, \( T \) is the absolute temperature and \( \text{Const} \) is a symbol which means constant. The interface self-diffusion is given by

\[
D_b = D_{bo} \exp \left( - \frac{Q_b}{RT} \right),
\]

where \( D_{bo} \) is the frequency factor, \( Q_b \) is the activation energy of interface self-diffusion, and \( R \) is the gas constant.

If the increase of bond-strength after bonding is due to the recovery of residual strain by the interface self-diffusion, the time required to increase the same quantity of the bond-strength is given by

\[
\frac{T}{t_r} = k \cdot \exp \left( - \frac{Q_b}{RT} \right),
\]

where \( k \) is a constant. The time \( t_r \) required to obtain \( F_p \)

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**Figure 14** shows comparisons between calculated and experimental results for Au-wire/Au-foil (Au pad). The applied pressure \( P \) is given by \( P = f/2R \), where \( f \) is the applied compressive force and \( R \) is wire radius \( (R = 50 \mu m) \). Au small solids with a diameter of 100 \( \mu m \) have the yield stress condition \( \sigma_{\text{yield}} = 294.7 \) MPa, although the smaller solids with sub micron diameters have higher yield stress or become a perfect elastic solid because they have few glissile (mobile) dislocations in the bulk. In Fig. 14, because of \( \sigma_{\text{yield}} \), plastic deformation contributes the adhesion. The experimental points agree with the dotted line of elasto-pastic adhesional contact \( \alpha_{ep} \), where \( \alpha_{ep} \) is half the elasto-plastic adhesional contact width, \( \alpha_{p} \) is half the plastic contact width and \( a \) is half the contact width. However, the experimental points agree with half the elastic adhesional contact \( \alpha_{j} \) at \( P < 3 \) MPa. Initial adhesion of small solids will be governed by elastic deformation if the bonding pressure is zero. When \( R \) is less than 1 \( \mu m \), Figs. 5 and 6 will predict that the elastic adhesion occurs.

**Figure 15** shows experimental results of change in the bond-strength \( F_p \) (pull strength) with holding time after adhesional bonding under room temperature (298K) and \( P = 5 \) MPa. As seen in Fig. 15, the bond-strength increases with the holding time. As the holding temperature increases, the bond-strength increases rapidly. This is due to stress induced vacancy diffusion. At first, the central area bonded has a higher density than the peripheral area. The experimental points agree with the dotted line of elasto-pastic adhesional contact \( \alpha_{ep} \), where \( \alpha_{ep} \) is half the elasto-plastic adhesional contact width, \( \alpha_{p} \) is half the plastic contact width and \( a \) is half the contact width. However, the experimental points agree with half the elastic adhesional contact \( \alpha_{j} \) at \( P < 3 \) MPa. Initial adhesion of small solids will be governed by elastic deformation if the bonding pressure is zero. When \( R \) is less than 1 \( \mu m \), Figs. 5 and 6 will predict that the elastic adhesion occurs.
after stress induced diffusion. The stress induced diffusion can quickly occur by reducing the radius $R$ of small solids. It is rather difficult to form one nano wire (or particle) contact bonding and measure the bond strength. In the present study, it has not been carried out. However, we can easily deduce that the stress relaxation time $t_r$ required to remove the maximum tensile stress $\sigma_{max}$ decreases down to several seconds as the radius $R$ decreases to nano level. Also, the bond-ratio $a_j/R$ goes up to 80% 4,5,10).

Figure 17 shows the stress relaxation time $t_r$ and the ratio $a_j/R$, depending on the radius $R$. For nano level small solids, pressureless room temperature bonding can be produced. This can be residual stress free in a short time. The initial adhesional contact area of small particles and fine wires will be predicted by equations (1) and (12), respectively. However, residual stress occurs around the contact interface. This residual stress changes vacancy concentration. As a result, the stress induced diffusion occurs along the contact (bonded) interface and the bonded area can be residual stress free.

8. Summary
The theory of adhesional contact joining for small solids (particles and wires) was introduced and the contact behaviors were discussed. Elastic contact phenomena can be predicted well but as the actual adhesion of small solids can be influenced by plastic deformation and diffusion, it is difficult to understand the actual adhesion behavior. Also, surface activating processes such as a Ar ion irradiation changes the surface situation of solids. Materials constant may be changed. The surface roughness is largely changed by strong Ar ion irradiation. It is important to activate the bonding surface, i.e., to remove oxide surface film but too much ion irradiation gives a damage to the surface. As solids (particles) becomes small to nano level, the bonding ratio goes up to 80% and the relaxation time required to remove the residual stress becomes several seconds.

The adhesional bonding behavior of small solids were reviewed based on the experimental results carried out under the conditions of low bonding pressures and an ultra-high vacuum. A lot of unknown problems have remained about adhesion such as tribology. The further researches are expected to solves a lot of these problems.

References

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