Effective friction in adhesive mixtures intended for inhalation: Simulation of oblique impact of adhesive units
Sohan Sarangi, Göran Frenning ∗
Department of Pharmaceutical Biosciences and Swedish Drug Delivery Centre, Uppsala University, Box 591, 751 24 Uppsala, Sweden

GRAPHICAL ABSTRACT

HIGHLIGHTS
• Adhesive units with different fine shapes were formed in silico.
• Binary collisions with different impact angle to study effective friction.
• Effective friction is significantly lower compared to bare carrier particles.
• Tetrahedral shape with angles towards grazing impact showed highest friction.

ARTICLE INFO

Keywords:
Oblique impact
Adhesive mixtures
Inhalation
Modelling

ABSTRACT

Oblique impact between adhesive units was studied using the Discrete Element Method (DEM) as a means to assess the effective friction coefficient and its dependence on particle shape, surface coverage ratio (SCR; 0.5, 0.75 and 1) and impact angle (15° – 75°) in a low handling velocity regime (0.6 – 1.6 m/s). Adhesive units were created in silico from a spherical carrier particle (100 μm diameter) and monodisperse fine (drug) particles (3 μm) of a spherical, triangular bipyramidal or tetrahedral shape. The particle interaction was modelled using the Hertz–Mindlin contact model with JKR adhesion (surface energy 0.03 J/m²). A total of over 300 simulations were performed and the effective friction coefficient was extracted from the normal and tangential forces experienced by the carrier particles. The adhered fines resulted in a significantly reduced friction coefficient in comparison to the bare carriers but no additional reduction was observed with increasing SCR, suggesting a saturation already at an SCR of 0.5. The effective friction coefficient was independent of the impact velocity.

https://doi.org/10.1016/j.powtec.2022.118075
Received 16 September 2022; Received in revised form 20 October 2022; Accepted 3 November 2022
Available online 9 November 2022
0032-5910/© 2022 The Authors. Published by Elsevier B.V. This is an open access article under the CC BY license (http://creativecommons.org/licenses/by/4.0/).

* Corresponding author.
E-mail address: goran.frenning@farmbio.uu.se (G. Frenning).
1. Introduction

The inhalation pathway for delivery of drugs/Active Pharmaceutical Ingredients (APIs) to the lungs has been extensively studied during the last few decades. Over time, Dry Powder Inhalers (DPIs) have become a standard device for delivery of APIs with local and systemic effects [1]. Most DPIs use a special type of mixture known as an adhesive mixture [2,3] that consists of micronised API (fine particles) of size < 5 μm and comparatively larger sized carrier particles (~ 100 μm). Owing to their high surface-to-volume ratio, the fine particles have a significant effect of van der Waals forces, leading to high cohesiveness and agglomeration, resulting in low flowability [2,4]. Adhesive units, consisting of fine particles on the surface of carrier particles, improve flowability, maximize homogeneity and improve dispersibility of the API during the aerosolisation process [4]. Understanding the effective behaviour of these units during formation and handling would aid optimization of drug delivery to the lungs and pave the way for high drug loads [5,6] using DPIs.

Adhesive mixtures are complex and difficult to understand. The interplay between interdependent parameters is critical for the understanding of the formulation, handling and dispersion performance of DPIs [7]. Even the most simplified systems consisting of lactose carriers and lactose fine particles seem to depend on an array of factors such as the cohesive–adhesive balance [8] and the size, shape and morphology of the fines and carriers [9–11], making correlation between the factors difficult to disentangle. Numerical modelling and simulation enable systematic parametric variations that avoid confounding of different factors, providing a clearer picture.

Numerical studies have been used to better understand the formation mechanisms of adhesive mixtures [12–14], their micromechanics during handling [15–17] and the dynamics of particles during aerosolisation [18]. There have been studies conducted focusing on agglomeration and deagglomeration of aggregates in DPIs [19,20] and the influence of surface energy thereon [21]. Computational Fluid Dynamics (CFD) and the Discrete Element Method (DEM) are the two most commonly used tools to study dynamics of particles in air streams. Recent studies have addressed the effects of carrier morphology [14] and fine-particle shape [22] as well as surface energy during the manufacturing and handling phase of adhesive mixtures.

Previous studies of the interaction between adhesive units have to a large extent focused on normal (head-on) impact. The overall mechanical response may in this case be summarized in terms of a (normal) coefficient of restitution. The situation is more complex for non-normal, i.e. oblique, impacts. Experimental studies and theoretical analysis of (large) non-cohesive spherical particles indicate that two regimes can be identified; a sticking regime for low impact angles (near head-on impacts) and a sliding regime for larger angles (near grazing impacts) [23–25]. The response in the sticking regime is typically characterized by a tangential coefficient of restitution and that in the sliding regime by a friction coefficient. This situation corresponds to oblique impact between bare carriers, in which case two parameters, the tangential coefficient of restitution and the friction coefficient, are typically at play.

Oblique impact of adhesive elastic particles was studied by Thornton et al. [26] who explained the departure of from the results obtained with the Hertz–Mindlin model. However, studies trying to explain the effective behaviour of adhesive units undergoing oblique impact are scarce. Adhered fines are expected to dramatically influence the effective behaviour of the particles, at least when present in sufficient amounts, since the interaction then is mediated via the fines whereas the carrier particles typically do not touch.

The current study is the first attempt to assess the effective (frictional) behaviour of adhesive units undergoing oblique impact in a handling velocity regime [27–29]. To this end, the DEM was used to study the effect of particle shape, impact angle, impact velocity and number density of fines (represented as a surface coverage ratio) on the effective tangential force between adhesive units. The shape of the particle was derived from our previous study [16] which focused on spherical, triangular bipyramidal and tetrahedral fine particles. The current study thus complements our previous work [15–17], in which normal (head-on) impact between adhesive units was investigated, and provides information needed for a more complete understanding of the effective mechanical behaviour of adhesive units. For brevity the carrier particles are referred to as carriers and fine (drug/API) particles are referred to as fines.

Numerical framework

1.1. The Discrete Element Method (DEM)

The interaction between the adhesive units was modelled using the DEM. The constituent particles are treated as discrete entities in the DEM and interact predominantly by short-range forces. The contact forces are calculated based on the effective overlap between the particles at the point of contact. The translational motion is governed by Newton’s laws represented as,

\[ m_i \frac{d^2 x_i}{dt^2} = m_i g + \sum_j f_{ij}^n + f_{ij}^e + f_{ij}^{fluid} \quad (1) \]

where \( i \) is the target particle and \( j \) is a neighbouring particle, \( m_i \) is the mass and \( x_i \) is the position of particle \( i \), \( g \) is acceleration due to gravity, \( f_{ij}^n \) and \( f_{ij}^e \) are normal and tangential forces acting on particle \( i \) due to contact with particle \( j \) and \( f_{ij}^{fluid} \) is the force on particle \( i \) resulting from fluid interaction. As in our previous study [22], a low velocity regime is considered to create an effective multiscale model to study the stability of adhesive mixtures during handling. The fluid forces in Eq. (1) are neglected owing to the low drag forces resulting from small velocities. The impact of gravity, though implemented, is negligible for individual particles owing to the small mass and time scale of study. The equation of motion for the rotational degrees of freedom could be expressed as,

\[ \frac{d\omega}{dt} + \omega \times (I\omega) = M \quad (2) \]

where \( I \) is the inertia tensor, \( M \) is the applied torque and \( \omega \) is the angular velocity about the centre of mass of the particle.

In this work, cohesive contact laws similar to the ones stated by Thornton et al. [30,31] are used. The effective properties between the particles could be defined as follows:

- The effective radius \( (R^e) \), and effective mass \( (M^e) \) of the two particles are represented as the harmonic mean of the respective mass and radii as stated in Eqs. (3) and (4)

\[ \frac{1}{R^e} = \frac{1}{R_i} + \frac{1}{R_j}, \quad (3) \]
\[ \frac{1}{M^e} = \frac{1}{M_i} + \frac{1}{M_j}. \quad (4) \]

where \( R_i, M_i \) are the radius and mass for particle \( i \) and \( R_j, M_j \) for particle \( j \).
The adhesive/cohesive interaction between the particles could be governed by the DMT (Derjaguin–Muller–Toporov) [33] model or the JKR (Johnson–Kendall–Roberts) model. The Tabor parameter was calculated for the appropriate contact model for adhesive/cohesive contacts given by,
\[ \mu_{\text{Tabor}} = \left( \frac{R_1 R_2}{E^c} \right)^{1/3}, \]  
where \( E \) is the surface energy, representing the strength of adhesion/cohesion between the two particles.

The normal elastic force \( F_{\text{ne}} \) between two particles is represented as,
\[ \frac{F_{\text{ne}}}{F_c} = 4 \left( \frac{a}{a_0} \right)^3 - \left( \frac{a}{a_0} \right)^{3/2}, \]  
where \( F_c = 3 \pi G R^2 \) is the pull-off force between the two particles, \( a \) is the contact radius at a given time and \( a_0 = \left( \frac{9 \pi G R^2}{E^c} \right)^{1/3} \) is the finite contact radius due to the attractive force in the absence of external load [39,40]. The contact radius is obtained from the normal overlap \( \delta_n \) via an expression of the form
\[ \frac{\delta_n}{\delta_c} = 6 \left[ \frac{2 \left( \frac{a}{a_0} \right)^2 - 4 \left( \frac{a}{a_0} \right)^{3/2}}{2(6)^{1/3} R} \right], \]  
where \[ \delta_c \equiv \frac{a_0^3}{2(6)^{1/3} R}. \]  
Likewise, the tangential elastic force \( F_{\text{te}} \) is defined as [37],
\[ F_{\text{te}} = S_t \delta_t, \]  
where \( S_t \) is the tangential stiffness and \( \delta_t \) is the tangential overlap. The detailed calculation of overlap and tangential stiffness is described in our previous work [15]. Normal \( (F_{\text{nad}}) \) and tangential \( (F_{\text{tid}}) \) damping forces are defined as [38],
\[ F_{\text{nad}} = 2 \sqrt{\frac{5}{9}} \beta \sqrt{S_n} M^2 v_n, \]  
\[ F_{\text{tid}} = 2 \sqrt{\frac{5}{9}} \beta \sqrt{S_t} M^2 v_t, \]  
where \( \beta \) is a dimensionless factor dependent of the coefficient of restitution, \( S_n (S_t) \) is normal (tangential) stiffness and \( v_n (v_t) \) is the normal (tangential) relative velocity between the particles. Hence the coefficient of restitution is used to parameterize the magnitude of the damping forces. Although alternative contact models predict a (weak) dependence of the coefficient of restitution on impact velocity [41], experimental data indicate that this parameter can be treated as a constant for a relatively large range of conditions [24], suggesting that the model used is sufficient in the present context.

Standard Coulomb-type friction was used, such that the maximal tangential force is given by
\[ F_{\text{max}} = \mu_s F_n, \]  
where \( \mu_s \) is the coefficient of static friction. Likewise, standard rolling friction was used, with a torque \( M_s \) given by
\[ M_s = -\mu_s F_n R, \]  
where \( \mu_s \) is the coefficient of rolling friction and \( R \) is the distance from the particle centre to the contact point.

### 2. Methods

#### 2.1. Primary particles

The carrier particles were modelled as elastic spheres with contact damping. The diameter of the carrier particles was 100\( \mu \)m with properties mimicking those of lactose (Table 1). Fine particles were modelled as multispheres composite particles of three different shapes; spherical, triangular bypripalidal and tetrahedral, in line with our previous work [22] and also illustrated in Fig. 1. The maximal diameter of the fine particles was fixed at 3\( \mu \)m. The composite shapes with multispheres were designed so as to ensure conservation of volume across shapes [15]. The multispheres represent various degrees of complexity of particle shape which controls their interlocking and rolling ability. Thus, the triangular bypripalidal particles represent a modest (in relative terms) and the tetrahedral particles a significant deviation from a spherical shape. The surface energy that controls the strength of adhesion/cohesion between the carrier–carrier, carrier–fines and fines–fines was fixed at 0.031/m\(^2\) [12,42,43].

#### 2.2. Adhesive units

The adhesive units were created in-silico by pseudo-randomizing the fine particles on the surface of the carrier particles [15]. This was attained by creating a uniformly spaced envelope of the desired particle number density. Each of the fine particles was given a random velocity and the system was evolved until smaller agglomerates of fine particles were formed. Thereafter the fine particles were given a velocity directed towards the centre of the carrier particle. The system was evolved until an envelope of weakly attached fine particles on the surface of the carrier particle was formed. The number density of fine particles on the surface of the carrier particle was quantified in terms of the surface coverage ratio (SCR) represented as,
\[ \text{SCR} = \frac{N \times \pi r^2 \text{fines}}{4 \pi r^2 \text{carrier}}, \]  

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Coarse particle (carrier)</th>
<th>Fine particle (fines)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Particle Size (Diameter, ( \mu )m)</td>
<td>100</td>
<td>3</td>
</tr>
<tr>
<td>Poisson’s ratio (-)</td>
<td>0.35</td>
<td>0.35</td>
</tr>
<tr>
<td>Density (kg/m(^3))</td>
<td>1540</td>
<td>1540</td>
</tr>
<tr>
<td>Young’s Modulus (GPa)</td>
<td>3.5</td>
<td>3.5</td>
</tr>
<tr>
<td>Coefficient of Static Friction (-)</td>
<td>0.5</td>
<td>0.5</td>
</tr>
<tr>
<td>Coefficient of Rolling Friction (-)</td>
<td>0.01</td>
<td>0.01</td>
</tr>
<tr>
<td>Surface Energy (J/m(^2))</td>
<td>0.03</td>
<td>0.03</td>
</tr>
<tr>
<td>Coefficient of Restitution (-)</td>
<td>0.85</td>
<td>0.65</td>
</tr>
<tr>
<td>Surface Coverage Ratio (-)</td>
<td>0.03</td>
<td>0.03</td>
</tr>
</tbody>
</table>
Basic and detailed analysis and post processing were performed using EDEM integration scheme with a time step of 30 ns. A total of over 300 independent simulations were performed to represent handling conditions and is in line with our previous work [27, 28] were studied. The velocity range (0.6–1.6 m/s) was chosen of 0.5 and an impact angle of 75°.

Hence head on collisions between the two particles correspond to an angle of 0° and grazing impacts correspond to 90°.

Binary collisions between adhesive units with varying angles (15°, 30°, 45°, 60° and 75°) and velocity (0.6, 0.8, 1.0, 1.2, 1.4, and 1.6 m/s) [27, 28] were studied. The velocity range (0.6–1.6 m/s) was chosen to represent handling conditions and is in line with our previous works [15, 16, 22]. The procedure was repeated for different SCRs and shapes of fine particles. A total of over 300 independent simulations were performed.

DEM modelling and simulation were performed using the Altair® EDEM™ 2021 bulk material simulation software package. A Verlet integration scheme with a time step of 1 ns was used to maintain numerical consistency. The initial setup was created using Matlab and Visual Basic and detailed analysis and post processing were performed using MATLAB® and the graphs, linear regression and statistical analysis were performed using Graphpad Prism. Total force, position and velocity of particles were extracted for analysis.

2.4. Analysis

The total force $F_{\text{tot}}$ experienced by one of the carrier particles (carrier A in Fig. 2) was recorded from the simulation. A unit normal vector $\hat{n}$ along $d = x_A - x_B$ (where $x_A$ and $x_B$ represent the location of particle A and B) and a unit tangent vector $\hat{t}$ in the $xy$ plane were introduced (Fig. 2). These enabled the total force to be decomposed into normal ($F_n$) and tangential ($F_t$) components,

$$F_n = F_{\text{tot}} \cdot \hat{n},$$

$$F_t = F_{\text{tot}} \cdot \hat{t}.$$  

The effective coefficient of friction ($\mu$) could be represented by,

$$\mu = \frac{F_t}{F_n},$$

and was extracted as the slope of the graph of $F_t$ vs. $F_n$. This procedure was considered satisfactory in the present context, because the adhered fines precluded direct interaction between the carriers and, as a result, sticking was not observed.

2.5. Statistical analysis

To establish an understanding of the data trends and the overall behaviour of $\mu$ with changing impact angle ($\alpha$), SCR and shape of fine particles; two-way ANNOVA followed by Tukey’s multiple comparison test was performed with the level of significance fixed at 0.05. While calculating the coefficient of friction ($\mu$), relative velocities above 1 m/s were considered (as explained further in Section 3.3). The standard deviation calculated from the three velocities exceeding 1 m/s is represented as error bars in the plots.

3. Results and discussion

Micromechanical studies of adhesive units during oblique impact provide insights into the effective frictional behaviour between the units and aid the development of a holistic multiscale model to improve the understanding of adhesive unit dynamics both during manufacturing and handling. It also throws light on the complex behaviour of adhesive units and their departure from a simplistic elastic collision with adhesion as studied by Thornton et al. [47]. The implications could be extended to further studying the interaction with a hopper wall and the dynamics inside inhalers, capsules and blisters as a few examples.

In order to quantify and predict the dynamics of the adhesive units during oblique collision, the normal and tangential forces between them were determined and the coefficient of friction was calculated and compared.
3.1. Evolution of contact forces with time

Fig. 3 shows the variation of normal and tangential forces with respect to time as measured during collision between adhesive units. The representative systems shown in Fig. 3a, b and c represents adhesive units with an SCR of 0.5, an impact angle $\alpha$ of $45^\circ$ and a relative impact velocity of $1.2 \, \text{m/s}$ with (a) spherical, (b) triangular bipyramidal and (c) tetrahedral fine particles.

It could be observed from Fig. 3a that the collision between adhesive units with spherical fine particles results in multiple peaks, henceforth termed events, in the normal and tangential force in the time range of 0.2 to 0.4 $\mu$s. The smaller peaks are characterized as minor events and are likely due to dynamic interlocking of the fine particles between the carrier particles at the point of initial contact. The minor events are analogous to a stick–slip behaviour. The event with longest time span and highest value of the normal force is most significant and hence characterized as the major event. An SCR of 0.5 is sufficient to preclude direct interaction between carriers, and hence this event represents rolling of fines on the surface of the carriers analogously to a ball bearing.

Fig. 3b represents the variation in force for adhesive units with triangular bipyramidal fine particles. Multiple minor events and one major event could be observed between 0.5 and 0.7 $\mu$s. The minor event is characterized by the initial interaction of the fine particles resulting in a stick–slip type behaviour. The major event (0.6–0.7 $\mu$s) is representative of rolling of the fine particles between the interacting carrier particles.

Lastly, Fig. 3c represents the force due to collision of adhesive units with tetrahedron fine particles. There are no significant minor events but a single major event is observed with a tangential force that is significantly higher than that for the spherical and triangular bipyramidal fines. This results is a direct consequence of the particle shape; tetrahedral particles are expected to interlock due to the presence of ridges and valleys to a larger extent than the rounder triangular bipyramidal particles and the spherical particles.

In summary, there is a significant effect of the shape of the fine particles on the magnitude of the tangential force. The tangential forces are lowest for the spherical fines, intermediate for the triangular bipyramidal fines and highest for the tetrahedral fines. This is expected, since composite particles tend to approach a spherical shape when formed from a larger number of spheres, as for the triangular bipyramidal fines compared to the tetrahedral fines. The observations stated above were consistent across the entire data set for the given shape of fines on the adhesive unit. The major event was captured and analysed further to understand the parametric dependence of the coefficient of friction on shape, SCR and collision velocity.

3.2. Calculation of coefficient of friction

The effective coefficient of friction ($\mu$) is defined as the slope of the curve between the normal and tangential force. As explained in Section 3.1, the analysis was restricted to data corresponding to the major events, characterized by the largest normal force and generally also the longest duration. In Fig. 4, the tangential force is plotted vs. the normal force for the major events in Fig. 3. A linear regression line was fitted to each of the data sets, with a regression coefficient ($R^2$) close to 0.99, and the slope of the curve was recorded. It could be observed that adhesive units with spherical fines have the smallest coefficient of friction followed by triangular bipyramidal and tetrahedral as anticipated from the results presented in Fig. 3 and elaborated upon in the following sections.

3.3. Effect of velocity

The coefficient of friction was calculated for the major events for the interactions between adhesive units during oblique impact. Variations with the impact angle, SCR and shape of fine particles were also noted. It was observed that the adhesive units tended to stick to each other
for low relative impact velocity ($\leq 1 \text{ m/s}$) as illustrated in Fig. 5. In these cases, other forces than friction dominate the response and it is not meaningful to extract any friction coefficients. The particles stick because the initial kinetic energy cannot compensate for the energy loss caused by breakage of adhesive bonds, interlocking and rearrangement of particles. It was observed from the data set that a relative impact velocity greater than $1 \text{ m/s}$ ensured that the adhesive units moved apart after the collision. The change in coefficient of friction was minimal for higher relative impact velocities as could be seen from the representative data set displayed in Fig. 6. In contrast, normal restitution coefficients for head-on impact between adhesive units with spherical fines generally increase with increasing velocity in this range, irrespective of the SCR [17]. Such a behaviour is also observed for triangular bipyramidal and tetrahedral fines at low SCRs (up to about 0.5), until larger interlinked chains (contact chains/force chains) of fines as formed [16]. Ultimately, these results have their roots in the contact law governing the behaviour of binary particle interaction: For normal interactions, contact damping and cohesion result in a velocity-dependent energy loss. However, for tangential interactions, the energy loss caused by contact damping and friction is velocity independent.

To further process the data, the mean and standard deviation of the friction coefficients obtained at 1.2, 1.4 and 1.6 m/s were calculated.

### 3.4. Impact of shape and impact angle

Fig. 7 shows the effect of the impact angle ($\alpha$) on the coefficient of friction. Each data point represents the mean value obtained for higher velocity as elucidated in Section 3.3 and the error bars represent the corresponding standard deviations. In order to better represent and interpret the data, Tukey multiple comparison tests were performed as discussed below.

#### 3.4.1. Spherical fines

Fig. 7a shows how the friction coefficient varies with impact angle for spherical fine particles. No significant difference was observed across SCR for the same impact angle (Table 2). A pattern emerges when comparing the changes across impact angles for the same SCR (Table 3). The adhesive units with an $\alpha$ of 15°, 30° and 45° have similar friction coefficients whereas the friction coefficients for 60° and 75° are significantly higher. The data set can thus be separated into two clusters, one characterized by relatively low friction coefficients obtained for head-on-like impacts ($\alpha \leq 45^\circ$) and the other characterized by larger friction coefficients obtained for grazing-like impact ($\alpha \geq 60^\circ$) with larger interaction between the fine particles.

#### Table 2

<table>
<thead>
<tr>
<th>Angle (°)</th>
<th>SCR 0.5</th>
<th>SCR 0.7</th>
<th>SCR 1</th>
</tr>
</thead>
<tbody>
<tr>
<td>0° – – T</td>
<td>– – S</td>
<td>– – S</td>
<td>– – S</td>
</tr>
<tr>
<td>30 – T</td>
<td>– – S</td>
<td>– – S</td>
<td>– – S</td>
</tr>
<tr>
<td>45 – T</td>
<td>– – S</td>
<td>– – S</td>
<td>– – S</td>
</tr>
<tr>
<td>60 – T</td>
<td>– – S</td>
<td>– – S</td>
<td>– – S</td>
</tr>
<tr>
<td>75 B</td>
<td>– – S</td>
<td>– – S</td>
<td>– – S</td>
</tr>
</tbody>
</table>

#### Table 3

<table>
<thead>
<tr>
<th>Angle (°)</th>
<th>SCR 0.5</th>
<th>SCR 0.7</th>
<th>SCR 1</th>
</tr>
</thead>
<tbody>
<tr>
<td>0° – – T</td>
<td>– – S</td>
<td>– – S</td>
<td>– – S</td>
</tr>
<tr>
<td>30 – T</td>
<td>– – S</td>
<td>– – S</td>
<td>– – S</td>
</tr>
<tr>
<td>45 – T</td>
<td>– – S</td>
<td>– – S</td>
<td>– – S</td>
</tr>
<tr>
<td>60 – T</td>
<td>– – S</td>
<td>– – S</td>
<td>– – S</td>
</tr>
</tbody>
</table>

These results can be contrasted to the one obtained for (considerably larger) bare spheres. For these, the effective friction coefficient exhibits a nearly linear increase with impact angle for small angles [23,25] in the sticking regime, followed by an angle-independent value for larger angles in the sliding regime. The much lower effective friction coefficients observed for adhesive units in the current study are most likely due to the fact that interaction is mediated via the fine (spheres) and that, as a consequence, sticking does not occur.

On comparing adhesive units with different SCRs and different impact angles, a similar behaviour as explained in the previous paragraph is found. Significant differences are observed between the grazing angle and head on angle on comparing among SCR values for 0.5, 0.7 and 1. A distinct effect is observed for extreme angles such as an $\alpha$ of 75°. One possible explanation for this behaviour is that the region of interaction increases with increasing impact angle, and that the rearrangement of more fines are involved for larger SCRs, leading to an increased friction coefficient.

#### 3.4.2. Triangular bipyramidal fines

Fig. 7b shows the variation of coefficient of friction with impact angle for adhesive units with triangular bipyramidal fine particles. On comparing the dependence of $\mu$ on SCR for the same value of $\alpha$ there were significant changes observed which was corroborated through ANOVA and Tukey’s test (Table 2). The angles close to grazing angle such as 60° and 75° result in a higher coefficient of friction compared to the smaller angles (Table 3). The larger angles increase the contact region and therefore involve more fine particles, resulting in stronger interlocking and thus higher resistance to the motion.

On comparing adhesive units as a function of SCR and $\alpha$, a pattern similar to that for adhesive units with spherical fine particles was observed with higher friction coefficient. The grazing-like impacts (60° and 75°) result in significantly different friction coefficient and the 45° impact angle behaves either as a grazing-like or an head-on-like angle. The triangular bipyramidal fine particles offer a higher resistance than spherical particles as their tendency to roll reduces which results in a higher coefficient of friction.

#### 3.4.3. Tetrahedral fines

Fig. 7c shows the variation of coefficient of friction with changing impact angle for different SCRs for adhesive units containing tetrahedral fine particles. On comparing the $\alpha$ across each SCR it could be...
observed that for an SCR of 0.5, the friction coefficients obtained for impact angles of 15° and 30° are lower compared to those obtained for higher angles (Table 3). In contrast, for SCRs of 0.7 and 1, a difference is observed only for comparison with an impact angle of 75°.

Significant differences between coefficients of friction obtained for different SCRs (0.5 vs. 0.75 and 0.75 vs. 1) are obtained for some impact angles \( \alpha \) (Table 2). There is some tendency for the differences in \( \mu \) to increase with an increase in the impact angle. On cross comparing \( \alpha \), a similar trend as that within the SCR is observed. The variation in friction coefficient is not as distinct as that for adhesive unit with spherical fines. One possible explanation for this is that the tetrahedral fines tend to interlock more compared to that of triangular bipyramidal and especially spherical fine particles for all impact angles. It could also be noted from Fig. 3 that adhesive units with tetrahedral fines only have a major event as opposed to those with the spherical and triangular bipyramidal particles, for which a number of minor events were also observed.

3.4.4. Cross comparison

Fig. 8 shows \( \mu \) vs. \( \alpha \) for all SCRs and fine particle shapes. Adhesive units with spherical fines have a lower coefficient of friction compared to the triangular bipyramidal and tetrahedral fines for \( \alpha \leq 45^\circ \) (head on-like collisions). The reason is that tetrahedral and triangular bipyramidal fines have ridges and valley and require a higher torque to initiate the rolling motion as compared to spherical particles and in increased tendency to interlock. On comparing across the different shapes of fine particles it could be observed that the maximum value for coefficient of friction is attained for tetrahedral followed by triangular bipyramidal and spherical fine particles. This is also observed in Fig. 3 where adhesive units with tetrahedral fines have a single event compared to units with spherical and triangular bipyramidal fines which have multiple smaller events followed by a larger event before the particles start to roll. For higher values of SCR and \( \alpha \geq 60^\circ \) (grazing-like angles) \( \mu \) tends to merge with one another. Spherical fine particles tend to form smaller aggregates which effectively behaves as composite particles, which makes the effect of shape minimal between spherical and triangular bipyramidal particles. As could be observed from Table 4, the tetrahedral particles have higher \( \mu \) because of their nonspherical shape.

3.5. Self agglomerates

It was observed that near grazing impact between adhesive units with an SCR of 1 resulted in a peeling of the fine particle envelope from the surface of carrier. The peeling predominantly took place between the two interacting carriers and the particles that were peeled off tended to cohere. The peeling effect could be observed distinctly for tetrahedron followed by triangular bipyramidal and spherical fine particles as illustrated in Fig. 9. The particles peeled off the adhesive units formed smaller self agglomerates. Such self agglomerates could potentially coalesce to form larger self agglomerates of the type observed by Rudén et al. [6] during the formulation of adhesive mixtures with high drug loads. With increasing drug loads, corresponding to higher number density of fines and higher SCRs, secondary and tertiary layers are formed. This results in the formation of larger interlinked contact or force chains [22] so that the particles in the adhered layer start to act cooperatively, explaining the increased tendency of the fine particle envelope to peel off.

4. Conclusion

Oblique impacts between adhesive units were studied as a means to extract their effective friction coefficient. The effect of fine-particle shape, impact angle, surface coverage ratio and collision velocity was assessed. The fine particles ranged from spheres via triangular bipyramids (formed from 5 spheres) with a relatively small deviation from a spherical shape (in relative terms) to tetrahedra (formed from 4 spheres) with a significant deviation. Simulations were performed for an impact angle between 15° and 75° and three different surface coverage ratios (0.5, 0.7 and 1) for a fixed surface energy of (0.03 J/m²) in a handling velocity regime (0.6 – 1.6 m/s).

<table>
<thead>
<tr>
<th>Angle (°)</th>
<th>Comparison</th>
<th>S vs. T</th>
<th>S vs. B</th>
<th>T vs. B</th>
</tr>
</thead>
<tbody>
<tr>
<td>15</td>
<td>S vs. T</td>
<td>0.5, 0.7, 1</td>
<td>0.5, 0.7, 1</td>
<td>0.5, 1</td>
</tr>
<tr>
<td>30</td>
<td>S vs. T</td>
<td>0.5, 0.7, 1</td>
<td>0.5, 0.7, 1</td>
<td>0.5, 1</td>
</tr>
<tr>
<td>45</td>
<td>S vs. T</td>
<td>0.5, 0.7, 1</td>
<td>0.5, 0.7, 1</td>
<td>0.5, 1</td>
</tr>
<tr>
<td>60</td>
<td>T vs. B</td>
<td>0.5, 0.7</td>
<td>0.5, 1</td>
<td></td>
</tr>
<tr>
<td>75</td>
<td>T vs. B</td>
<td>0.5, 0.7</td>
<td>0.5, 1</td>
<td></td>
</tr>
</tbody>
</table>
It was observed that the adhesive units stick to each other for low impact velocity and that similar force profiles were obtained for higher velocity, demonstrating that the coefficient of friction is independent of velocity in the non-sticking velocity range. The presence of fines significantly reduced the friction coefficient compared to bare carriers but no tendency for increased reduction with increased SCR was observed, indicating that a saturation is achieved already at an SCR of 0.5.

The impact angle and the fine-particle shape have significant effects on the coefficient of friction. The highest friction coefficient was observed for near-grazing impacts (75°) and the lowest for near-head on impacts (15°). Tetrahedral particles provided highest resistance followed by triangular bipyramidal and spherical fine particles. The tendency of particles to roll and the number of fine particles involved in the interaction dictate the ability to lower the friction coefficient.

It was finally observed that part of the adhered particle envelope was peeled off as a result of near-grazing impacts of adhesive units with a high SCR. This pealing effect was most evident for tetrahedral fines and may provide the mechanism behind the formation of self agglomerates.

The knowledge of the effect of particle shape and interaction parameters on the coefficient of friction is a crucial micromechanical model which could be used to better understand the particle dynamics during formulation and handling of adhesive mixtures. Our results could also shed further light on the mechanism of self-agglomerate formation and may be used as a basis for a multiscale model of adhesive mixtures.

CRediT authorship contribution statement

Sohan Sarangi: Conceptualization, Methodology, Software, Investigation, Validation, Formal analysis, Data curation, Visualization, Writing – original draft. Göran Frenning: Conceptualization, Resources, Writing – review & editing, Supervision.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

Acknowledgement

This study is a part of the science program of the Swedish Drug Delivery Centre (SweDeliver) and financial support from Vinnova, Sweden (Dnr 2019-00048) is gratefully acknowledged.

References


Fig. 9. Interaction between adhesive units with a surface coverage ratio of 1, tetrahedral fine particles and impact angle of 75° showing formation of self agglomerates of size ∼50 μm.
S. Sarangi and G. Frenning
Powder Technology 414 (2023) 118075


