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**Review Paper** 

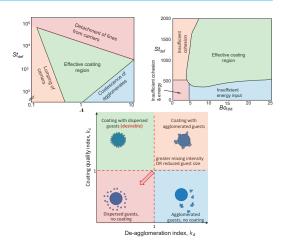
# A Review of Dry Powder Coating: Techniques, Theory, and Applications †

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Dry powder coating, characterised by the blending of poorly flowing powders with finer coating powders to optimise flowability, represents a sophisticated and evolving approach to powder processing. The optimisation of this method involves precise formulation, carefully combining powders with different particle properties to achieve a desirable blend aimed at enhancing the flow characteristics during the application process. Over the last decade, this field has witnessed increasing activity, focusing on key mixing parameters, such as mixer type and mixing power, as well as understanding the influence of constituent powder characteristics, including size ratio, density, and cohesion. Various techniques have been used to assess the flowability improvement or quantify the degree of coating. This review aims to provide a comprehensive exploration of the literature on powder coating research, highlighting its significance in both academic research and industrial applications. This paper discusses current coating analysis techniques using state-of-the-art equipment and reviews recent findings, particularly the nascent attempts to establish regime maps for dry powder coating.

**Keywords:** dry powder coating, adhesion, flowability, powder mixing, powder spreading



### 1. Introduction

Dry powder coating is a process by which material properties are adjusted by coating with a fine powder in order to adjust the surface properties. This review focusses on the dry coating of one powder with this finer powder, which in most cases is used to improve powder flowability. In this context, dry powder coating inherently involves the interaction of (at least) two powder populations, with the aim of ensuring the finer material sticks to the coarser material, so the combined interactions of both types need to be considered. This review therefore summarises these matters in Section 2. Section 3 examines the actual process of coating, considering the mixing mechanisms, coating quantification methods and performance indicators, as well as insights from modelling, environmental effects and coating stability. Finally, Section 4 provides a conclusion of the key matters to be considered in dry powder coating applications and the open questions that remain.

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### 2. Theory and current understanding

### 2.1 Attractive interaction forces

In dry powder systems, particle adhesion occurs primarily through liquid bridges (adsorbed moisture), electrostatic interactions, and van der Waals interactions. The relative force of these depends largely on particle size, although in the fine particle size range of guests used in dry powder coating, the van der Waals attractive forces usually dominate.

### 2.1.1 Liquid bridges

The presence of liquid between two surfaces causes attractive boundary forces to arise due to the surface tension,  $\gamma$ , of the liquid. Even in nominally dry systems, moisture is typically adsorbed on surfaces, causing liquid bridges to form at contact points between particles. The curved nature of the liquid-air interface results in a pressure difference between its concave and convex sides. The capillary pressure can be determined by Laplace's equation:

$$P = \gamma \left( \frac{1}{R_1} - \frac{1}{R_2} \right) \tag{1}$$

where  $R_1$  and  $R_2$  are the principal radii of curvature at the liquid surface, as indicated in **Fig. 1(a)** (Rumpf, 1990). If the external pressure exceeds that within the liquid bridge,



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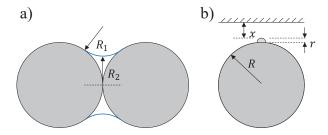


Fig. 1 Schematic diagrams of (a) a liquid bridge between two particles and (b) a particle with an asperity approaching a surface.

the capillary pressure is positive; otherwise, it is negative. The liquid bridge force is defined by the summation of the boundary and capillary forces and depends on the size of the particles, surface tension of the liquid, gap between the two particles, angle of liquid-solid contact, and the half-angle at the centre of the bridge. The latter three parameters depend on the amount of liquid present in the bridge. As noted by Rumpf (1990), the liquid bridge force for identically sized spheres is governed by a linear differential equation for which a closed solution exists only in some cases.

### 2.1.2 Electrostatics

When two dissimilar surfaces contact and separate, electric charge is transferred (Matsusaka et al., 2010). Charge transfer may be due to the transfer of electrons, ions, or material, with several mechanisms and theories proposed, which differ based on conductor-conductor, insulatorconductor and insulator-insulator contacts (Matsusaka and Masuda, 2003). For conductor-conductor contacts, the charge transfer is proportional to the difference in the work function (energy required to remove an electron from inside the material to outside) of the two materials. For insulators, the contact charge is strongly dependent on the density of electrons on the surface (Krupp, 1971). Electrostatic forces can be either attractive (opposite charge) or repulsive (same charge) and decrease at greater separation distances; however, they can exist over longer distances than liquid bridge and van der Waals forces (Rumpf, 1990).

### 2.1.3 van der Waals

Rumpf (1990) considered an ideal representation of rough surfaces as a sphere of radius R, with a hemispherical asperity with radius r, with the asperity centre lying on the particle surface. Taking the Derjaguin (1934) approximation, the van der Waals adhesion force between the sphere and a plate is given by

$$F_{\rm ad} = \frac{A}{6x^2} \left( \frac{rR}{r+R} + \frac{R}{(1+r/x)^2} \right)$$
 (2)

where A is the Hamaker constant and x is the separation distance, as indicated in Fig. 1(b). Rumpf (1990) suggests that if fine particles of  $\sim 10-100$  nm exist between large

particles, as in the case of dry powder coating, the above asperity model is applicable. Rabinovich et al. (2000) adapted this theory to account for the root mean square roughness,  $r_{rms}$ , as shown in **Eqn. (3)**, and further extended it to remove the assumption that the asperity centre lies on the particle surface.

$$F_{\rm ad} = \frac{AR}{6a^2} \left( \frac{1}{1 + R/1.48r_{\rm rms}} + \frac{1}{(1 + 1.48r_{\rm rms}/x)^2} \right)$$
(3)

Johnson et al. (1971) introduced the JKR model, which extended Hertz (1896) theory for normal contact of elastic spheres to include adhesive van der Waals interactions, such that the normal force,

$$F_{\rm n} = \frac{4E^*a^3}{3R^*} - 4\sqrt{\pi \Gamma E^*a^3} \tag{4}$$

where  $E^*$  is the reduced modulus of the contact, a is the contact radius,  $\Gamma$  is the interfacial energy and the reduced radius.

$$R^* = \frac{R_i R_j}{R_i + R_j} \tag{5}$$

where subscripts i and j represent the two contacting particles. Under such adhesive interactions, the two spheres separate when a sufficient 'pull-off force',  $F_{\rm p}$ , acts in the tensile direction:

$$F_{\rm n-IKR} = 3\pi \Gamma R^* \tag{6}$$

In this model, JKR assumes that the adhesive force influences particle deformation. An alternative theory (DMT) was offered by Derjaguin et al. (1975), who proposed that the deformation is driven by Hertz theory, while the adhesive forces act outside of the contact zone (Seville and Wu, 2016). There is no analytical expression for the adhesive force; however, the pull-off force is given by

$$F_{\rm p-DMT} = 4\pi \Gamma R^* \tag{7}$$

Muller et al. (1980) further analysed the two models and proposed that the DMT model is more applicable to small, hard spheres with low surface energy, whereas the JKR model is more applicable to larger, softer spheres with high surface energy. Since dry-coated systems typically consist of very small, hard guests of moderate-high surface energy and moderately sized hosts with variable surface energy and stiffness, it may not always be clear which model is more applicable.

### 2.2 Surface area coverage

For dry powder-coated particles, there are interactions between host-host, guest-host and guest-guest, with the dominating interactions progressing in this order as the guest fraction increases. The surface area coverage (SAC) of the guest particles on the host can be defined from the total projected area of the guest particles on the surface area

of the host particles, as stated by Chen et al. (2009a),

$$SAC = \frac{WD^3 \rho_D}{d^3 \rho_d} \frac{d^2}{4D^2} \times 100\%$$
 (8)

where W is the weight ratio of guest to host particles, d and D are guest and host particle sizes, respectively,  $\rho$  is particle density and subscripts d and D refer to the guest and host particles, respectively. Note that the weight fraction, w, is invariably reported and selected for a dry powder coated system, rather than the weight ratio; however, the two are related by Eqn. (9), and furthermore have very similar values for the low weight fractions typically employed in dry powder coating.

$$W = w / (1 - w) \tag{9}$$

Chen et al. (2009a) established that the critical surface area coverage required to transition from host-host to guest-host dominated contacts,

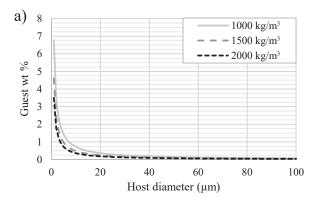
$$SAC_{g-h} = \frac{1.21}{1 + 2(D/d)} \times 100\%$$
 (10)

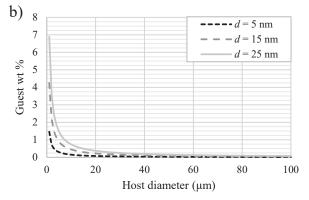
They also proposed that the critical surface area coverage to transition from guest-host to guest-guest dominated contacts depends solely on the geometrical arrangement enabling a single guest on one host particle to contact three equally spaced guests on another host particle, and is thus independent of the properties of the involved particles, and is given by

$$SAC_{g-g} = \frac{\pi}{3\sqrt{6}} \times 100\% \approx 30\%$$
 (11)

Of course, the guest mass fraction required to achieve this surface coverage depends on the particle properties, governed by Eqn. (8) and is demonstrated in Fig. 2. Within the ranges of host particle size and density, and guest particle size and density shown in Fig. 2, it is clear that the typical industrial application of dry powder coating with >0.25 wt% (Conesa et al., 2004) is sufficient to result in guest-guest contacts dominating the interaction for host particles of at least 30 µm. Below this host size, the required guest mass percentage increases quite rapidly, and increases significantly if guest particle size increases beyond 25 nm. Additionally, it is important to note that this analysis assumes that every guest particle is evenly dispersed and distributed among all hosts, i.e., there are no agglomerates of the guests and no multi-layers covering the hosts. In reality, a considerable portion of the mixing effort is given to dispersing these guest agglomerates, as explored in Section 2.3.

Chen et al. (2008) developed the Rabinovich et al. (2000) model to be more relevant for dry powder coating by relating the adhesive force to the surface area coverage, assuming that guests are evenly dispersed across the host surface, arriving at Eqn. (12). Their analysis of this theory





**Fig. 2** Guest mass percentage required to achieve guest–guest dominated contacts (**Eqn. (11))** (a) for Aerosil® R972 as a guest, with d=16 nm and  $\rho_{\rm d}=2650~{\rm kg/m^3}$  and varying host diameter and density and (b) for α-lactose monohydrate as host, with  $\rho_{\rm D}=1530~{\rm kg/m^3}$  and Aerosil as guest, with varying host and guest diameter.

shows that increasing SAC has negligible effect until  $SAC_{g-h}$  (defined by **Eqn. (10)**) is reached, beyond which  $F_{ad}$  reduces dramatically, by approximately two orders of magnitude for the example shown. The adhesive force further reduces, though much less than an order of magnitude, as SAC increases to approximately the value of  $SAC_{g-g}$ , beyond which there is negligible change.

$$F_{\text{ad}} = \frac{Ad}{4x^2} + \frac{A}{24\left(\sqrt{\left(1 + \frac{d}{D}\right)^2 - \frac{1.21}{\text{SAC}}\left(\frac{d}{D}\right)^2} - 1\right)^2 D}$$
(12)

This suggests that  $SAC_{g-g}$  represents the critical SAC that practitioners should aim to achieve to maximise flowability. In contrast, Fulchini et al. (2017) found that optimum flowability occurs at an SAC of 10–20 %, with higher degrees of coverage leading to a gradual reduction in flowability. This reduction in flowability was hypothesised to be caused by the increase in contact between guests, who themselves had poor flowability. Dry powder coating should reduce the host–host contacts, but whether primarily guest–host or guest–guest contacts should be sought remains an open question. It may even be case-dependent, for example, due to the relative flowability or surface properties of the host and guest powders.

### 2.3 Guest dispersion

Nguyen et al. (2015) assessed the de-agglomeration of guests by mixing micronised lactose agglomerates of various sizes with D-Mannitol pellets. The agglomerate fractions continually decreased in the early stages of mixing, with the largest fractions disappearing first. This indicates that bond strength distribution is similar within agglomerates of different sizes, but due to the larger number of guest-guest contacts in larger agglomerates, breakdown is slower. It was also shown that coating of the D-Mannitol pellets is slower when the initial size of guest agglomerates is increased. These two observations indicate that agglomerate breakage primarily occurs by erosion of the surface or removal of small portions from the edge, rather than by agglomerate rupture or fragmentation. Liu et al. (2022) also observed this agglomerate dispersion mechanism for TiO<sub>2</sub> in a rotor-stator in a wet system.

Nguyen et al. (2015) found that breaking down guest agglomerates took most of the time required to reach blend uniformity, suggesting that de-agglomeration of guests is the bottleneck to mixing and coating uniformity. DEM simulations by Deng et al. (2018) supported this finding by showing that the equilibrium of guest-guest contact reduction (indicating guest de-agglomeration) coincides with an increase in guest-host contacts. An increase in surface energy extends the time required to reach this equilibrium point. Above a given surface energy, the detachment energy required to break guest-guest contacts exceeds the average collision energy in guest-host contacts, resulting in an inability to fully disperse individual guest particles and direct them onto the hosts. It can be expected that this threshold surface energy is system dependent and influenced by particle properties and process conditions. A more intense mixing process would enable increasingly cohesive guests to be broken down into single particles (or smaller agglomerates) and directed onto the hosts.

Zheng et al. (2020) examined guest dispersion and coating success based on the de-agglomeration index,  $k_{\rm d}$ , and coating quality index,  $k_{\rm c}$ ,

$$k_{\rm d} = \frac{E_{\rm de}}{E_{\rm kin}} \tag{13}$$

$$k_{\rm c} = \frac{E_{\rm vdW} + E_{\rm p} + E_{\rm de}}{E_{\rm kin}} \tag{14}$$

where E is energy and subscripts de, kin, vdW and p refer to the energy required to de-agglomerate the guest clusters, the relative kinetic energy, the van der Waals energy between the two deformed particles, and the plastic deformation energy of the deformed particles, respectively. For further details on the equations and assumptions, see Zheng et al. (2020). Successful sticking of the guest and host occurs when  $k_c > 1$ , whereas successful de-agglomeration occurs when  $k_d < 1$ . However, both indices decrease as

mixing intensity increases; thus, more intense mixing increases the propensity for effectively dispersing the guests but may ultimately lead to a reduction in surface coverage as guests are easily detached from the host. Reducing guest size significantly increases the values of both indices, meaning that guest dispersion becomes less successful but guest-host sticking is improved, as would intuitively be expected. Zheng et al. (2020) also showed that if the host particle size is close to that of the guest (less than an order of magnitude difference), then the host size influences the coating performance since total detachment, plastic, van der Waals, and (particularly) relative kinetic energy increase with increasing host size, but otherwise it has negligible effect. This analysis highlights the fact that fine guests exhibit a great adhesive force relative to their weight; thus, once stuck to the surface, they are difficult to remove. However, dispersing them effectively to break up cohesive clusters is the primary challenge, and thus, high energy input may be needed. Larger guest particles may more easily detach; thus, the recommended energy input may be lower to prevent their detachment once they initially adhere to the host. Kim et al. (2024a) analysed several silicas of different sizes and surface properties and concluded that finer sizes mean a much smaller mass is required to achieve a given SAC, and a greater reduction in cohesion occurs for the coated powders; however, there is an increased tendency for the guests to exist as agglomerates on the host particle surfaces.

A further implication of the above analysis by Zheng et al. (2020) is that a (narrow) operational window exists within which guest agglomerates are well dispersed and distributed to the hosts, as shown schematically in Fig. 3. Depending on the system considered, there may be no such

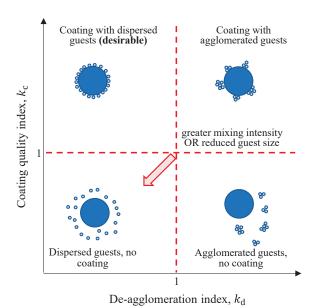


Fig. 3 Window of successful coating operation. Inspired by Kim et al. (2024b).

operational window available within which coating coincides with effective guest dispersion, meaning that coating hosts with guest agglomerates would be the best achievable outcome in such cases.

### 3. Coating

## 3.1 Powder mixing: mechanisms, techniques, and challenges

Powder mixing involves the complex interplay of various mechanisms to achieve homogeneity. Convective, diffusive, and shear mechanisms are the three mechanisms by which powder mixing can occur (Lacey, 1954). Convective mixing relies on bulk movement, where particles are transported through the mixing medium, such as air or a liquid to achieve a more uniform distribution. Diffusion, on the other hand, involves the gradual dispersal of particles due to concentration gradients, which promotes blending at the molecular level. Shear mechanisms occur when forces act upon the powder bed, causing particles to slide and rotate against each other, enhancing mixing. Moreover, based on the type of mixture obtained, Hersey (1975) introduced the concept of ordered mixing. According to Hersey, an ordered mixture is characterised by a standard deviation (SD) of zero across all sample concentrations, given that the sample size exceeds a single ordered unit, in contrast to a random mixture where the SD diminishes as the sample size increases. Later, extending this concept of adhesiondriven ordered mixing, which represents more idealised systems, the term interactive mixture was introduced, which has gained more widespread usage in practical applications (Sallam et al., 1986).

The mixing performance of the components depends on many particle and process parameters (Kaialy, 2016; Venables and Wells, 2001). The consideration of material properties is crucial in powder mixing operations, particularly regarding their cohesive-adhesive nature and the resultant flow characteristics. Free-flowing materials are prone to random mixing and segregation, necessitating the selection of a mixer that prevents dead spots, ensures rapid randomisation, and avoids segregation. Cohesive materials, although mainly poorly flowing, undergo ordered mixing mechanisms and are not subject to segregation. Hence, in such cases, particle size reduction or the addition of adhesive agents may be necessary to enhance mixing. Accordingly, the mixer selection for ordered mixing operations involves avoiding dead spots, breaking down particle agglomerates, and creating ordered mix units, with options ranging from tumbling mixers to high-speed mixer mills.

From an application perspective, most of these powder processing industries often prefer fine particles because of their exponentially enhanced surface areas and surface properties, such as chemistry and roughness. However, this comes with inherent issues of poor flow and nonuniform mixing. Obtaining blend homogeneity, content uniformity,

and reducing batch-to-batch variability in multicomponent mixtures with fine particle fractions is critical. Fundamentally, decreasing the interparticle cohesive and frictional forces appears to be the most effective approach to enhance powder flow and mixing. Dry coating, which is an ordered mixture category, of adhered guest particles on host surfaces serves both the purposes of decreasing cohesivity and friction between particles. The use of dry particle coatings has been associated with the development of diverse functionalities, such as enhanced flowability, content uniformity, and dissolution, which are crucial in pharmaceutical drug delivery systems (Dahmash and Mohammed, 2016; Kim et al., 2023; Qu et al., 2015a). The performance enhancements of the powders and blends are discussed in detail in Section 3.3.

Unlike the traditional convective and diffusive mixing mechanisms required for powder blending, dry particle coating also necessitates shear mixing. From a mechanistic viewpoint, the dry coating process involves the spreading of the guests and the adhesion of the guests to the host surfaces. The spreading and adhesion stages predominantly depend on the particle properties and the high-shear mixing conditions employed. To achieve dry coating, researchers typically use either traditional mixing or milling devices, with more recent applications using external forces to assist mixing. The high-shear mixing offered by milling devices is sought after compared with the majority of low-tomedium shear mixers used in the powder processing industry (Soulier et al., 2022). Some of the mixing devices frequently used for dry coating include low-shear mixers such as rotating drums or Turbula® mixers, high-shear mixers, e.g., the Hosokawa Cyclomix®, Hybridizer®, Mechanofusion® devices, Co-mills, fluidised bed coaters, magnetically assisted impact coater ResonantAcoustic® mixers (RAM), and electrostatic coating devices. These devices are discussed further below, and Table 1 lists some of their advantages and disadvantages when applied to dry coating applications.

### 3.1.1 Low-shear mixers

The rotating drum is the simplest mixer, with a cylinder rotating slowly around a near-horizontal axis, often with baffles or other accessories around the perimeter to ensure that the powder grips to the periphery and does not slip. The Turbula® mixer provides two further planes of motion to minimise segregation that can occur in other low-shear tumblers (Sommier et al., 2001). Applying low-shear mixers to dry powder coating often involves adding grinding balls to the vessel to aid de-agglomeration.

### 3.1.2 High-shear mixers

These devices typically use rotating impellers, rods, or paddles to intensively mix particles through friction with the vessel walls and through impact and shear to break up

Table 1 Summary of c	Summary of dry powder coating methods.	rethods.		
Method	Mixing Intensity	Pros	Cons	Comments
Low-shear mixers	+ (Otles et al., 2011)	<ul> <li>Simple, scalable process suitable for laboratory and industrial scales</li> <li>Effective de-agglomeration using grinding balls</li> </ul>	<ul> <li>Ineffective without grinding media</li> <li>Potential challenges in scaling up due to reliance on convective mixing modes</li> </ul>	
High-shear mixers	++ (Otles et al., 2011)	<ul> <li>Effective mixing via friction and conical vessel design</li> <li>Capable of altering particle morphology for specific applications</li> </ul>	<ul> <li>Potential for undesirable particle shape changes</li> <li>Requires precise control of rotational speed to prevent material degradation</li> </ul>	Applicable to ceramics and composite materials where particle shape modification is advantageous
${ m Hybridizer}^{ m  ext{ iny }}$	+++ (Otles et al., 2011)	<ul> <li>Rapid coating due to high-speed rotor</li> <li>Continuous recirculation ensures thorough processing</li> <li>Short processing times due to high impact forces</li> </ul>	<ul> <li>Batch operation limits scalability</li> <li>Potential temperature build-up requiring careful parameter control</li> </ul>	Delivers high-throughput coating processes
Mechanofusion <sup>®</sup>	+++ (Barling et al., 2015)	<ul> <li>High shear and compressive forces create strong physical and/or chemical bonds</li> <li>Effective coating with minimal temperature increase</li> <li>No substantial particle micronisation or shape change</li> </ul>	<ul> <li>Requires precise control of operational parameters</li> <li>Potential local temperature build-up necessitates careful monitoring</li> </ul>	For advanced materials requiring strong bonds
Co-mill	‡	<ul> <li>Efficient de-agglomeration and coating via high shear stress</li> <li>Continuous process potential with controlled residence time</li> <li>Minimal temperature increase and dust generation</li> </ul>	<ul> <li>Limited effectiveness with a low mean residence time</li> <li>Requires multiple passes or recycling for enhanced coating efficiency</li> <li>Prior blending preferred</li> </ul>	Suitable for processes requiring low dust generation and minimal temperature increase
Fluidised bed coater	+	<ul> <li>Uniform coating due to fluidisation</li> <li>Ideal for small pellets and similar dosage forms</li> </ul>	<ul> <li>High energy consumption due to hot air requirement</li> <li>Long processing times increase operational costs</li> </ul>	Used in pharmaceutical industry for coating small dosage forms
Magnetically Assisted Impact Coater (MAIC)	‡	<ul> <li>Suitable for coating soft organic particles without significant shape or size changes</li> <li>Negligible macroscopic heat generation, suitable for temperature-sensitive powders</li> <li>Operable in both batch and continuous modes, enhancing versatility</li> </ul>	Potential contamination from uncoated magnetic particles     Requires prior blending for optimal performance	Suitable for temperature-sensitive materials
ResonantAcoustic® Mixer (RAM)	‡	<ul> <li>No substantial micronisation or particle shape change</li> <li>Effective mixing via acoustic pressure waves</li> <li>Adjustable mixing intensity for customised processing</li> </ul>	<ul> <li>Potential heating of the powder bed at high accelerations</li> <li>Requires careful analysis of powder properties to avoid melting</li> </ul>	Suited to fine powders and sensitive materials requiring precise mixing intensity control
Electrostatic coating	+	<ul> <li>Capable of coating complex designs</li> <li>Potential for sustained-release pharmaceutical applications</li> </ul>	<ul> <li>Application of plasticisers is required in the pharmaceutical industry to improve material conductivity</li> <li>Requires careful pre-treatment to enhance electrical properties</li> </ul>	Used in pharmaceutical industry for coating tablets with complex designs and sustained-release profiles

agglomerates. One example is the Hosokawa Cyclomix®, which contains several rows of paddles in a frustrum-shaped vessel. The upward motion of the paddles ensures thorough mixing and de-agglomeration, although it can alter particle shape (Ouabbas et al., 2009; Sato et al., 2013).

### 3.1.3 Hybridizer®

Hybridizer® consists of a high-speed rotor with multiple blades, a stator, and a powder-recirculation circuit. The device's capability for high rotation speeds (up to 16,000 rpm) results in a short processing time (Pfeffer et al., 2001). The high rotation speeds of the rotor create high impaction and promote effective de-agglomeration and dispersion of the fine particles, leading to the quick deposition of guest particles onto the surface of the host particles. The recirculating unit continuously moves particles in and out of the processing vessel, thereby enhancing the efficiency. High impact forces due to excessive rotor speeds cause frictional heat build-up, which aids the deposition of guest particles onto the host particle surface by increasing ductility.

### 3.1.4 Mechanofusion® devices

Mechanofusion® uses a cylindrical vessel with an axial shaft and paddles set at alternating angles. The high-speed rotation of the shaft causes the paddles to sweep close to the vessel wall, ensuring turbulent powder movement. This motion induces high shear stress at the paddle face and compressive stress between the paddle and wall, thereby breaking agglomerates and promoting coating. This process is associated with local heat accumulation due to the strong forces acting on the particles, which can result in the fusion of the surface of the host and guest particles. This produces very strong physical and/or chemical bonds, which enhance the coating process (Pfeffer et al., 2001; Yokoyama et al., 1987).

### 3.1.5 Co-Mills

The Co-Mill has a conical chamber in which the powder is fed through a hopper. Inside the chamber, a rotating impeller generates a vortex flow that forces the particles through a screen via centrifugal forces. The particles residing between the screen and tip of the impeller are subjected to high shear stresses, leading to the de-agglomeration of host and guest particles (Sharma and Setia, 2019). The optimal residence time ensures a sufficient number of repeated collisions between these particles, resulting in uniform coating. Several studies have investigated the effects of impeller speed, feed flow rate, and screen size on the residence time (Capece et al., 2021; Huang et al., 2015; Mullarney et al., 2011). The screen size determines the desired size reduction, and the finished product is discharged at the bottom of the milling chamber.

### 3.1.6 Fluidised bed coaters

Fluidised bed coating involves fluidisation of both host and guest materials using air, which is introduced via a distributor, to achieve a uniform coating film. This method requires substantial hot air and extended processing times because of the low coating efficiency. Other designs of fluidised bed coaters involve spraying coating material onto a rotating bed (Watano et al., 2004). Drum rotation subjects the particles to high shear and centrifugal forces, which aid breakage of agglomerates (Qian et al., 2001).

### 3.1.7 Magnetically Assisted Impact Coater (MAIC)

MAIC uses electromagnets surrounding the processing vessel to agitate magnetic particles within the powder mixture. These magnetic particles transfer energy to the host and guest particles through collisions, promoting interparticle and particle—wall collisions. This energy transfer results in the de-agglomeration and deposition of guest particles onto host particles (Pfeffer et al., 2001). The magnetic particles are typically coated to prevent contamination. Particle collisions generate some heat on a microscopic scale, but this is negligible on a macroscopic scale, making MAIC suitable for heat-sensitive materials.

### 3.1.8 ResonantAcoustic® Mixer (RAM)

The RAM operates at the resonant frequency of the system to induce microscale turbulence via acoustic pressure waves. This energy transfer promotes de-agglomeration and coating without significant particle shape changes (Sharma and Setia, 2019). The mixing intensity can be controlled by adjusting the energy supplied to the vessel through the load plates. Acceleration of the mixing vessel can be set up to 100 g to generate intense powder movement; however, very high accelerations and prolonged mixing times can cause temperature build-up in the vessel (Soulier et al., 2022).

### 3.1.9 Electrostatic coating

Electrostatic coating involves the charging of particles, atomisation and deposition of charged particles onto a grounded substrate, typically using a pan coater. This is usually followed by heat curing to facilitate film formation (Prasad et al., 2016). Electrostatic coating requires a conductive material to allow for charge differential formation and to promote the adhesion of charged particles onto the substrate. Most pharmaceutical powders have low conductivity; therefore, a liquid plasticiser is typically applied to reduce the electrical resistance of the material, enhancing the effectiveness of the electrostatic coating process (Yang et al., 2015).

In general, milling devices generate the highest shear forces due to their specialised design for milling and blending. On the other hand, mixing equipment, like drum blenders, MAIC, and Turbula<sup>®</sup>, produce comparatively low

to moderate shear for coating. RAM systems utilise acoustic energy for mixing with varying shear levels.

As touched upon in Section 2.3, in addition to the commonly observed challenges while handling fine particle systems, particle de-agglomeration at the beginning of the mixing operation is a critical but often overlooked step. Ideally, fine cohesive particles or aggregates of particles should be dispersed into their primary particles to achieve good quality mixing and coating (with high SAC). Hence, a two-step mixing process is often used when dealing with mixtures of guest-host compositions. Thus, an additional pre-treatment step of 'pre-blending' the guest and host particles can be employed. Pre-blending predominantly helps with the de-agglomeration of the guest particles to offer a higher degree of initial guest-host interaction within the powder blend. A few studies on dry coating to improve host particle properties are listed in Table 2. This operation is of course an additional process step that can bring complexity to the coating operation; however, in most cases, it is found that its benefits overcompensate for the added process complexity (Tamadondar et al., 2018). Overall, the

dry coating and mixing can lead to the formation of complex coating structures on the particles, which may have significant effects on their bulk behaviour during compaction and in other applications. Recently, Yoshida et al. (2024), from their work on the admixtures of spherical silica particles of two significantly different sizes, showed that the coating structures on large silica particles are affected by compression forces by influencing the agglomeration of finer particles.

The different stages of the dry coating process, which describe two distinct coating scenarios, are depicted in Fig. 4. During mixing, the guest and host particles deagglomerate, with some initial adhesion. Subsequently, this is followed by intimate guest–host collisions and the spreading of coating material on the host surface. Overall, these stages guide the extent of surface coating. The adhesion of the coating composite on the surface is responsible for the coating strength. However, guests that can bind more strongly to the hosts are often more prone to forming cohesive guest clusters. Thus, as mentioned previously, the initial state of the component particles is critical; hence,

Table 2 Examples of dry coatings using pre-treatment approaches.

Method	Components	Pre-treatment	Comments	Reference
Magnetically Assisted Impaction Coating (MAIC)	Hosts: corn starch, cellulose Guest: silica	No	<ul> <li>Poor de-agglomeration of the very fine guest particles</li> <li>Coating efficiency inversely proportional to guest particle size</li> <li>Increased magnetic particle size increased coating</li> </ul>	(Ramlakhan et al., 2000)
Mechanofusion®	Hosts: salbutamol sulphate, salmeterol xinafoate, triamcinolone acetonide Guest: magnesium stearate	No	<ul> <li>Aerosolisation improved as indicated by higher fine particle fraction and smaller agglomerate sizes</li> </ul>	(Q.T. Zhou et al., 2010)
Co-mill	Hosts: ibuprofen-50, mannitol, lactose Guests: Aerosil R972, Aerosil 200	Pre-mixing in V-blender	Bulk density and flow improvement without significant attrition	(Mullarney et al., 2011)
Co-mill	Host: Avicel PH105 Guest: Cabosil M5P	Geometric dilution and Sieving	<ul> <li>40 co-milling cycles required for optimised SAC</li> <li>Tabletability (tensile strength) inferior for nano- coated powder as compared to uncoated powder</li> </ul>	(Chattoraj et al., 2011)
Fluid Energy mill (FEM)	Hosts: ibuprofen110, PVP 40 Guests: Cabosil M5P, Aerosil R972	Pre-mixing in V-blender	Simultaneous milling and dry coating achieved with improved flow dissolution and less static charge accumulation during milling	(Han et al., 2011)
ResonantAcoustic® mixer (LabRAM)	Host: acetaminophen, ibuprofen, ascorbic acid, lactose, starch Guest: silica grades	No	Dry coating improved powder bed porosity	(Capece et al., 2014)

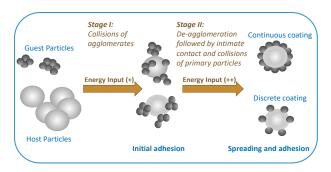


Fig. 4 Two-step dry coating process.

particle de-agglomeration at the initial stages appears to be the rate-limiting step in achieving a guest-host type of adhesive mixing (Deng et al., 2018; Nguyen et al., 2015). There are different approaches utilised to minimise or circumvent the issue of particle de-agglomeration, employing different de-agglomeration strategies like sieving, aerodynamic dispersion, ultrasonication etc., as a pre-treatment protocol before dry coating. Some of the strategies used are listed in Table 2.

### 3.2 Methods to quantify coatings

In the preceding sections, we extensively explored the significance of coating and compared various coating techniques. However, it's imperative to quantify coating efficiency to optimise systems and operational costs. This section discusses different techniques for coating quantification.

### 3.2.1 Functionality analysis

The primary purpose of dry coating is to improve functionality. Therefore, measuring flow properties such as flowability and dispersity, and bulk properties such as bulk density, size distribution, and porosity can serve as valuable indicators of coating efficiency. By dispersing guest particles within the bulk of cohesive powder through mixing, the contact between host particles is minimised, thus enhancing overall powder flowability, dispersity, and fluidisability (Zhou et al., 2010). Similarly, this can influence particle packing, affecting the bed porosity, bulk density, and particle size distribution (Ghoroi et al., 2013b).

Numerous studies examining powder flowability have consistently observed a decrease in the angle of repose for coated powders (Ghoroi et al., 2013b; Yang et al., 2005). Additionally, shear cell tests have been employed to quantify coating efficiency by calculating the flow function coefficient (FFC), showcasing an enhancement from poor flowing (FFC < 4) to easy flowing (FFC > 4) for coated powders (Davé et al., 2022; Ghoroi et al., 2013b; Kim et al., 2023). Fluidisability tests conducted via aeration have further demonstrated that well-coated powders exhibit increased fluidity owing to reduced interparticle cohesive

forces (Zhou et al., 2010).

Moreover, due to the introduction of fine particles between host particles, coated powders typically exhibit higher bulk and tapped density than uncoated powders (Castellanos, 2005; Ghoroi et al., 2013a, 2013b; Zhou et al., 2010). Furthermore, porosity is another property affected by particle surface modification. Surface modification through coating reduces van der Waals forces, leading to a more densely packed bed with reduced porosity. Capece et al. (2014) investigated this phenomenon using pharmaceutical powders and reported up to a 50 % reduction in porosity for coated powders, depending on the size of the guest material. Furthermore, particle size distribution can be used as an indicator of coating because deagglomeration of cohesive clusters can cause a shift in the size distribution of the powder bed (Liu et al., 2021). Soulier et al. (2022) examined the particle size distribution before coating, finding two distinct peak sizes corresponding to the guest and host particles. Upon mixing, the distribution shifted to a mono-modal size distribution like that of host particles, implying de-agglomeration, dispersion, and attachment of guest particles onto host particles.

Despite providing indicative measures of flowability, this method lacks detailed information on coated particle surfaces. Determining the dispersion of guest material or the extent to which they are attached to individual host particles remains a subject of interest.

### 3.2.2 Surface imaging

To gain deeper insight into the powder coating, particle surface profiles can be examined using various imaging techniques. These are described below.

# 3.2.2.1 Scanning Electron Microscopy (SEM) and Energy Dispersive X-ray Spectroscopy (EDX)

SEM has been extensively employed as a qualitative method for determining the quality of coating (Dahmash and Mohammed, 2016; Ghoroi et al., 2013b; Ishizaka et al., 1989; Qu et al., 2015a; Weiss et al., 2015; Zhou et al., 2010). In combination with EDX, this technique can detect the elemental composition of a particle surface, which can be an indicator of the presence of guest material on the host particles (Chen et al., 2009c; Ghoroi et al., 2013a, 2013b).

While this technique primarily serves as a qualitative measure of coating efficiency, other researchers have introduced image analysis algorithms to quantitatively analyse surface coverage (Lecoq et al., 2015). However, the two-dimensional nature of SEM images limits the calculation accuracy. A recent advancement by Friebel et al. (2024), with the aid of computed tomography, developed an image processing workflow to generate three-dimensional images. Leveraging machine learning models, this technique enabled the segmentation of host and guest particles and the determination of coating coverage and thickness.

However, this was not without assumptions such as uniform coating on the unscanned side of the particles.

### 3.2.2.2 X-ray Photoelectron Spectroscopy (XPS)

X-ray Photoelectron Spectroscopy (XPS) involves irradiating a sample with high-energy X-rays, causing the ejection of core-level electrons through the photoelectric effect. These ejected photoelectrons are then detected and analysed to determine their kinetic energy, which provides insights into the elemental composition, chemical state, and bonding environment of the sample surface. Given that the guest materials used for coating can be on the nanoscale, this technique is more advantageous over SEM-EDX because it can detect the presence of guest materials on host particles at shallow depths of 3-10 nm without a challenge of the beam passing through the guest particle to the substrate. XPS was used to detect magnesium stearate used as a coating material for ibuprofen (Qu et al., 2015a) and lactose (Q. Zhou et al. 2011). In both cases, Mg was not detected for the uncoated powder, whereas for the coated powder, the composition and depth of the Mg coating were obtained.

## 3.2.2.3 Fourier Transform Infrared Spectroscopy (FTIR)

This mechanochemical technique is used to assess the quality and composition of dry powder coatings. By directing infrared light through a powder sample, FTIR spectroscopy can identify the characteristic absorption patterns associated with molecular vibrations within the coating material. These spectral fingerprints provide valuable insights into the chemical composition, functional group presence, and coating integrity (Fransisco and Fransisco, 2018; Ishizaka et al., 1989). Pfeffer et al. (2001) used FTIR spectroscopy to investigate changes in hydroxyl (OH) groups before and after coating of cornstarch and cellulose materials coated with silica. They observed reductions in the OH group absorbance upon coating application, indicating altered surface properties. Notably, variations in absorbance levels were correlated with processing time, indicating time-dependent effects on OH group availability. Similarly, Dahmash et al. (2018) evaluated the properties and molecular interactions of ibuprofen powder (guest material) and microcrystalline cellulose (MCC) in dry particle coating applications. The coated powder exhibited reduced carbonyl band intensity, indicating the formation of hydrogen bonds between ibuprofen and MCC.

### 3.2.2.4 Atomic Force Microscopy (AFM)

Atomic Force Microscopy (AFM) is a precise method for determining the coating of guest material on host particles in dry powder coating processes. By scanning the surface of coated particles with an AFM probe, the topographic features of the particles at nanoscale resolution can be de-

tected. The presence of a coating layer alters the surface morphology of the host particles, leading to changes in the surface roughness, thickness, and particle size distribution. AFM enabled the direct observation of these changes, allowing the quantification of coating thickness and uniformity. Additionally, AFM can probe mechanical properties such as adhesion strength between the host and guest materials, providing insights into the quality and integrity of the coating (Cui et al., 2014; Traini et al., 2007; Weiss et al., 2015). Overall, AFM serves as a valuable tool for characterising coated particles, offering detailed information about their surface structure, morphology, and mechanical properties, essential for optimising dry powder coating processes in various industrial applications (Shur and Price, 2012).

Although imaging techniques offer valuable insights into coating efficiency, they are not without drawbacks. One major limitation is the small sample which can be assessed compared to functionality tests, which makes it challenging to extrapolate observations to the entire powder sample. Additionally, depending on particle size, imaging may necessitate high-resolution or specialised instruments, which can be costly.

### 3.2.3 Colourimetry

Coloured tracers have been used in studies to assess blend uniformity or to investigate powder flow mechanisms. Through the analysis of colour variations in the blend, researchers have gained insights into its homogeneity (Legoix et al., 2017). For powder coating, colourimetry is a valuable technique in which contrasting colours of guest and host materials result in a blend displaying a colour intermediate between the two. A colorimeter is used to track colour changes in the mixed powder by detecting alterations in chroma (colour concentration) and luminance. These changes indicate how effectively the guest particles have dispersed and adhered to the host particles (Emady et al., 2015).

The integration of colourimetry in dry coating studies has provided invaluable insights, often visualised through the sophisticated CLE (Colour, Lightness, and Chroma Evaluation) colour space. For instance, Nguyen's work (Nguyen et al., 2016) reported dark pink host particles contrasted with white guest materials to investigate the time required to reach steady coating. Leveraging a monodimensional colour space, the increase of lightness signalled the efficient dispersion of guest particles, implying superior coating performance. Barling et al. (2015) considered beyond colour lightness and considered colour concentration when examining the coating performances of different mixers. Using coloured guest materials, the shifts in chroma and brightness values corresponded, respectively, to the de-agglomeration and dispersion phases crucial to coating efficacy. Further colorimetric analyses have

**Table 3** A summary of different quantification methods.

Quantification method	Advantages	Limitations
Function characterisation	<ul> <li>Bulk measurements hence more representative</li> <li>Can be achieved using simple methods</li> <li>Requires less expertise</li> </ul>	<ul><li> Does not quantify coating</li><li> Intrusive method hence may affect sample</li></ul>
Surface imaging	<ul> <li>Provides the surface details</li> <li>Can quantify coating</li> <li>Can provide details about the coating thickness</li> </ul>	<ul> <li>A small amount of sample may not be representative</li> <li>Requires pretreatment of samples</li> <li>Requires expertise</li> <li>Advanced equipment and hence expensive</li> </ul>
Colourimetry	<ul> <li>Uses a large sample quantity so more representative</li> <li>Non-intrusive method</li> <li>Inexpensive</li> </ul>	<ul> <li>Requires samples with contrasting colours</li> <li>Less sensitive</li> </ul>

recently been presented using CIE XYZ colour space, where the influence of mixing speed and time on coating was examined (Karde et al., 2023).

Colourimetry offers advantages over imaging techniques as it analyses a larger portion of the powder sample. However, it has limitations, such as the necessity for contrasting colours of constituent materials. Furthermore, even for powders of different colours, subtle colour changes between mixtures may be challenging to capture accurately using a colorimeter.

### 3.2.4 Other techniques

Another distinctive technique for quantifying coating efficiency is the isoelectric point method. This method involves dispersing coated powders in various buffer solutions with known pH levels and determining the point of zero charge, also known as the isoelectric point. By identifying the isoelectric points of individual powders (both guest and host), the efficiency of the coating in the mixed powder can be assessed by examining where its isoelectric point lies between the two extremes. Although this method is commonly used in other fields, such as colloid science and surface chemistry (Kosmulski, 2002; 2006), it has potential application in dry powder coating. The advantages of the isoelectric point method lie in its cost-effectiveness and the ability to analyse a large amount of powder.

In addition, another method is based on the Brunauer–Emmett–Teller (BET) theory. This method, originally developed for measuring surface area in porous materials, involves analysis of the adsorption and desorption of gas molecules on the powder surface, which enables the calculation of specific surface area and pore volume (Brunauer et al., 1938). This technique has been extensively applied, including to determine the powder coating efficiency. Coated particles involve filling pores with guest particles, which in turn reduces the specific surface area (Qu et al., 2015b).

Furthermore, the technique of Near Infrared spectros-

copy (NIR) technique involves measurements of homogeneity in powder flow. The noise in the NIR spectra represents interference in the flow, which may result from the introduction of the guest material. This method was used to measure the flow consistency of ibuprofen powder and determine the optimum weight percentage of the guest material for a good coating (Ropero et al., 2009).

This section outlines various characterisation techniques tailored to understand particle functionalities and their impacts on coating effectiveness. Acquaintance with these techniques enables researchers to select the most suitable method for quantifying coatings. While conventional flowability characterisation methods like angle of repose (AoR), bulk, and tapped densities remain valuable for identifying the quality of coating, emerging techniques such as surface profile analysis offer additional benefits by quantitatively assessing the extent of dry coating needed to achieve the desired functionality. It is worth noting that surface imaging and scanning methods may not always provide a comprehensive view of coating quality, as they involve only a small sample size. Therefore, distinguishing between good and poor coatings is not always trivial. In summary, the methods discussed here have both advantages and limitations (see Table 3). Therefore, combining different methods can provide a more comprehensive understanding of the coating process.

### 3.3 Insights from modelling

Various numerical approaches have been employed for modelling of the dry powder coating process such as Discrete Element Modelling (Khala et al., 2023a, 2023b, 2023c; Nguyen et al., 2016, 2015; Yang et al., 2013, 2015, 2022) and Population Balance Modelling (PBM) (Alonso et al., 1989). Numerical studies, particularly Discrete Element Method (DEM), allow tracking particle positions at each instant. With this information, many earlier studies evaluated coating by examining the concentration of guest particles in a particular region, collision frequency, mixing

index, and/or calculating the average number of guest particles per host particle (Alonso et al., 1989; Deng et al., 2018; Tamadondar et al., 2018; Zheng et al., 2020). Another advantage of knowing every particle's position is the simplicity with which SAC can be determined. DEM is extensively used to simulate bulk particle flow behaviour by computing the translational and rotational motion of discrete particles over a specified period. However, DEM simulations of dry coating processes demand very high computational costs due to the large number of fine particles involved. Nevertheless, DEM has been used to evaluate the effects of material properties such as cohesion, particle size distribution, particle size, and density ratio on coating efficiency in a bladed mixer (Deng et al., 2018; Khala et al., 2023b). The granular Bond number (Capece et al., 2015), which indicates the adhesion strength of guest particles to host surfaces, is a critical parameter which dictates coating performance

$$Bo = \frac{F_{\rm w}}{F_{\rm c}} \tag{15}$$

where  $F_c$  is the cohesive force between the contacting particles, which is often determined by JKR contact theory using **Eqn.** (16), and  $F_w$  is the harmonic mean of the gravitational forces given by **Eqn.** (17).

$$F_{\rm c} = 1.5\pi R^* \Gamma \tag{16}$$

$$F_{\rm w} = \left(\frac{2m_i m_j}{m_i + m_j}\right) g \tag{17}$$

where m represents the particle weight, g is gravitational acceleration and subscripts i and j represent the two contacting particles. As shown in **Eqns.** (15–17), the granular Bond number is influenced by the particle size, surface energy, and particle mass or density.

A higher granular Bond number indicates stronger adherence of the guest particles to the surface of the host material, whereas a lower Bond number indicates weaker affinity between the guest and host material. The granular Bond number increases with reducing guest particle size and density, leading to improved stickiness of the guest particles on the host surface. Conversely, very dense guest particles tend to percolate through the granular bed and settle at the bottom of the mixing vessel (Khala et al., 2023b).

DEM simulations by Khala et al. (2023b) of spherical particles in a bladed mixer revealed that coating performance transitions from poor to good beyond a critical granular Bond number between 160 and 212. The granular Bond number also increases with increasing surface energy, resulting in a higher agglomerate strength or detachment energy. This requires a prolonged period of mixing or higher shear stress to break up the agglomerates and disperse the guest particles. For low surface energies, guest

particles can be easily dispersed; however, the coating may be unstable because of its weak adhesion strength. Deng et al. (2018) concluded that the particle collision energy must exceed the detachment energy of fine particles to improve the mixing quality. The particle size distribution of the host particles can also profoundly affect coating efficiency since the total host surface area available for coating varies with the width of the size distribution. As an alternative to the computationally expensive DEM technique, Alonso et al. (1989) employed a PBM approach to model the timeevolution of the fines distribution on carrier particles based on collision probabilities. The model considers the adhesion of fines to host particles in their neighbourhood, spread of fines throughout the bulk via collisions between coated and uncoated particles, and the rearrangement of guest particles over the host's surface. Although PBM is less detailed in terms of physical interactions than DEM, it is useful for understanding the effects of guest concentration and host size ratio on coating efficiency. However, there are some limitations to the model, particularly the lack of understanding of the dependence of the model parameters on the physical properties of the powder system.

DEM simulations were also employed to assess the impact of process parameters such as mixer geometry, impeller rotational speed, guest concentration, initial fill location of guest material, fill height, and impeller design on coating efficiency. Dave et al. (2003) investigated particle dynamics in Mechanofusion®, Hybridizer® and Magnetically Assisted Impact Coating (MAIC) mixers. They demonstrated that DEM provides insights into the governing mechanisms of coating in each system based on the particle collision frequency, mixing energy, and impact force. However, the simplification of mixer geometries and the consideration of only host particles in the simulations limit the applicability of the findings of the study, particularly given that deagglomeration of guests has been shown to be the bottleneck (Deng et al., 2018; Nguyen et al., 2015). In high-shear mixers, higher rotational speeds are typically associated with higher shear stresses, which promote breakage of guest particle agglomerates and spreading (Khala et al., 2023c; Serris et al., 2013). However, excessive impeller rotational speeds can fluidise the powder bed, reducing the collisional frequency required for optimal coating performance (Khala et al., 2023c) or inducing particle attrition, which can be detrimental to coating efficiency (Sato et al., 2013). Similarly, higher vibration intensities are desirable for vibrating systems, particularly for highly cohesive guest materials (Yasunaga et al., 2022; Zheng et al., 2022). The guest particle loading and the initial location of the guest material can also affect the dry coating efficiency. High guest loading leads to an increase in the mixing time required to reach mixture equilibrium due to the increased number of particle contacts (Deng et al., 2018; Khala et al., 2023c). An increased guest fraction leads to higher overall surface area coverage but less efficient use of the guest particles, i.e., a lower percentage of the guests stick to the hosts (Deng et al., 2018). For bladed mixers, the coating rate is accelerated when the guest material is initially positioned closest to the impeller zone, which leads to instantaneous rupture of the guest agglomerates (Khala et al., 2023c). Khala et al. (2023c) also demonstrated that high fill ratios can result in mixing dead zones of significant volume; hence, a longer mixing time is required to reach mixing equilibrium.

Regime maps, characterised by dimensionless numbers, have been proposed in the literature to identify the optimal combination of material properties and process parameters for efficient dry coating. Khala et al. (2023a) and Tamadondar et al. (2018) described coating efficiency based on Stokes deformation number,  $St_{\rm def}$ , a dimensionless number expressed as the ratio of mechanical mixing energy,  $E_{\rm m}$ , to the cohesive energy between the particles,  $E_{\rm c}$ ,

$$St_{\text{def}} = \frac{E_{\text{m}}}{E_{\text{c}}} \tag{18}$$

Depending on the coating system, various expressions for the cumulative mixing energy imparted on the particles can be derived from parameters such as power and impeller torque. For DEM simulations, the system kinetic energy calculated from the kinetic energy of discrete particles can be used to define the mixing energy. The cohesive energy of agglomerates can be estimated from the JKR contact theory of adhesion, in which the work of adhesion for a single contact is expressed as

$$E_{\rm c} = 7.09 \left( \frac{\Gamma^5 R^{*5}}{E^{*2}} \right)^{\frac{1}{3}} \tag{19}$$

Khala et al. (2021) developed an alternative expression for estimating cohesive energy from experimental data:

$$E_{c} = \sum_{i=1}^{n} \sum_{j=1}^{m} \Gamma_{ij} f_{sa,i} f_{sa,j} R_{ij}^{*2}$$
(20)

where  $f_{\rm sa}$  is the fractional surface area of each species. Tamadondar et al. (2018) described coating efficiency as a function of a further dimensionless number, referred to as the reduced intermixing coefficient,  $\Lambda$ , which is the ratio of guest–guest interfacial energy to guest–host interfacial energy. This describes particle attribute effects, albeit ignoring particle weight effect.

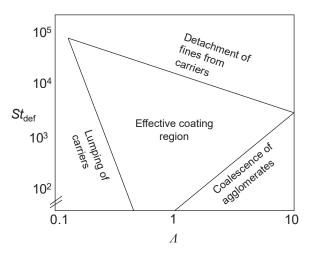
Using simulations of a Couette shear cell, Tamadondar et al. (2018) found that poor mixing occurred at low  $St_{\rm def}$  and low  $\Lambda$  values whilst excessive agglomerate formation occurred at low  $St_{\rm def}$  and high  $\Lambda$  values due to insufficient mixing energy, as shown in Fig. 5. Furthermore, rapid breakage of guest–host contacts occurred at high  $St_{\rm def}$  values due to excessive mixing energy. However, it should be noted that only the particle surface energy was varied in this study; therefore, the results may not be applicable to

coating systems with other particle properties such as size and density ratios.

**Fig. 6** shows the regime map proposed by Khala et al. (2023a), in which surface area coverage is expressed as a function of Stokes deformation number and harmonic mixture Bond number accounting for host–host, guest–guest and host–guest particle interactions, to delineate regions of poor and good coating efficiency,

$$Bo_{\text{HM}} = \left(\sum_{i=1}^{n} \sum_{j=1}^{m} \frac{f_{\text{sa},i} f_{\text{sa},j}}{Bo_{ij}}\right)^{-1}$$
 (21)

where  $Bo_{ij}$  is the granular Bond number between particle species i and j. For low  $Bo_{\rm HM}$ , poor coating efficiency was observed due to weak interparticle forces between the guest and host materials. However, for high  $Bo_{\rm HM}$ , coating performance is determined by the  $St_{\rm def}$ , whereby low  $St_{\rm def}$  implies insufficient mixing energy, leading to poor guest



**Fig. 5** Dry powder coating regime map proposed by Tamadondar et al. (2018), adapted with permission from Tamadondar et al. (2018). Copyright: (2018) Elsevier B.V.

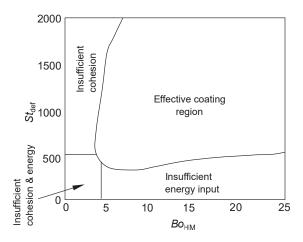


Fig. 6 Dry powder coating regime map proposed by Khala et al. (2023a), adapted from Ref. (Khala et al., 2023a) under the terms of the CC-BY 4.0 licence. Copyright: (2023) The Authors, published by Frontiers.

dispersion, whilst high  $St_{\rm def}$  leads to improved breakage and dispersion of guest agglomerates, resulting in better coating efficiency. However, at very high  $St_{\rm def}$ , guest detachment from host surfaces or demixing can occur, which was not captured in the regime map.

### 3.4 Coating performance

For dry powders, the existence of strong interparticle forces among fine particles often results in several processing and manufacturing problems with respect to their flowability, packing, dispersibility, and compressibility. Poor or variable powder flow and packing can result in various processing problems across powder processing industries, such as incomplete die fill resulting in weight variation and content uniformity problems during tableting and capsule filling operations (Sun, 2010; Tan and Newton, 1990), dispersibility of a dose from dry powder inhalers (DPI's) (Das et al., 2009), and blend uniformity issues during mixing (Chaudhuri et al., 2006). Thus, dry coating of powders plays a crucial role in enhancing their performance and suitability for various applications, including food, pharmaceuticals, and additive manufacturing. This section presents a comprehensive overview of performance testing methodologies for coated powders, focusing on key indicators such as flow, dispersion for dry powder inhalers (DPIs), tabletability, dissolution of active pharmaceutical ingredients (API), and spreading for additive manufacturing. Each performance indicator is discussed in detail, highlighting its importance and relevance in evaluating the quality and functionality of coated powders from the literature. The quantitative techniques for analysing these coating performance indicators and their associated parameters are extensively discussed in Section 3.2.

### 3.4.1 Flow improvement

Powder flow improvement is the most prevalent performance indicator used throughout the literature for assessing dry coating efficiency. The improvement in powder flow through different mechanisms after dry coating has been shown. Specifically, two mechanisms have been identified: the reduction in interparticle cohesion due to increased nanoscale roughness created by guest particles (Chen et al., 2009b; Yang et al., 2005), and the passivation of higher energy sites on the host surface leading to decreased dispersive surface energy (Han et al., 2013). It is also well known that the flow properties of coated powders are crucial for their processability during manufacturing processes such as die filling, mixing, and compaction. A well-flowing powder ensures uniformity and efficiency in downstream processes, preventing issues such as segregation or inconsistent dosing. To highlight these, Davé and co-workers have extensively employed dry particle coating to reduce interparticle cohesion and improve the flow behaviour of pharmaceutical powders (Huang et al., 2017; Kim et al., 2023). Han et al. (2011) reported that the reduction in interparticle cohesion through surface modification of drug particles pre-blended with nanosilica could be achieved simultaneously during the micronisation process using a fluid energy mill. Additionally, Jallo and Dave (2015) also suggested that dry coating could be helpful in reducing the electrostatic charging of micronised powders. A hydrophilic coating was found to be more effective than a hydrophobic coating for electrostatic charge reduction and flow improvement.

### 3.4.2 Content and blend uniformity

Dry particle coating has been recognised as a valuable technique for improving both the content uniformity of tablets and the blend uniformity of powders. The proposed method offers advantages over conventional blending methods in terms of achieving higher content uniformity for the same amount of additive. Studies have shown that dry particle coating can improve blend content uniformity by reducing API cohesion and agglomeration, thus enhancing the overall uniformity of the blend (Huang et al., 2017). Beach et al. (2010) demonstrated the application of dry particle coating to enhance the flow properties of powders, leading to better blend uniformity of API formulations.

Furthermore, research has demonstrated that the use of dry particle coating can lead to improved powder flowability (Mullarney et al., 2011; Yang et al., 2005; Q.T. Zhou et al., 2011), which is essential for ensuring the homogeneity of powder blends and subsequently the content uniformity of tablets. By reducing interparticle cohesion and modifying surface properties, dry particle coating contributes to better blend uniformity, which translates into improved content uniformity in the final tablet product. Additionally, dry coating processes increase the flowability of powders, thereby enhancing the dosing accuracy during tablet production. Recently, Kim et al. (2023) examined the characteristics of multicomponent blends containing low levels of API (1, 3, and 5 wt%) for blend uniformity, flowability, and drug release rates. It was found that dry coating reduced the aspect ratio of API agglomerates and improved blend properties like flow and uniformity, facilitating accurate dosing. Overall, these findings suggest that using finer excipients and dry coating the API can enhance blend uniformity and flow without sacrificing dissolution rates, particularly in low drug-loaded blends of fine APIs.

### 3.4.3 Improved dispersibility

Efficient powder dispersion is essential for dry powder inhaler (DPI) formulations to ensure effective drug delivery to the respiratory system. The powder coating should facilitate optimal de-agglomeration and dispersion upon inhalation, ensuring consistent and targeted drug delivery to the lungs. This surface engineering technique has been applied to improve powder properties for inhalation applications

(Healy et al., 2014; Scherließ et al., 2022; Zhou et al., 2010).

The surface modification through dry coating modifies the cohesive-adhesive balance between the carrier and API particles, improving the drug delivery efficiency (Zhou et al., 2012; Zhou et al., 2010). This study investigated how the inherent interparticle cohesion of model pharmaceutical powders affects their aerosolization from dry powder inhalers. Two cohesive lactose powders with different particle sizes were studied, and dry coating with magnesium stearate markedly improved their flowability, fluidisation, and de-agglomeration behaviours. The reduction of the cohesive forces through surface modification was responsible for these enhancements, underscoring the potential for improving powder aerosolization by decreasing interparticle cohesion. The findings highlight the crucial relationship between intrinsic cohesive properties and bulk characteristics, offering valuable insights for optimising dry powder inhaler formulations and suggesting the potential of mechanical dry coating to enhance the efficiency of fine cohesive powders in aerosolization. The influence of dry coating on the fluidisability of powders was also investigated to determine the tensile strength of the powder bed (Chen et al., 2009a).

Morton and co-workers have demonstrated through their work a significant impact on powder aerosolization or dispersion behaviour after dry coating to improve the drug delivery efficiency of DPI formulations by modifying the cohesive—adhesive balance in the carrier and drug particles (Begat et al., 2004; Jong et al., 2016; Zhou and Morton, 2012; Zhou et al., 2010). In addition to the flow enhancement, Ghoroi et al. (2013b) reported an improvement in the dispersibility of dry-coated pharmaceutical powders by the virtue of a reduced cohesion tendency. Other potential benefits include reducing electrostatic charging in dispersed micronised powders (Jallo and Dave, 2015). Studies have also highlighted the importance of dry particle coating in improving the fluidisation behaviour of cohesive particles, resulting in better powder film consistency and deposition in electrostatic coating processes (Ricks et al., 2002).

### 3.4.4 Tabletability

In tablet manufacturing, the strength of tablets is a critical parameter influencing their integrity, handling, and dissolution characteristics. Tablet hardness, friability, and tensile strength tests are commonly used to evaluate tablet strength. The coating of powders should enhance tablet strength by promoting interparticle adhesion and reducing friability by enabling the use of finer excipients and improving the packing density (Huang et al., 2015). Thus, this ensures robust tablets capable of withstanding handling and shipping without degradation. Previous studies have shown an improved tabletability of the dry-coated API formulation blends, predominantly due to improved flow

and packing characteristics, without compromising the tablet strength expected from the reduced adhesion of the dry-coated APIs (Han et al., 2013; Mullarney et al., 2011). In fact, these studies demonstrated that improved tabletability of the formulations is possible even at high drug loading. Huang et al. (2015) measured the flow and packing enhancements of fine pharmaceutical powder blends consisting of fine excipients and dry-coated micronised acetaminophen. It was found that along with the flow and packing property improvements obtained through dry coating, the presence of fine excipients also enhanced tablet compatibility, indicating its suitability for direct compaction.

### 3.4.5 Wettability and dissolution

The dissolution of active pharmaceutical ingredients (API) from coated powders is critical for controlling drug release kinetics and ensuring therapeutic efficacy. Dissolution testing, utilizing paddle or basket apparatus, evaluates the rate and extent of API release from the coated powder. The coating should not hinder API dissolution but may be tailored to modulate the release profile, enabling controlled and consistent drug release according to therapeutic requirements. Han et al. (2011; 2013) reported improved dissolution of tablets of directly compacted hydrophobic ibuprofen powders coated with hydrophilic silica through a simultaneous milling and coating process. Interestingly, Qu et al. (2015b) found an increase in the dissolution rates of dry-coated ibuprofen powders coated with hydrophobic guests like MgSt and Aerosil R972. The authors attributed this unexpected increase in the dissolution rate to the improved dispersibility of the powders in the dissolution medium. Recently, similar observations on the dissolution improvement of hydrophobically coated fine ibuprofen powders were reported, which were ascribed to the reduced agglomerate size of the API after coating (Kim et al., 2021). These results indicate that the influence of increased effective surface area availability after coating for solvent interactions is more dominant than the nature (hydrophilic or hydrophobic) of interactions on the dissolution mechanism. Further advancements in dry coating techniques for producing fine spherical drug particles with improved drug delivery mechanisms and dissolution properties, particularly for poorly water-soluble drugs, have been reported (Saeki et al., 2019). In this case, the drug particles were coated over a corn starch carrier or core particles. They explored two approaches, firstly, incorporating hydrophilic PEG into the coated drug layer to enhance wettability, which showed some improvement but was limited due to unexpected agglomeration of drug particles. Secondly, using swellable core particles that rapidly swell upon contact with water, facilitating the detachment of fine guest drug particles from the surface and leading to excellent dissolution behaviour. Overall, these enhancements demonstrate promising avenues for improving drug delivery of poorly water-soluble compounds.

### 3.4.6 Spreading

Powder spreading applications, especially in additive manufacturing processes like powder bed fusion, are crucial. The spreading behaviour of coated powders directly affects the layer uniformity, build quality, and part accuracy. Spreading tests, including powder flowability measurements and powder bed characterisation, assess the ability of the coated powder to uniformly spread and adhere to the build platform. Optimal spreading ensures precise control over part dimensions and surface finish, resulting in high-quality additive manufacturing (AM) outcomes. Recent applications include nanoparticulate coatings on filter fibres to adjust the wetting and mobility of collected droplets, while ongoing research is exploring the impact of nanoparticles on properties of additively manufactured products, with benefits primarily seen in improved flow, reduced agglomeration, and enhanced packing properties. Blümel et al. (2015) reported that dry particle coating enhances the flowability and bulk density of cohesive PE-HD particles, making it suitable for Laser Beam Melting (LBM). The nanoparticle coating improved powder deposition, with flowability and bulk density serving as reliable indicators of the polymer layer quality.

To ensure uniformly spread powder layers with consistent, high packing densities across the build volume, it is essential to use feedstocks with excellent flow characteristics and optimal bulk density. Dry coating applications for metal AM feed stocks like steel, aluminium, nickel, and copper alloys, have been reported. Overall, these studies demonstrate the enhancement of processability and sintered part properties by improving flowability and laser absorption and reducing defects like cracks through dry coating (Lüddecke et al., 2021; Pannitz et al., 2021). For example, coating steel tool and steel powders with additives like SiC or Y<sub>2</sub>O<sub>3</sub> enhances laser absorption, leading to improved part density and strength (Zhai et al., 2020). Similarly, coating aluminium alloys with nanoparticles like TiC or TiN refines the grain structure, reducing defects and improving mechanical properties (Heiland et al., 2021; Zhuravlev et al., 2021). Similarly, copper alloy powders can be functionalised with materials like carbon black or TiC to enhance laser absorption and processability, enabling the production of dense, high-strength parts (Jadhav et al., 2019). Furthermore, the coating of conductive and radiation-absorbing nanoparticles like carbon black (CB) has been demonstrated as an effective radiation-absorbing additive for polymeric powders for selective laser sintering (SLS), enhancing absorptance and leading to improved melt pool temperatures and sintered part properties (Athreya et al., 2010; Xi et al., 2020).

Thus, it is clear from these studies that generating func-

tionalised particles through dry coating is effective for creating composite particle systems with the required bulk powder properties for additive manufacturing (AM), not only optimising flowability but also adjusting other key material properties such as absorptivity and microstructure.

### 3.4.7 Aesthetics and taste masking

A powder coating technique has also been used to apply taste-masking and moisture-protective films onto tablets, eliminating the need for solvent-based coatings typically employed for these applications. Cerea et al. (2004) reported dry powder coating of micronised acrylic polymers to generate polymeric films, which resulted in taste masking and a modified drug release. The proposed approach is a promising alternative to conventional coating methods, demonstrating potential for taste masking and controlled release applications.

### **3.4.8 Summary**

As can be surmised from the above, the choice of quantitative indicators for dry-coated powders is highly dependent on the material properties, specific industrial application and desired properties of the coating. Drawing from the existing literature, **Table 4** provides a guiding framework for researchers to navigate the process of selecting appropriate coating performance indicators and critical parameters for specific needs and applications highlighted.

In conclusion, dry particle coating is a versatile technique that can significantly enhance performance in various processes. By reducing cohesion, improving flowability, and modifying surface properties, dry particle coating offers a promising approach for achieving better product quality and performance in various applications.

### 3.5 Environmental effects

In dry coating processes, the interaction between guest and host particles varies significantly based on the mechanical properties of the materials. For high Young's modulus materials such as ceramics and metals, guest particles remain adhered to the host surface primarily due to van der Waals forces. However, for softer materials like polymers, guest particles can be partially embedded into the host surface, which is influenced by the host material's Young's modulus and the mechanical stresses during the coating process (Naito et al., 1993; Pfeffer et al., 2001). Temperature and humidity play critical roles in dry coating process efficiency. Higher process temperatures above the glass transition temperature can soften polymers, enhancing the adhesion of guest particles. Additionally, heat generated through mechanical energy during the coating process can further soften materials although this effect is usually minor. For electrostatic dry powder coating, which relies on Coulomb forces for particle adhesion, the moisture content on the powder surface can significantly influence the

Table 4 Guiding frameworks and performance indicators for dry powder coating.

A 11	Performance indicators					
Application	Powder flow	Powder packing	Surface coverage	Powder dispersion	Wettability	
Pharmaceuticals and chemicals	Gravimetric feeding: FFC >3 (Karttunen et al., 2019; Tan and Newton, 1990) DPI: FFC >6 for API-coated carrier particles	Bulk density >0.35 g/cm <sup>3</sup> Hausner ratio <1.22 Carr's Index <20 % (Karttunen et al., 2019; Tan and Newton, 1990)	Flow: 10–20 % (Fulchini et al., 2017) Lubrication: >60 % (Q. Zhou et al., 2011) Aerosolisation: >10 % (Mangal et al., 2019)	Aerosolisation efficiency: FPF >50 % (Bungert et al., 2021)	5–15 %w/w MgSt coating: >90° (Ouabbas et al., 2007)	
Additive manufacturing	LPBF for PE powders: FFC >3.8 (Wencke et al., 2021) SLM for metal powders: FFC >10, Cohesion <0.26 kPa (Hatami et al., 2017)	LPBF for PE powders: bulk density >0.25 g/cm <sup>3</sup> (Wencke et al., 2021) SLM for metal powders: bulk density ~0.42 g/cm <sup>3</sup> (Hatami et al., 2017)			PE powders: hydrophobic coatings performed better by increasing flowability (FFC 5–7) (Blümel et al., 2015)	

process. Düsenberg et al. (2022) found that higher process temperatures reduce the surface moisture on polypropylene, increasing the powder's electric charge and improving the coating efficiency with nanosilica particles. Moreover, humidifying surfaces can increase surface conductivity, making electrostatic coatings more feasible for pharmaceutical excipients such as MCC, lactose, and magnesium stearate, which have high resistivity. Plasticisers are typically used to humidify the surfaces of pharmaceutical tablets or pellets during the electrostatic dry coating process (Vanamu and Sahoo, 2022). Surface modification via dry coating can also enhance the humidity resistance of hygroscopic powders, thereby improving product stability. Karde et al. (2015) reported that coating corn starch with hydrophobic Aerosil significantly improved its flowability and stability under high humidity conditions. The hydrophobic coating reduces capillary bridge formation and interparticle friction, enhancing the performance at various humidity levels.

### 3.6 Coating stability

Throughout their lifespan, dry-coated powders are exposed to various mechanical, chemical, and thermal stresses based on the type of application during handling, conditioning, storage, and transport. Therefore, the physical or structural stability of dry particle coatings is of utmost importance because their performance characteristics, such as flow, wettability, and dispersibility, are heavily dependent on the surface coating integrity and strength. If a coating lacks stability, it can lead to various issues that compromise the functionality of the coated particles. Strong and stable coatings ensure that particles can move past each other smoothly without clumping or sticking to

each other. This is particularly crucial in pharmaceuticals, where consistent flow properties are required for accurate dosing and efficient processing. A stable, well-adhered coating can enhance wettability, making the particles more compatible with solvents or other liquids, which is essential in formulations for improved dissolution and bioavailability. In one such study, Ouabbas et al. (2009) investigated the adverse effects of relative humidity on the storage of dry-coated silica gel particles. The silica gel particles were dry-coated with MgSt to make the surface more hydrophobic. However, the research revealed that after storage, especially at a relative humidity between 30 % and 50 %, the MgSt coating disappeared after 36 days, returning the silica gel to its hydrophilic state due to water vapour absorption and silanol group formation. In contrast, the coating remained stable under drier conditions (below 12 % humidity), and thermogravimetric analysis suggested that MgSt diffuses into silica gel pores under higher humidity (Ouabbas et al., 2009).

Moreover, coating stability under mechanical, chemical, and thermal stress is essential for maintaining the functional properties of particles. Coatings that maintain their integrity under mechanical and chemical stresses prevent particles from degrading or losing their functional properties during handling, transportation, and chemical synthesis and storage when exposed to harsh chemicals. Similarly, thermal stability ensures that coatings can withstand temperature variations without losing their protective properties, which are important for applications such as thermal insulation and catalysis. Overall, the structural stability of dry particle coatings ensures that the coated particles retain their enhanced properties throughout their lifecycle, leading to better performance and reliability in their

applications. These benefits provided from dry-coated powder systems can be diminished or lost, undermining the effectiveness and efficiency of the product.

### 4. Concluding remarks

Dry powder coating has been achieved using various mixers, which, for the most part, provide high shear stresses to break up cohesive guest agglomerates. In some cases, this mixing is supplemented by additional forces, e.g., vibration or magnetic manipulation of the added media. Although the exact theoretical weight fraction required to achieve a given surface area coverage based on particle properties is known, this relies upon the assumption that guest agglomerates are fully dispersed. However, the literature suggests that the dispersion of guest agglomerates is critical to the process and is, in fact, the rate-determining step; hence the need for such intense mixing conditions. These intense mixing conditions may still be insufficient to fully disperse guest agglomerates; however, they can lead to the detachment of guests from the hosts, depending on the particle properties and the mixing intensity employed. An additional consideration is the use of a pre-mixing step, which has the primary aim of breaking up the guest agglomerates, followed by mixing to direct the dispersed guests to the host particles and spread them evenly over the surfaces. At present, there is no guidance on when such a pre-mixing step is necessary. Furthermore, the ideal relative intensities of the pre-mixing and mixing steps have not been widely examined. Referring to the de-agglomeration and coating indices of Zheng et al. (2020), one might hypothesise that an initial high-intensity pre-mixing step would effectively disperse guest agglomerates, which could be followed by lower-intensity mixing to enable them to stick to the hosts. Whether the guests would remain well-dispersed in the mixing step in such a scenario remains to be seen and warrants further research.

Throughout the dry powder coating literature, studies have invariably considered a binary system of a single guest and host material; however, industrial processes are often more complex than this. An obvious example is that of dry powder inhalers, in which highly active sites of the carrier lactose are preferentially coated with fine lactose, leaving only the less active sites to be coated by the API, so that it can be more easily detached from the carrier when inhaled. This inevitably requires a pre-mixing step with fine lactose before the API is added. In other formulated products, the order of mixing is not pre-defined in this way and inevitably influences the structuring of the guests/fines on and around the hosts/larger particles. Karde et al. (2022) demonstrated that in a ternary system, the order of mixing dictates the fractional coverage of two guest materials on the host, with the first additive preferentially coating the host, while the second additive is more influential on the surface energy. This approach opens opportunities for careful formulation control to achieve desirable multicomponent product properties and performance.

The results of dry powder coating processes immediately following the mixing step have been extensively reported. There has been very little examination of the stability of dry powder-coated powders. Detachment of guests from the hosts, migration and clustering of guests along host surfaces, and agglomeration of coated powders are all potential problems that could arise following storage. These problems ultimately affect the flowability and other functional properties of the product. Progress in this direction is somewhat hampered by the impracticality of long-term sample storage to test its effectiveness. This can be accelerated to a degree, e.g., by exposure to high humidity and/or temperature (Calvert et al., 2013), but for other driving forces, such as exposure to vibrations during transportation, the method to accelerate the effect is less clear.

Numerical studies have provided useful insights into dry powder coating processes. Although it is intuitive that an increase in the guest weight fraction should lead to an increase in the surface area coverage, DEM simulations have shown that additional guest particles are less effectively utilised at higher weight fractions (Deng et al., 2018; Khala et al., 2023c). It has also been shown that surface area coverage is notably affected by the initial location of cohesive guests, with placement close to a high-shear mixing zone leading to a more rapid increase in surface area coverage, thus ensuring a more efficient mixing process (Khala et al., 2023c).

Despite the myriad applications of dry powder coating, since the properties of the guest dominate the coating success, a considerable number of dry powder-coated products have successfully used fumed silica (e.g., Aerosil®) as the coating material. One only needs to look at the recent ban on titanium oxide in European markets in the food and dietary supplements industry (Juelicher, 2022), and proposed ban in the pharmaceutical industry (Schoneker, 2024), to see how concerning is this reliance on a single material. Kleinschmidt et al. (2024) explored the use of 'clean label' flow additives using fine, food-safe powders (maltodextrin, lactose and milk powder) to coat food powders, demonstrating a marked improvement in flowability but falling short of the improvement offered by Aerosil®. In fact, the flow improvement was driven by the particle size for the clean-label flow aids, but extrapolation of the trend to the size of Aerosil® powders still suggested a substandard improvement in flow performance. Industries should be more prepared to adapt to alternative guest materials. This is of particular importance when considering the robustness of a supply chain. Furthermore, seemingly identical powders offered by different suppliers do not always perform in the same way (Jiménez Garavito et al., 2022; Sunkara and Capece, 2018), limiting flexibility.

The final pressing requirement of industries is to ensure

the sustainability of their processes. In the context of dry powder coating, beyond the sustainability of the raw materials used and the limitation of waste from failed batches, the greatest strides in this direction may be a reduction in the amount of guest material and energy input used. Regime maps, such as those proposed by Khala et al. (2023a) and Tamadondar et al. (2018), are promising tools for this purpose and are perhaps the most beneficial lines of numerical research on dry powder coating. For wider application, these must be strengthened by encompassing the complexities of disparate systems, such as the type and scale of the mixer, the distribution of particle properties, and the mixing order. In addition, it would be beneficial to establish reliable relationships between the material properties of guest(s) and host, surface area coverage, and the resulting powder bulk properties, e.g., flowability or tabletability. Combined with robust regime maps, such a development would enable a process workflow to be developed, whereby knowledge of the material properties can be used along with the regime map to specify the optimum mixer type and conditions to be used and the minimum guest powder quantity to be added to achieve specified product properties.

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### Nomenclature

```
contact radius (m)
A
           Hamaker constant (-)
Bo
           granular Bond number (-)
d
            guest diameter (m)
D
           host diameter (m)
E
           energy (J)
E^*
           reduced modulus of contact (Pa)
            fractional surface area (-)
            force (N)
           pull-off force (N)
g
            gravitational acceleration (m·s<sup>-2</sup>)
k_{\rm c}
           coating quality index (-)
            de-agglomeration index (-)
           particle mass (kg)
m
P
           capillary pressure (Pa)
r
           asperity radius (m)
           root mean square roughness (m)
r_{
m rms}
           sphere radius (m)
           principal radii of curvature of liquid surface (m)
R_{1}, R_{2}
R^*
           reduced radius (m)
SAC
            surface area coverage (%)
           Stokes' deformation numbers (-)
St_{\rm def}
            weight fraction (-)
W
            weight ratio (-)
х
            separation distance (-)
            surface tension (N·m<sup>-1</sup>)
           interface energy (J)
           reduced intermixing coefficient (-)
Λ
           particle density (kg·m<sup>-3</sup>)
```

```
Subscripts
          van der Waals adhesion
          cohesive
d
          guest
de
          de-agglomerate
D
          host
DMT
          based on DMT theory
          critical point for guest-guest dominated contacts
g-g
          critical point for guest-host dominated contacts
g-h
HM
          harmonic mean
          entity i
          entity j
JKR
          based on JKR theory
kin
          relative kinetic
          mechanical mixing
          normal
          plastic deformation
```

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van der Waals

vdW

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### **Authors' Short Biographies**



**Dr. Francisco Kisuka** is a Research Associate at Newcastle University. His research interests include FEM and DEM modelling, experimental investigations, and data analysis. Recently, he worked on a project aimed at developing a regime map for dry powder coating. In this project, he conducted experiments using a pilot-scale ribbon mixer to explore the influence of the operating conditions on dry powder coating. Additionally, he performed post-analysis of powder samples using various methods, including flowability analysis, SEM imaging, colorimetry, and isoelectric point charge analysis. His expertise in numerical and experimental techniques provides invaluable experience in research involving powders and granular materials.



**Prof. Jerry Heng** is currently a Professor in Particle Technology at the Department of Chemical Engineering, Imperial College London. He holds a PhD in Chemical Engineering from Imperial College London and a BEng in Chemical Engineering from Universiti Teknologi Malaysia. His group's research focuses on surfaces and interfaces of particulate materials. A current key research activity of the group is nucleation and crystal growth, with the aim of developing crystallisation as a separation strategy for the purification of biopharmaceuticals.



**Dr. Colin Hare** is a Senior Lecturer in Chemical Engineering at Newcastle University. His research aims to discover fundamental relationships between material properties, process conditions and product performance in powder-based manufacturing processes. He specialises in powder flow, particle breakage, mixing, powder coating and DEM. He has received funding from the International Fine Particle Research Institute (IFPRI), Corning Inc., GSK, the European Commission, and the EPSRC. He was awarded the Young Researcher Award at the UK Particle Technology Forum in 2012 and the IChemE Nicklin Medal in 2015. He is the vice-chair and former secretary and treasurer of the IChemE Particle Technology Special Interest Group.