

# Adhesion of powders to metal surfaces during compaction

Renewal proposal

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with

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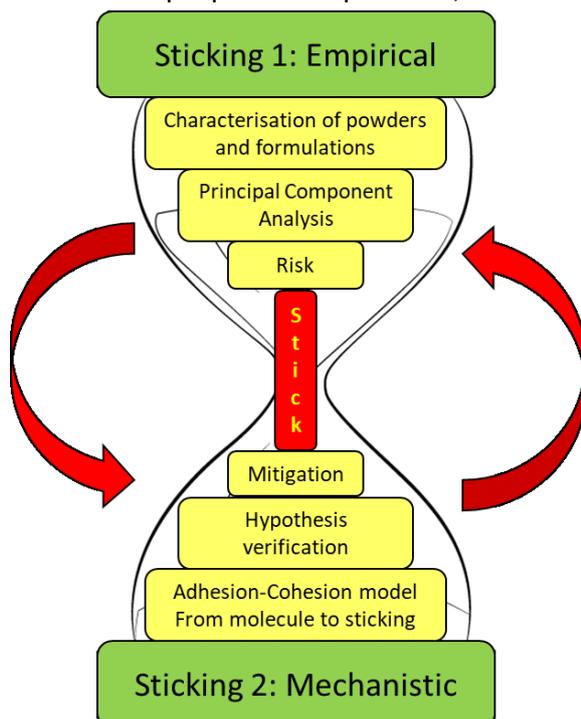
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## Overview of current project

The current project addresses undesired adhesion of powders to metal punch surfaces during compaction of pharmaceuticals, detergents and food products at pressures larger than 20 MPa (ARR-101-02). The key deliverable is a **predictive toolkit** comprising of a database of approximately 20 powder materials (including sticking and non-sticking APIs, sticking and non-sticking excipients, powder formulations and lubricated formulations), characterised in terms of sticking under different levels of compaction stress and loading rate as well as environmental conditions, including temperature and relative humidity.

Given the complexity of the relationships between material properties (chemical and crystallographic information, physical characteristics of the particles and bulk powders, mechanical properties of particles, interaction properties between particles, thermal properties and humidity related properties)

and the process and environment conditions, the first 3 years were focussed on empirical work to create the database, extract functional relationships using Principal Component Analysis and assign risk factors to predict sticking behaviour for new entities.



## Motivation for follow on proposal

In addition to predicting sticking for new entities, the functional relationships derived from the database can also be used to develop and verify the hypotheses behind the sticking phenomenon, prioritised as: temperature, humidity and breakage. These will be explored in the follow-on proposal through targeted studies.

The sticking was quantified using the % area covered by the sticking powder. However, in addition, approximately 20 characterisation methods were also considered and explored as described in the annual report. Extending the database by 20 characterisation methods is out of scope of the original or indeed of the follow-on project. Nevertheless, we are in a position to propose targeted studies to develop the science for the prioritised sticking mechanisms and construct mechanistic models. It is envisaged that mechanistic models combined with the empirical based toolkit will significantly extend our predictive capabilities for sticking.

In the following we propose a series of series of work packages to cover the scientific challenges involved in sticking and refine the proposal by the AGM in June 2023 according to the interests of IFPRI members.

## Work package 1: Extension of existing database

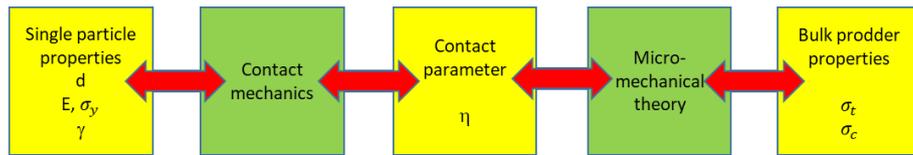
By the time of renewal the database will have contained approximately 20 materials, mostly pharmaceutical APIs, excipients, mixtures and lubricated formulations. The main goal of this WP is to provide further experimental support for validation of the mitigations strategies developed using the predicted tool:

- 1) add **new materials of interest**, including pharmaceuticals, detergents and food materials, particularly formulated and lubricated mixtures which will be prepared using a Turbula.
- 2) Evaluate the **effect of humidity**, which requires experimental work in addition to the current project, targeting detergent and food.
- 3) In addition to single compression events, this stage of the project will focus on **progressive material deposition** over multiple compression events.
- 4) Sticking quantification. % surface area measurement was proven advantageous in its simplicity, however, it does not make distinction between a thin film and larger amounts of particles covering the same area. We are currently cutting the tips of new commercial B tooling using spark erosion to create trilayer disk set-ups using representative tooling material and finishes. At the time of writing we are trialling surface roughness measurement using **high resolution confocal topography** which appears promising to determine the volume of powder material attached to the surface. We also propose to develop a method whereby targeted confocal topography employed for a limited number of powder systems will improve the sticking quantification ability of the entire database.

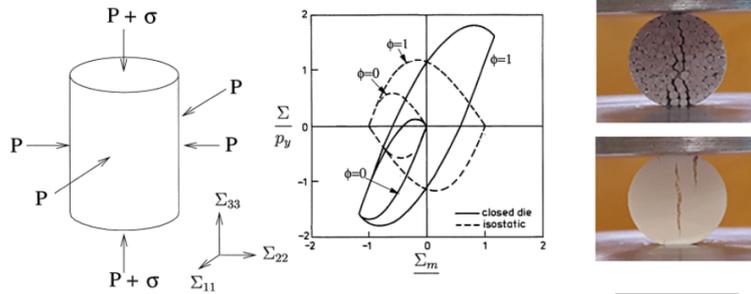
## Work package 2: Mechanistic model from particle properties to sticking

In other research projects we developed a framework to link particle properties to bulk behaviour such as compressibility and compactability. The link between particle and bulk scale was based on contact mechanics and the contact strength parameter  $\eta$  (illustrated in Figure WP2).

We propose to develop a similar framework to establish the strength of the adhesive forces between bulk powder and the metal surface. This framework will require two further inputs described in WPs 3-4.



Fleck, 1995:



Mesarovic and Johnson, 2000:

$$\eta_{MJ}(E, \nu, w, a_0, \sigma_y) = \frac{4\sqrt{\pi-2}}{3\pi\sigma_y} \sqrt{\frac{\pi w E^*}{(2\pi-4)a_0}}$$

Figure WP2: Framework for linking particle to bulk powder compressibility/compactibility

### Work package 3: From chemical information to particle properties. Collaboration with Professor Jerry Heng at Imperial College

This work package complements WP2 by linking chemical information to particle properties. The concept, illustrated in Figure WP3 links molecular structure and unit cell packing to crystal habit which exhibiting heterogeneous surface properties affecting sticking behaviour.

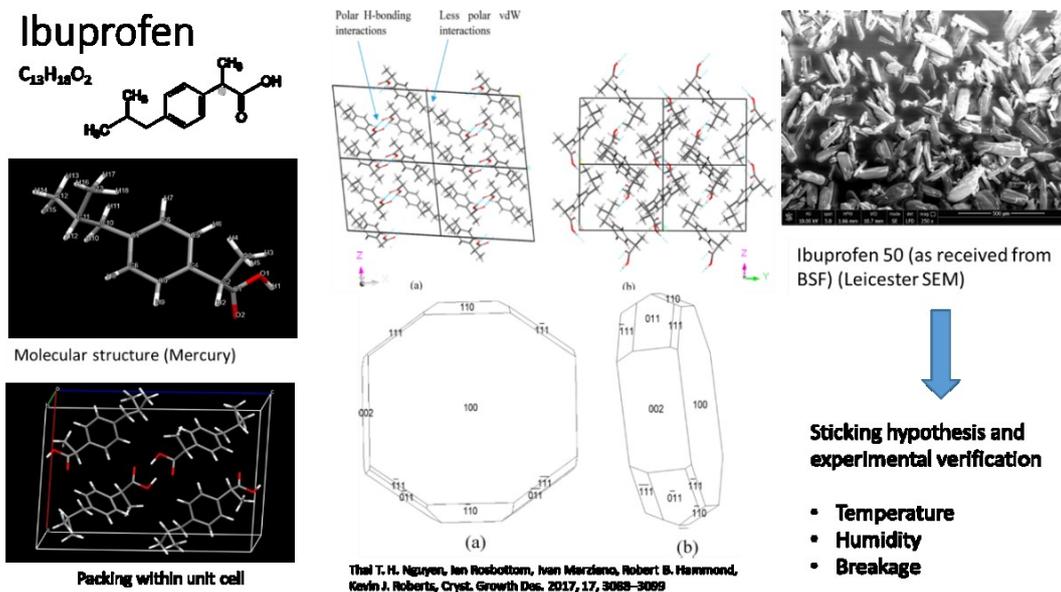


Figure WP3: Linking chemical information to sticking

Powder compaction incurs rearrangement, deformation and breakage of particles which can induce a range of bulk and surface changes in crystalline solids with a range of practical implications including upon sticking. Such physicochemical changes in particle properties include the introduction of defects, amorphisation and polymorphic transformations. To study these processes we propose a collaboration with Professor Jerry Heng at Imperial College.

## Work package 4: Characterisation of adhesion using AFM

We described the use of AFM for surface topography and Kelvin Probe AFM to distinguish between the surface features and attached particles in the annual report. We propose the use of AFM probes where the tips are made of API particles, and also silicon probes functionalised with a range of functional groups in order to characterise the adhesive forces directly. This WP provides input into the mechanistic model proposed in WP2.

## Work package 5: Verification of sticking hypotheses

Three main mechanisms for sticking have been prioritised during the project: temperature, humidity and breakage.

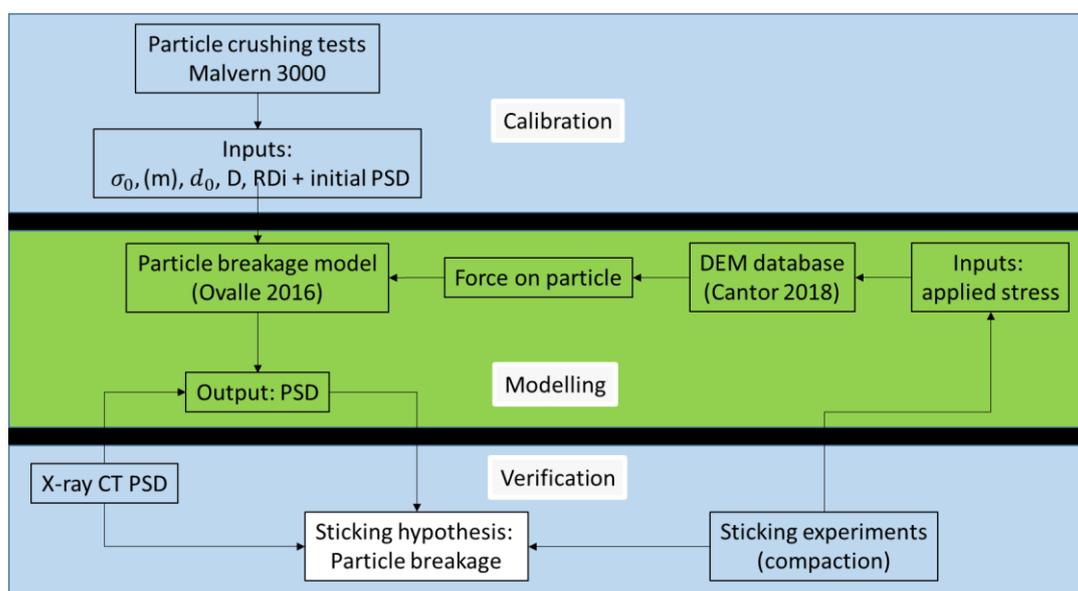
**Temperature.** Sticking can be understood as a coupled thermo-mechanical problem with two sources of heat: 1) Powder compaction involves dissipative processes that generate heat and 2) At the tool interface heat is also generated due to friction. As sticking (gradual deposition of the material to the tool surface) progresses the properties of the materials and surfaces evolve, e.g. phase transformations due to stress, strain rate, temperature.

Coupled thermo-mechanical finite element analysis is proposed to develop an understanding of heat generation and changes of the state of the material at 5 nanometre asperities scale.

**Humidity.** Experimental work planned to study the effect of humidity is included in WP1.

**Breakage.** Brittle materials or granules break during compaction, creating new surfaces. These new unlubricated surfaces give cohesion/strength to compact but also lead to sticking. Examples: mannitol, sorbitol.

A numerical modelling framework was developed at Leicester during other projects, using input such as particle size, PSD and particle strength, a DEM database established by Cantor (2018) to determine the forces applied to the particles during densification and a the population balanced method of Ovalle (2016) to update PSD will be combined with breakage experiments and sticking observations as illustrated in Figure WP5.



WP5: Experimental characterisation of breakage and model development

This work requires two developments:

- 1) determination of particle breakage using a Malvern laser diffraction based analyser with dry feeder using the method of (Rajniak et al. 2018)
- 2) characterisation of particle breakage during die compaction using X-ray CT (fixture design and manufacturing is currently ongoing).

The experimental characterisation methods described in the annual report will be employed as needed to validate the sticking hypotheses. These are electron microscopy, roughness profilometry, AFM profilometry, Kelvin Probe AFM (work function), particle size measurement, energy-dispersive X-ray spectroscopy (EDS, EDX, EDXS or XEDS) elemental analysis, tensile pull-off force measurement. In addition we explored the applicability of several other experimental techniques to provide further understanding on sticking, including AFM with functionally coated probes, PF-QNM - Peak Force Quantitative Nanomechanical Imaging. We sent disks with sticking ibuprofen to our collaborator at Swansea University to explore if XPS or Micro-FTIR could be used to identify the functional groups that are attached to the surface of the punch. The indications were that electrostatic forces or mechanical interlocking of the particles with the surface roughness features are more likely than chemical bonds with the metal. We ruled out experiments that require high vacuum or ultra-high vacuum depending on the sublimation point of the material tested. (The SEM images presented the annual report were taken at Leicester using one of our systems that can be operated with low vacuum).

Furthermore, within the timeframe of the follow-on project the following capabilities are or will become available to us: Electron backscatter diffraction (EBSD) (available at Leicester) to study crystal orientations, Rahman (available at Leicester) to provide information about bonds), Water activity. Future analytical methods that could be considered part of collaborations include: Diffraction Contrast Tomography (£2.7m, Leicester, September 2023), DMTA, FDIGC (part of the collaboration with Jerry Heng at Imperial College). Drop test and shear test are possible to perform at Greenwich to determine the Bond number and to characterise the friction and adhesion of material during ring shear testing.

## **Work package 6: Predictive tool development and mitigation of sticking**

The hourglass diagram on the front page indicated how the empirical work carried out in the first stage of the project trickled down leading sticking hypotheses. The predictive tool developed is based on empirical correlations and principal component analysis. Turning the hourglass around the follow-on project will focus on science and the development of a mechanistic understanding of sticking. By integrating the empirical toolkit with the mechanistic model the predictive capabilities will be mechanism based and thus significantly enhanced. The confidence of using the toolkit beyond the experimental space mapped will also be increased. The integration of the WPs is illustrated in Figure WP6.

Currently sticking is mitigated mostly by lubrication, however, lubrication normally has detrimental effects. The combined toolkit will be used to develop regime maps for sticking and strategies for mitigating sticking through formulation design targeted to address specific effects due to temperature, humidity and breakage.

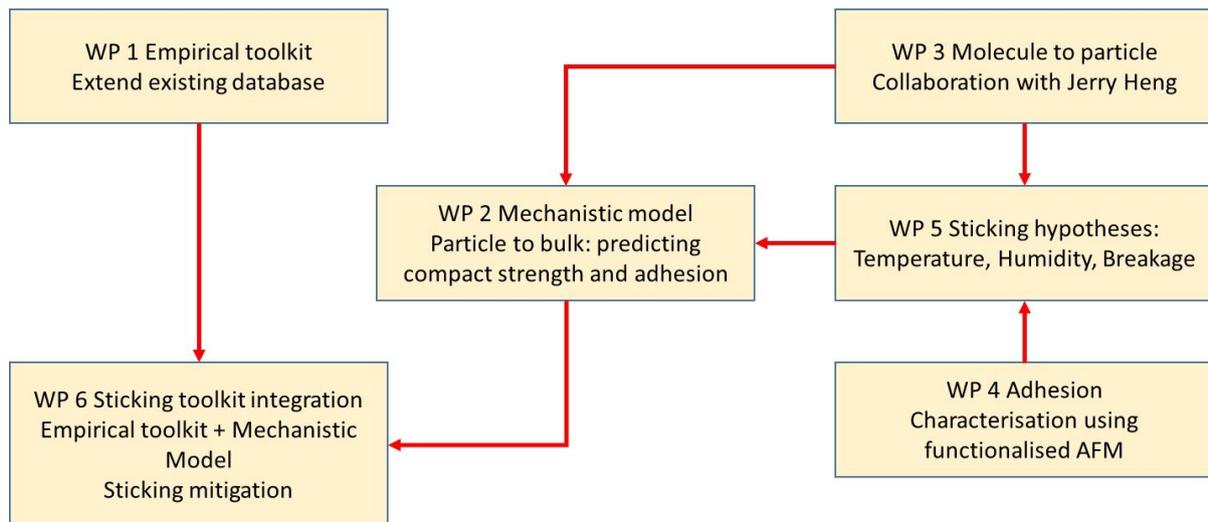


Figure WP6: Integration of WPs

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