



# Fine powder flow under humid environmental conditions from the perspective of surface energy



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## ARTICLE INFO

### Article history:

Received 26 December 2014

Accepted 11 March 2015

Available online 12 March 2015

### Keywords:

Pharmaceutical powders

Humidity

Moisture sorption

Powder flow

Surface energy

Interparticle force

## ABSTRACT

The influence of humidity on surface energetics and flow behavior of fine pharmaceutical powders was investigated. Amorphous and crystalline fine powders with hydrophilic (Corn starch and Avicel PH105) and hydrophobic (ibuprofen) nature were considered for this study. The surface energy was determined using surface energy analyzer and flow behavior was measured in terms of unconfined yield stress (UYS) using a shear tester. The study showed that unlike hydrophobic ibuprofen powder, surface energy and flow of hydrophilic excipient powders were affected by relative humidity (RH). The Lifshitz–van der Waals dispersive ( $\gamma^{LW}$ ) component of surface energy barely changed with varying RH for all pharmaceutical powders. For hydrophilic excipients, the specific component of surface energy ( $\gamma^{SP}$ ) was found to increase with increasing RH. Furthermore, for these excipients, flow deterioration at elevated RH was observed due to increased capillary bridge formation. Detailed analysis showed that  $\gamma^{SP}$  component of surface energy can be an effective indicator for flow behavior of fine powders under varying humid conditions. The present study also brought out the existence of different regimes of probable interparticle forces which dictate the bulk flow behavior of fine hydrophilic powder under humid conditions.

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## 1. Introduction

Increase in relative humidity (RH) increases the particle adhesion forces mostly due to increased capillary bridge formation tendency (Podczek et al., 1996, 1997; Price et al., 2002). This is true, especially in the high humidity region where the amount of adsorbed moisture on the particle surface is more. The atmospheric humidity thus can greatly affect powder materials due to presence of unbound moisture, which is available for chemical or physical interactions (Zografis, 1988; Bravo-Osuna et al., 2007; Faqih et al., 2007; Mollan and Celik, 1995). While the moisture induced chemical changes often results into chemical degradation or incompatibilities with other components of the formulation (Adeyeye et al., 1995; Kuu et al., 1998; Teraoka et al., 2009), changes in the physical properties are usually manifested in terms of their altered flow behavior especially observed with fine excipients or

APIs during handling and processing in pharmaceutical industry (Amidon and Houghton, 1995; Emery et al., 2009). Although the influence of humidity on powder behavior is well established (Amidon and Houghton, 1995; Emery et al., 2009; Stevens et al., 2009), a detailed insight with respect to the surface level changes and interaction between particles under humid condition is still missing from the perspective of surface energetics of fine powder.

Surface energy consists of two components namely “Lifshitz–van der Waals” component ( $\gamma^{LW}$ ) also termed as the dispersive component ( $\gamma^d$ ) and Lewis acid–base component ( $\gamma^{AB}$ ) which is known as the polar ( $\gamma^p$ ) or specific component ( $\gamma^{SP}$ ). In general, higher the Lifshitz–van der Waals (LW) component ( $\gamma^{LW}$ ) of surface energy, higher is the interparticle cohesion or adhesion force (Castellanos, 2005). The specific component ( $\gamma^{SP}$ ) of surface energy basically involves all other interaction forces like Keesom, Debye, hydrogen bonding and other polar interactions. Although surface energy is an important property of powder systems, which governs interparticle interactions and affects powder flow behavior across many industries, including pharmaceutical, there exists very limited number of studies investigating the influence of RH on the surface energies of fine powders. Sunkersett et al. studied the changes in surface energy parameters ( $\gamma^{LW}$  and specific free

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energy,  $\Delta G_{SP}$ , components) of crystalline paracetamol and carbamazepine drugs and found very less variation in  $\gamma^{LW}$  surface energy of the two powders as the humidity was increased. In contrast, the specific free energy component either remained constant or decreased slightly. They concluded that these insignificant variations in these surface energy components were due to differences in the binding sites of water molecules and the interacting probes (Sunkersett et al., 2001). On the other hand, Newell et al. observed a consistent decrease in  $\gamma^{LW}$  component of amorphous lactose with an increase in relative humidity from 0% to 40% and also suggested that this decrease was greatest in the range where water uptake was greatest (Newell et al., 2001). Similarly, Ohta and Buckton also reported a decrease in  $\gamma^{LW}$  of ground amorphous cefditoren pivoxil when the humidity was increased from 0% to 50%. The shielding effect of surface sites by water molecules were held responsible for these changes (Ohta and Buckton, 2004). Also, from water sorption studies in mineral oxides, Sun and Berg found that the  $\gamma^{LW}$  component decreased with an increase in water coverage and vice versa for the free energy of acid–base interaction (Sun and Berg, 2003).

A recent study performed by Das et al. correlated the surface energy changes with dispersibility of salmeterol xinafoate (SX) inhalation formulation before and after storage at 75% RH. They reported an increase in  $\gamma^{SP}$  of all the formulation components when stored at high RH. However,  $\gamma^{LW}$  component of carrier lactose powders decreased and remained unchanged for API. While an increase in  $\gamma^{SP}$  component was attributed to the moisture present on the particle surface, a decrease in  $\gamma^{LW}$  was explained as a result of shielding of surface sites on lactose particles. Non-competitive behavior between the water molecules and non-polar probes for surface sites was held responsible for the unchanged  $\gamma^{LW}$  of SX. Accordingly, they observed decreased dispersibility of the inhalation formulation due to increased adhesion at higher RH (Das et al., 2009). Although previous studies offer some understanding about the influence of humidity on solid surface energetics, lack of clarity and some conflicting observations inspired us to investigate the present subject. Moreover, comparative surface energy studies performed under humid condition and dealing with different solid-state structure (amorphous or crystalline) along with their distinct surface chemical nature are also absent in the available literature. Particularly, the  $\gamma^{SP}$  component of surface energy which indicates the polar interaction capabilities of a surface could be used as an indicator for determining the presence or absence of polar water molecules on the surface. Thus, the crucial information obtained from inverse gas chromatography (surface energy analyzer) can be effectively utilized for analyzing and predicting the powder bulk behavior of fine powder under humid environment.

The present work investigates the influence of RH on Lifshitz–van der Waals ( $\gamma^{LW}$ ) and specific component ( $\gamma^{SP}$ ) of surface energy of different amorphous and crystalline fine powders with either hydrophobic or hydrophilic surface nature. Both,  $\gamma^{LW}$  and  $\gamma^{SP}$  components were evaluated in the range of 30–90% RH from surface energy analyzer (SEA). Furthermore, to explore the moisture induced adhesion behavior of these powders;  $\gamma^{SP}$  component of surface energy was correlated with the unconfined yield stress (UYS) which was determined using the powder rheometer.

## 2. Experimental

### 2.1. Materials

Corn starch and Avicel PH 105 were used as hydrophilic and ibuprofen drug was used as hydrophobic fine powder for the present study. Details of all three materials are given in Table 1.

Decane (Spectrochem, India), Nonane (Merck, USA), Octane (Spectrochem, India) and Heptane (RANKEM, India) were used as non-polar alkane probes; and dichloromethane (Finar, India) and ethyl acetate (Finar, India) were used as polar probes for surface energy studies using SEA.

### 2.2. Surface morphology studies

The surface morphology of all powders was examined using field emission scanning electron microscope (FESEM) (JEOL JSM 7600F, USA). Working distance (WD) of 6.0 mm and a voltage of 1.5–2.0 kV was used during FESEM analysis. To study the aggregate formation tendencies, all powders (unconditioned and 90% RH conditioned sample) were investigated under optical microscopy using an inverted light microscope (Nikon TS 100F, Japan) at 40× magnification.

### 2.3. Moisture sorption studies

Powder samples were placed and uniformly spread to a thin layer in a stainless steel tray. The samples were kept in a humidity chamber (HMG India, India) to equilibrate at preset relative humidity and temperature conditions. All the samples were exposed to five different humidity conditions of 30%, 45%, 60%, 75% and 90% RH in the increasing order for adsorption studies, with the temperature kept constant at  $25 \pm 2$  °C. Small amounts of samples were withdrawn from the chamber after 24 h conditioning and their moisture contents corresponding to percent loss on drying (LOD) were determined by keeping the samples in a hot air oven (Equitron 7053-250, India) to a temperature specified as per the United States pharmacopoeial method.

### 2.4. X-ray diffraction analysis

Crystalline and amorphous phase analysis of as received and humid powders were carried out using X-ray diffractometer (D8 Discover, Bruker, Germany) with a Cu K $\alpha$  X-ray source, operating at 40 kV and 30 mA (increment 0.02 and scan speed 0.2 s/step).

### 2.5. Surface energy studies

Inverse gas chromatography is an important tool for studying interactions of the probe molecules (polar and non-polar) with solid surface sites under controlled environmental conditions of temperature and relative humidity. These interactions can be analyzed at infinite dilution of probe in which the concentration of the interacting probe molecules is so small that the probe–solid interaction is predominant rather than the probe–probe interactions as well as these interactions are non-competitive in nature.

**Table 1**  
Properties of fine powders.

| S. no. | Materials    | Source          | Particle size, $d_{50}$ ( $\mu$ m) | Solid state structure | Surface nature |
|--------|--------------|-----------------|------------------------------------|-----------------------|----------------|
| 1      | Corn starch  | Suru Pharma     | 17.5                               | Amorphous             | Hydrophilic    |
| 2      | Avicel PH105 | FMC Biopolymers | 18.0                               | Crystalline           | Hydrophilic    |
| 3      | Ibuprofen    | BASF            | 33.6                               | Crystalline           | Hydrophobic    |

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