Stokes shape factor for lactose crystals
Konrad Shaffer¹, Clive E. Davies², A.H.J Paterson³, Gerald Hebbink⁴

¹ School of Engineering and Advanced Technology, Massey University, Palmerston North, New Zealand
² (k.shaffer@massey.ac.nz)
³ DMV-Fonterra Excipients GmbH and CoKG

ABSTRACT
The alpha-lactose crystal has a tomahawk shape which needs to be accounted for when designing settling equipment. A shape factor can be used to achieve this. A variety of shape factors have been used for lactose crystals in the literature. This paper sets out to experimentally determine a shape factor for lactose. Large undamaged tomahawk shaped lactose crystals were grown in a lactose agar gel and then recovered for use in settling experiments. Typical industry produced crystals were also tested for comparison. The settling experiments enabled the calculation of a Stokes settling diameter, the diameter of a sphere with the same density and settling velocity as the tomahawk shaped lactose crystal. Using crystal mass to calculate equivalent particle volume and Stokes diameter, the Stokes shape factor for lactose gel-grown crystals was calculated to be 0.99. A Stokes height factor (Bst) was formulated which, when multiplied by the height of the lactose crystal, gives the Stokes settling diameter. The lactose Bst value was determined to be a 0.595±0.007 and 0.643±0.008 for gel-grown and plant-grown lactose crystals respectively.

Keywords: lactose; shape factor; settling velocity; Stokes diameter

INTRODUCTION
The design of gravity settlers and classifiers used in the production of particles of a specific size range requires knowledge of their settling parameters. These can be readily predicted for spherical particles using established relationships for drag coefficients widely available in the literature and standard texts. Lactose crystals, however, are not spherical and while there are also several approaches to estimating the settling velocity of a non-spherical particle, these all require information on particle shape. Generally, lactose crystals conform to a well defined geometry, exhibiting a tomahawk shape [1] as depicted in Fig. 1; the two principal dimensions, as can be measured when the crystal is lying on a flat surface in its most stable position, are its height, B, and its width, A. But, despite their importance industrially, there is little definitive information on shape that can be used with confidence to predict their terminal velocity.

Factors for characterizing particle shape are used in widely differing contexts, and are defined in many different ways; for example Bouwman et al [2] who were interested in quantifying the shape and roughness of granulated pharmaceutical powders considered eight different shape factors. In this work, we are specifically interested in particle descriptors that enable the estimation of terminal settling velocity, \( \upsilon_t \), at very low particle Reynolds numbers, \( Re_p \), and have confined our attention to the Stokes shape factor, \( k_{st} \).

Figure 1. Diagram of tomahawk shaped alpha-lactose crystal [1].
For $Re_p \ll 1$, Stokes law applies, and the terminal velocity of a spherical particle is explicitly related to particle density, $\rho_p$, fluid density, $\rho_f$, fluid viscosity, $\mu$, particle diameter, $d$, and acceleration due to gravity, $g$, by the expression given in Eq (1); see for example, Rhodes [3]:

$$u_t = \frac{d^2 (\rho_p - \rho_f) g}{18 \mu}$$  \hspace{1cm} (1)

The utility of using $k_{st}$, defined in Eq. (2), in calculations with non-spherical particles was demonstrated by Pettyjohn and Christiansen [4]:

$$k_{st} = \frac{u_t}{u_{t,v}}$$  \hspace{1cm} (2)

where $u_t$ is the actual terminal settling velocity of a particle; $u_{t,v}$ is the terminal settling velocity of a spherical particle having the same density and volume as the particle volume, $V_p$; the equivalent volume diameter of the particle, $d_{eq}$, used in the calculation of $u_{t,v}$, is thus $\left(\frac{6V_p}{\pi}\right)^{1/3}$.

Using Eq. (1), writing $u_t$ in terms of a Stokes diameter, $d_{St}$, defined as the diameter of a spherical particle of the same density and the same terminal velocity, writing $u_{t,v}$ in terms of $d_{eq}$ and then substituting into Eq. (2), it follows that:

$$k_{st} = \frac{d_{St}^2}{\left(\frac{6V_p}{\pi}\right)^{1/3}}$$  \hspace{1cm} (3)

Thus if $V_p$ can be measured or estimated, and an appropriate value of $k_{st}$ is known, the terminal settling velocity of a non-spherical lactose crystal can be calculated. The work described here was undertaken to directly measure $k_{st}$ for lactose crystals and to investigate the relationship between sedimentation characteristics and crystal geometry.

MATERIALS & METHODS

Two populations of lactose crystals were used. One from a process line before milling had occurred. Secondly, for comparison, large undamaged tomahawk shaped alpha lactose crystals were grown in a supersaturated lactose agar gel and subsequently recovered.

The principal dimensions, $A$ and $B$, of individual crystals were measured under a microscope; see Fig. 1. The crystal weight for the first 9 gel-grown crystals was also measured. Settling experiments were carried out by dropping single crystals into a transparent sedimentation vessel so that they fell along its longitudinal axis; they were allowed to fall through 50 mm to reach terminal velocity, then the time taken to travel a known distance was measured using a stopwatch; the process was repeated five times. For the first 9 gel-grown crystals, the sedimentation vessel was a 500 mL measuring cylinder and the distance traversed was 100 mm; the crystals were collected for recycling using a stainless steel cup positioned below the traverse zone. For all the other crystals, both the remaining gel-grown crystals and the plant-grown crystals, the sedimentation vessel was a 1 L beaker and the traverse distance was 60 mm; instead of collecting the crystals in a cup, they were allowed to fall to the bottom of the beaker and were removed and recycled using a pipette.

The sedimentation vessel contained fluid consisting of glycerol and supersaturated lactose solution (30 grams of lactose per 100 grams of water). Glycerol was used to control the viscosity of the lactose solution so that $Re_p \ll 1$ and Stokes law was applicable.

RESULTS

Lactose gel-grown crystals traversing the sedimentation vessel were visually measured with the aid of a video camera for the first 9 gel-grown crystals. The video camera enabled the general orientation of the falling lactose crystals to be compared. The lactose crystals settled with the large tomahawk base positioned downwards and the Apex tip positioned upwards. This also occurred for crystals that were originally dropped into the fluid with a different orientation, the crystal changed to the specified orientation while settling.

Elongation ratio, $E$, was calculated for each crystal; $E$ is defined as the ratio of crystal height to crystal width, i.e. with reference to Fig 1, the ratio $[B/A]$. For the gel-grown crystals, for which $0.34\text{mm} \leq B \leq 1.70\text{mm}$, the mean value for the 35 crystals measured was $1.50\pm0.03$. The mean value for the 21 plant-grown crystals, $0.30\text{mm} \leq B \leq 1.69\text{mm}$, was $1.40\pm0.03$. Fig. 2 shows $E$ plotted as a function of crystal height, $B$, for both crystal populations; while there is some scatter, there is no apparent trend with crystal height.
Figure 2. Elongation ratio values plotted against height for gel and plant grown tomahawk alpha lactose crystals.

Plots of Stokes diameter, $d_{st}$, versus crystal height, $B$, for both the gel-grown and the plant-grown crystals suggest a strong linear correlation between these parameters as can be seen in Fig. 3; $R^2 > 0.96$ for both populations. The ratio $[d_{st}/B]$ has been termed the Stokes height factor, $B_{St}$, and can be obtained for each population from the slope of the trend line fitted to the experimental data; the fitted lines have been forced through the origin to be consistent with the definition of $B_{St}$, though for very small particles, Stokes law is not expected to apply[5]. The Stokes height factor, $B_{St}$, for which $0.30 \text{mm} \leq B \leq 1.70 \text{mm}$, was calculated to be $0.595 \pm 0.007$ and $0.643 \pm 0.008$ for gel-grown and plant-grown lactose crystals respectively.

Figure 3. Stokes settling diameter versus measured height for gel and plant grown tomahawk alpha lactose crystals.
Figure 4 is a plot of Stokes equivalent spherical volume, $V_{St}$, defined in Eq. (4), against particle volume, $V_p$, for the 9 gel-grown crystals which were weighed; $V_p$ was calculated using a value of 1540 kg.m$^{-3}$ [6].

$$V_{St} = \frac{\pi d_{St}^3}{6}$$  \hspace{1cm} (4)

It is evident that there is a strong linear relationship between $V_{St}$ and $V_p$, with the data in Fig. 4 well correlated by Eq. (5)

$$V_{St} = 0.98V_p$$  \hspace{1cm} (5)

The Stokes diameter is thus virtually the same as the equivalent volume diameter. Using this relationship in Eq. (4), the Stokes shape factor, Eq. (3), is 0.99.

**DISCUSSION**

**Particle Size Conversion**

Lactose crystal height was used as the description of particle size. Converting from other commonly accepted industrial definitions of particle size involves a simple conversion factor depending on the definition to be used. If using a particle size based on a sieve analysis, which in principle obtains the particle width, the sieve particle size will be multiplied by the elongation ratio. The elongation ratio is the ratio of the height, $B$, to the width, $A$. The elongation ratio was found to be 1.50±0.03 and 1.40±0.03 for gel-grown and plant-grown lactose crystals respectively, which falls within the range of 1.4-1.7 previously found for commercial grade lactose [7-9]. The converted lactose crystal height can then be multiplied by the Stokes height factor to obtain the Stokes settling diameter.

**Stokes Spherical Equivalent Volume**

The crystal weight was measured for the first 9 gel-grown crystal measurements. Fig 4 shows the relationship between the Stokes spherical equivalent volume and the particle equivalent volume. The particle volume was calculated from the measured mass of the corresponding lactose crystals and crystal density. Volume calculated from mass (and crystal density) can be considered as the true particle volume, as this volume excludes any pores or voids. A strong linear relationship exists between the Stokes spherical equivalent volume and particle equivalent volume, $V_{St} = 0.98V_p$. The approximately 1:1 ratio between the Stokes spherical equivalent volume and the particle equivalent volume was an unexpected result due to the irregularly shaped tomahawk lactose crystal. The Stokes diameter is the spherical diameter (of same density) which gives the same settling velocity for the crystal; however these results show that the Stokes settling diameter for lactose crystals is the same as the particle spherical volume equivalent diameter. Typical particle
sizing instruments calculate particle diameters using light scattering techniques. These light scattering techniques commonly calculate the particle spherical volume equivalent diameter. If it is desired to correlate diameters obtained from light scattering instrument to Stokes settling diameter, then these results imply no conversion is required. This is due to the 1:1 relationship with Stokes spherical equivalent settling diameter.

**Stokes Height Factor**

The plant-grown lactose crystals were obtained from a crystallization process line before milling. The reason the plant-grown lactose crystal $B_{St}$ is different has been attributed to being damaged during the crystallization and recovery stages. Under normal crystallization conditions highly concentrated mixed slurries are used, leading to particle-particle and particle-wall interaction. Similar interactions will also be occurring during the drying stages. The damage causes the plant-grown lactose crystals to be more spherical compared to the gel-grown crystals, and have a $B_{St}$ relationship closer to 1. This is because the $B_{St}$ relationship is based on particle height and Stokes spherical diameter, as crystal is damaged the height becomes closer to the Stokes spherical diameter. This damage can be studied by comparing the gel-grown and plant-grown crystal microscope images, see Fig. 5; the highlighted edges show the irregularity in the plant-grown crystal (c) compared to the smooth gel-grown crystals (a and b). To confirm if the $B_{St}$ values for gel-grown and plant-grown crystals were significantly different a statistical analysis was carried out. A T-test resulted in a P-value that was vanishingly small, much less than 0.05 and it is concluded that the $B_{St}$ values for gel-grown and plant-grown lactose crystals are significantly different.

![Figure 5. Tomahawk alpha lactose crystals viewed under the microscope. (a) and (b) are gel-grown, and (c) is plant-grown.](image)

The $B_{St}$ value can be used in settling equations which require the settling diameter of crystals. Assuming the geometrical proportions of lactose crystals are the same at different crystal sizes, the Stokes height factor can also be used for lactose crystals outside of the particle size range tested.

**Errors and Variation**

The simplicity of measuring terminal crystal settling velocity and using the corresponding Stokes law equations is an effective method for formulating a Stokes height factor for the irregularly (tomahawk) shaped lactose crystal. The scatter seen in the results of Figures 2-4 can be attributed in part to experimental errors. Although the settling experiments are relatively simple to carry out, many factors need to be controlled and kept constant to ensure reliability. Temperature changes can affect the fluid properties of solubility, density and viscosity. This affects the settling time of the falling crystal. The sedimentation vessel was equilibrated with ambient temperature. The measuring cylinder fluid temperature would have been better controlled if placed in a temperature controlled room. Another deviation in results could have arisen from foreign matter attached to the lactose crystals. This may have occurred when the lactose crystals were grown in the lactose agar gel (to ensure large undamaged tomahawk shaped crystals) and some remaining gel may have been present even after washing and drying. This was visually noticed for some of the crystals grown, and therefore these crystals were not used in the settling experiments. Foreign matter may have also been introduced when measuring the dimensions of the lactose crystals. Careful handling of the small crystals was also needed to avoid crystal surface damage (altering morphology) and lost crystal mass. Glycerol was used...
to alter the lactose solution velocity so the crystal was settling in the Stokes region. If the homogeneity of the glycerol and lactose solution was not 100%, this would have lead to periods of increased/decreased settling rates for the crystals, leading to variation.

Data variation could also arise from differences in the crystal faces due to growth rate dispersion. Growth rate dispersion happens when different growth rates occur, even when crystals are subjected to identical growth conditions. This gives rise to variation in morphology of the lactose crystal. The Stokes height factor multiplied by the lactose crystals height gives the Stokes settling diameter. This assumes that the ratio of height to other dimensions of crystal remains the same. For instance the elongation ratio was 1.50±0.03 and 1.40±0.03 for gel-grown and plant-grown lactose crystals respectively. There was some difficulty in accurately measuring the width of the lactose crystal, due to crystal not lying flat on surface. These results still indicate that even for crystals growing under identical conditions, the different faces of lactose crystals grow faster or slower leading to slight variation of geometrical proportions. This will ultimately cause variation in the results, regardless of how precisely the experiment was carried out.

CONCLUSION

The Stokes shape factor for gel-grown lactose crystals was found to be 0.99. The Stokes height factor multiplied by the height of the lactose crystal gives the equivalent Stokes spherical settling diameter. The Stokes height factor was measured to be 0.595±0.007 and 0.643±0.008 for gel-grown and plant-grown tomahawk alpha lactose crystals respectively. To convert from a standard measure of particle size such as sieve diameter or equivalent volume diameter to Stokes settling diameter requires a simple conversion step. The ability to convert from the lactose dimension of height to Stokes settling diameter aids in the design of classifiers or settlers to settle out a desired particle size.

ACKNOWLEDGEMENT

We would like to thank DMV-Fonterra-Excipients GmbH and CoKG for funding this work.

REFERENCES