EXPERIMENTAL STUDY OF SPRAY DRYING AND ATOMISATION WITH A TWO-FLUID NOZZLE TO PRODUCE FINE PARTICLES

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Abstract: A series of 36 experiments was performed to investigate particle size and yield from a Buchi laboratory spray dryer with a two-fluid nozzle. First principles theory suggests that the main parameters affecting final particle size should be atomization gas velocity and solution concentration, and the experiments confirmed this. Hybrid ratios such as the ALM (atomization gas to liquid flow ratio) did not correlate well to measured final particle size. Particles of 2-3 microns were obtained at high atomization gas flows, indicating initial droplet sizes of 4-7 microns, while lower flowrates gave larger particles.

Keywords: particle size distribution, yield, atomiser, respirable, inhaled particles, droplet size

INTRODUCTION

Spray dryers have conventionally given particles in the size range from about 20 to 500 microns. In the last 20 years, manufacturers have produced dryers capable of producing particles down to 1 micron in size, suitable for inhaled pharmaceutical products, using two-fluid or ultrasonic atomisation. These dryers can also be very compact, as the tiny droplets dry out before they hit the dryer wall, allowing operation at very small scale (1g product or below). Only limited information is available on how to control the dryer to achieve given product properties, particularly particle size distribution. This study looked systematically at the final particle size produced from an aqueous solution by a Buchi B-290 spray dryer with a two-fluid nozzle under a wide range of operating conditions.

THEORY

In spray drying, liquid is atomized into fine droplets and these then dry out in a hot gas stream. The droplets shrink as liquid is evaporated, and a crust may form, particularly with solution feeds.

The atomization step is clearly key. Most information is available for rotary atomizers and pressure nozzles, which are unsuitable for fine particles. Some correlations have been suggested for two-fluid nozzles, e.g. Thybo et al (2008):

\[
d_{\text{av}} = \left(1 + \frac{W_L}{W_{\text{mol}}^m}\right) k_2 \left(\frac{\mu_L \sqrt{\sigma}}{\rho_L \rho U^2 \sigma D^{1.5}}\right)\]

The ratio of the mass flowrates of the atomizing gas and liquid feed, \(W_{\text{mol}}/W_L\) (sometimes entitled ALM) has been suggested as a characterisation parameter.

However, from first principles, two-fluid atomization is caused by the high gas velocity at the nozzle (typically 100-500 m/s), unlike pressure nozzles and rotary atomizers which use high speed liquid flow. Hence one would expect droplet size to depend more on gas flowrate than on liquid flowrate.

The initial droplet size and the final particle size are related by a mass balance on the solids fraction, assuming no coalescence, agglomeration or attrition:

\[
m_s = m_p S_s = \frac{\pi}{6} d^3 p_s S_p = m_p S_p = \frac{\pi}{6} d^3 p_s S_p\]

Hence the final particle size can be estimated from the initial droplet size as:

\[
d_p = d \left(\frac{p_s S_s}{p_s S_p}\right)^{\frac{1}{3}}\]

Thus, for a given droplet size, low solids concentration and high particle density are expected to give smaller final particles.

EXPERIMENTAL PROCEDURE

The experiments were performed using a Buchi B-290 mini spray dryer with standard Buchi two-fluid nozzles and a high efficiency cyclone to collect the dried particles. Recycled nitrogen was the carrier gas (oxygen <6%) with solvent condensed in the B-295 inert loop. Atomisation gas flow was measured by a digital mass flow meter.

The material dried was egg albumin in aqueous solution, mixed with 32% excipients (mainly sugars) to produce a strong matrix on drying. The product was collected in the glass jar at the bottom of the cyclone, which was then weighed to give the yield.
Particle size distribution (PSD) was measured by a Malvern laser method in dispersion in an antisolvent (isooctane). A small number of runs were also viewed by optical microscopy or SEM (scanning electron microscope) as confirmation. The high efficiency cyclone appeared to be able to recover most particles down to 1 micron in size.

**EXPERIMENTAL PLAN**

A total of 36 experiments were performed and a total of 12 parameters were investigated:

- Nozzle cap diameter (gas passage)
- Nozzle tip diameter (liquid passage)
- Nozzle position (tip protrusion beyond cap)
- Solids composition (formulation)
- Solution concentration
- Atomisation pressure
- Atomisation flowrate
- Main gas flowrate
- Inlet gas temperature
- Liquid flowrate
- Liquid temperature
- Solvent identity (changing viscosity, surface tension, latent heat and heat balance)

Exhaust temperature also varied but is not directly controllable. It was limited to 40-50°C due to temperature sensitivity of some constituents.

A set of base case conditions was chosen and 3 replicate runs were performed. Most runs used a univariate variation from this, to clarify which variables had the most effect. The range of variation is shown in Table 1. Typical yield was 50-80%.

### Table 1. Experimental conditions used

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Base case</th>
<th>Variants</th>
</tr>
</thead>
<tbody>
<tr>
<td>Excipients level</td>
<td>30%</td>
<td>0%, 55%</td>
</tr>
<tr>
<td>Solids concentration</td>
<td>5%</td>
<td>10%, 2.5%, 1%</td>
</tr>
<tr>
<td>Inlet temperature</td>
<td>80°C</td>
<td>60, 100</td>
</tr>
<tr>
<td>Outlet temperature</td>
<td>40°C</td>
<td>45, 50</td>
</tr>
<tr>
<td>Aspirator setting</td>
<td>90%</td>
<td>60, 65, 100</td>
</tr>
<tr>
<td>Main airflow</td>
<td>600 l/min</td>
<td>400, 440, 670</td>
</tr>
<tr>
<td>Solution feedrate</td>
<td>3 ml/min</td>
<td>6, 8, 10</td>
</tr>
<tr>
<td>Atomisation pressure</td>
<td>4 barg</td>
<td>1, 2, 5.5, 6</td>
</tr>
<tr>
<td>Atomisation flowrate</td>
<td>0.2 g/s</td>
<td>0.1, 0.15, 0.3, 0.4, 0.5</td>
</tr>
<tr>
<td>Nozzle protrusion</td>
<td>0 (flush)</td>
<td>1mm (protruding)</td>
</tr>
<tr>
<td>Nozzle tip diameter</td>
<td>0.7mm</td>
<td>2.0</td>
</tr>
<tr>
<td>Nozzle cap diameter</td>
<td>1.5mm</td>
<td>1.4 with 0.7 mm tip 2.8 with 2.0 mm tip</td>
</tr>
<tr>
<td>Insulation</td>
<td>No</td>
<td>Yes</td>
</tr>
<tr>
<td>Solution heated</td>
<td>No</td>
<td>Yes</td>
</tr>
<tr>
<td>Solvent</td>
<td>Water</td>
<td>Acetone, ethanol/ water mixtures (25%, 12.5%, 6.3%), heated water</td>
</tr>
</tbody>
</table>

**RESULTS**

The variation in PSD between the 3 base case runs, shown in Fig.1, indicates the extent of random variability. Calculated mean particle size ($d_{50}$) was 5.0, 5.3 and 6.3 μm for the three runs.

![Particle Size Distribution](image.png)

**Fig. 1. Variability in base case**

**General results**

Most of the variables had little effect on final PSD and yield. Drying pure egg albumin without excipient gave a much lower final particle size and yield, confirming that the structure collapses without the robust matrix formed by the excipient. If the liquid nozzle projected beyond the cap, yield was reduced and PSD was wider, confirming that it should be set flush with the cap. Visual observation shows that having the nozzle projecting gives a wider spray angle and a greater danger of wall impingement.

**Effect of atomization flowrate**

Results indicate that atomisation gas flowrate was the main effect on particle size and runs were performed at 0.1, 0.15, 0.2, 0.3, 0.4 and 0.5 g/s, using both 1.5 and 1.4 mm caps and thus giving a wide range of gas velocities at the nozzle annulus. High flowrate and velocity gives smaller particle size. The effect levels off with increasing flowrate and little further change is observed at and above 0.4 g/s. Final mean particle size was 2-3 μm at high atomisation flowrate, 4-7 μm at 0.2 g/s and 12 μm for the single run at 0.1 g/s.

![Particle Size Distribution](image.png)

**Fig. 2. Effect of atomization gas flowrate**
The results were also plotted against ALM. Runs with the same liquid flow plot on to a smooth curve (Fig. 4), while higher flows lie off the curve and give slightly smaller particles, opposite to the prediction from ALM. Hence, it seems inappropriate to lump together atomization gas and liquid flowrates as a ratio, as their relationships to droplet size follow very different forms. Also, the trend was reasonably close to the Thybo correlation (1), with some scatter, but a simple plot against 1/U gave an equally good trend.

The measured bulk density was roughly 350-500 kg/m³, indicating a particle true density around 500-750 kg/m³. The back-calculated initial droplet size from (3) is 5-7 μm at high atomization flow (0.3-0.5 g/s), 10-15 μm at 0.2 g/s and 25-30 μm at 0.1 g/s.

The correlation for both particle and droplet size is slightly better with gas velocity than with flowrate, particularly for the one result with the large nozzle.

**Effect of solution concentration**

Particle size measured for solids concentrations of 1% and 10% is compared in Table 2 with predictions from Eq. 1, based on an average particle size for the base case runs at 5% concentration. Within the limits of scatter on the data, the final particle size varies exactly as predicted from theory.

**Table 2. Variation with solids concentration**

<table>
<thead>
<tr>
<th>Concentration</th>
<th>Measured d50</th>
<th>Predicted d50</th>
</tr>
</thead>
<tbody>
<tr>
<td>5%</td>
<td>5.5 μm (ave)</td>
<td>-</td>
</tr>
<tr>
<td>1%</td>
<td>3.25 μm</td>
<td>3.22 μm</td>
</tr>
<tr>
<td>10%</td>
<td>7.09 μm</td>
<td>6.93 μm</td>
</tr>
</tbody>
</table>

**CONCLUSIONS**

Atomisation gas flowrate (or velocity at the nozzle) and initial solution concentration had the largest effect on product particle size distribution when a two-fluid nozzle is used. Hybrid dimensionless ratios like ALM are misleading and should not be used to characterize the atomization process.

Further work is planned to study a material soluble in different solvents to show the effect of surface tension, viscosity and latent heat. Main airflow and inlet air temperature had little effect, but might affect crust formation if varied over a wider range.

**NOMENCLATURE**

- d, D: diameter (m)
- k: constant in Thybo equation (-)
- m: mass (kg)
- S: solids mass fraction (kg kg⁻¹)
- U: velocity (m s⁻¹)
- W: mass flowrate (kg s⁻¹)

- μ: viscosity (kg m⁻¹s⁻¹)
- ρ: density (kg m⁻³)
- σ: surface tension (N m⁻¹)

**Subscripts**

- atm: atomizing gas
- d: droplet
- g: gas
- L: liquid
- p: particle
- sm: surface mean

**REFERENCES**