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Lagrange Two-Dimensional Interpolation Method for Modeling Nanoparticle Formation During RESS Process

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Abstract

In the pharmaceutical industry, a great number of products are in the form of particulate solids. Since the mid-1980s, a new method of powder generation has appeared involving crystallisation with supercritical fluids (SCF). The rapid expansion of supercritical solutions (RESS) is a promising new process for the production of small and uniform particles. Several variables can influence the RESS process: the nozzle temperature, preexpansion pressure, the nozzle diameter, and geometry. In this work, a two-dimensional Lagrange interpolation method has been proposed to describe the size of nanoparticle forming through the rapid expansion of supercritical solutions, as a function of preexpansion pressure and nozzle temperature.

Keywords: 2D-Lagrange interpolation, RESS, Modeling, Nanoparticle generation.

1 Introduction

Interest in supercritical fluids and their potential use for process improvements has significantly increased in the past decade. Some of the extraction processes such as decaffeination, and some polymerization and foaming processes have become commercial. Particle formation will most likely be the next major commercial application area that uses supercritical fluids. The particle formation technology that uses supercritical fluids has evolved

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in many different forms during the last 20 years. Several review articles have already appeared in the literature ([1]-[10]). A wide variety of organic and inorganic materials have been processed in the form of particles, for example, to crystallize a supercritical fluid-soluble compound, or as non-solvent to precipitate supercritical fluid-insoluble materials. Micronization and recrystallization of pharmaceutical compounds using supercritical fluids has many advantages over conventional techniques such as spray drying, jet milling, grinding, and liquid antisolvent technique.

2 The RESS process

2.1 A two-step process: Solubilisation and particle formation

RESS is a two-step process: after having solubilised a substance in an SCF, the mixture is suddenly depressurized in a nozzle, causing fast nucleation and fine particle generation(Fig.1).

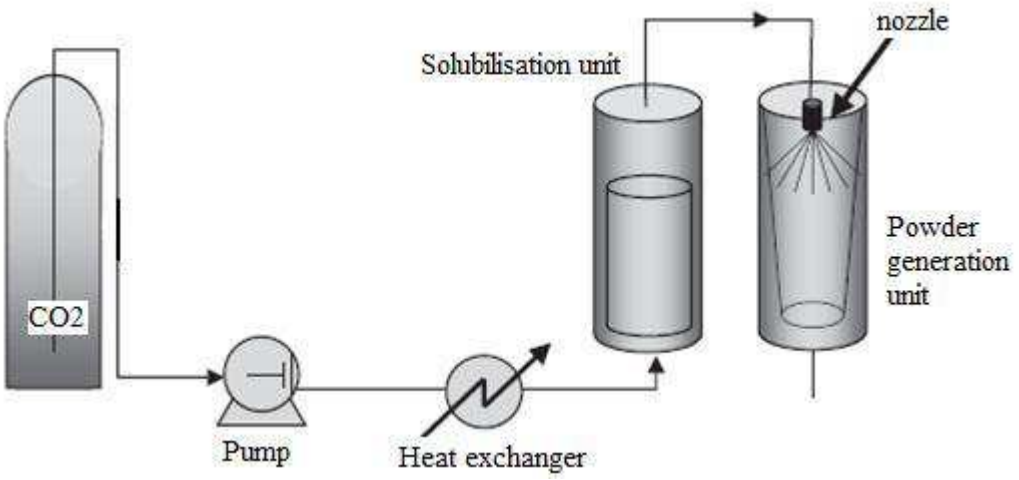

Fig. 1. The key parameters of the extraction step are obviously the operation T and P.

3 Mathematical modeling

A schematic representation of the RESS expansion device used to micronized ibuprofen is displayed in Fig.2. The RESS process can be generally described as follows: a dilute saturated solution at pre-pressure(P0) and pre-temperature(T0) expands across a well designed nozzle into a post-expansion chamber under atmospheric conditions.

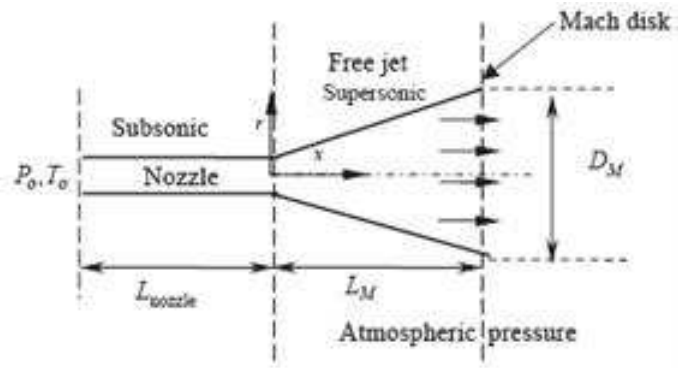

Fig. 2. Schematic presentation of the RESS expansion device.

An understanding of the underlying physical phenomena of the relationship between the mechanisms of particle formation and growth during the RESS process is still at a rather early stage. It sounds that fitting an adequate mathematical equation by experimental results, can predict the general trends of particle size.

3.1 2D-Lagrange interpolation

2D-Lagrange interpolation is based on 1D-Lagrange interpolation. In this method, one of the variables is forced to be constant and, with another variable, the Lagrange polynomials can be written by using the given data. Then, this value can be complicated for the final form of 2D-Lagrange interpolation. The result is a 2D-Lagrange polynomial whose functional agents are replaced by Lagrange polynomials. The interpolation formula of Lagrange may be a more straightforward way for obtaining a function in explicit form. 2D-Lagrange polynomials can be described as follows:

$$L_{ij} = L_i(x)L_j(y) \qquad 0 \le i \le n, \quad 0 \le j \le m$$

$$L_i(x) = \prod_{s=0, s\neq i}^{n} \frac{(x - x_s)}{(x_i - x_s)}, \qquad L_j(y) = \prod_{s=0, s\neq j}^{m} \frac{(y - y_s)}{(y_i - y_s)}$$

So we have

$$L_{ij}(x_r, y_s) = \begin{cases} 1 & i = r, j = s \\ 0 & otherwise \end{cases}$$

And then we have:

$$P(x, y) = \sum_{i=0}^{n} \sum_{j=0}^{m} f((x_i, y_j))L_{ij}(x, y)$$

Where $P(x, y)$ is a polynomial that interpolates $f(x, y)$ in the given data and is called the $2D$- Lagrange interpolant polynomial. Theorem: for $(n+1)(m+1)$ arbitrary support points

$$((x_i, y_j), f(x_i, y_j))$$

there exists a unique polynomial $P(x, y)$. After simplification, the nanoparticle size of ibuprofen as a function of thermodynamic conditions (preexpansion pressure and nozzle temperature) can be written as follows:

$$D = \tfrac{1}{3000}(p-85)(p-90)(p-100)(p-110)\times$$

$$[\tfrac{295}{15000}(T-85)(T-90)(T-95)(T-100) - \tfrac{325}{3750}(T-80)(T-90)(T-95)(T-100)+$$

$$\tfrac{380}{2500}(T-80)(T-85)(T-95)(T-100) - \tfrac{419}{3750}(T-80)(T-85)(T-95)(T-100)$$

$$+\tfrac{460}{15000}(T-80)(T-85)(T-90)(T-95)]$$

$$-\tfrac{1}{9375}(p-80)(p-90)(p-100)(p-110)\times$$

$$[\tfrac{91}{15000}(T-85)(T-90)(T-95)(T-100) - \tfrac{96}{3750}(T-80)(T-90)(T-95)(T-100)+$$

$$\tfrac{315}{2500}(T-80)(T-85)(T-95)(T-100) - \tfrac{498}{3750}(T-80)(T-85)(T-90)(T-100)$$

$$+\tfrac{769}{15000}(T-80)(T-85)(T-90)(T-95)]+$$

$$\tfrac{1}{10000}(p-80)(p-85)(p-100)(p-110)\times$$

$$[\tfrac{89}{15000}(T-85)(T-90)(T-95)(T-100) - \tfrac{93}{3750}(T-80)(T-90)(T-95)(T-100)+$$

$$\tfrac{251}{2500}(T-80)(T-85)(T-95)(T-100) - \tfrac{479}{3750}(T-80)(T-85)(T-90)(T-100)$$

$$+\tfrac{701}{15000}(T-80)(T-85)(T-90)(T-95)]$$

$$-\tfrac{1}{3000}(p-80)(p-85)(p-90)(p-110)\times$$

$$[\tfrac{287}{15000}(T-85)(T-90)(T-95)(T-100) - \tfrac{330}{3750}(T-80)(T-90)(T-95)(T-100)+$$

$$\tfrac{405}{2500}(T-80)(T-85)(T-95)(T-100) - \tfrac{551}{3750}(T-80)(T-85)(T-90)(T-100)$$

$$+\tfrac{812}{15000}(T-80)(T-85)(T-90)(T-95)]$$

$$\tfrac{1}{15000}(p-80)(p-85)(p-90)(p-100)\times$$

$$[\tfrac{2690}{15000}(T-85)(T-90)(T-95)(T-100) - \tfrac{3200}{3750}(T-80)(T-90)(T-95)(T-100)+$$

$$\tfrac{3600}{2500}(T-80)(T-85)(T-95)(T-100) - \tfrac{1050}{3750}(T-80)(T-85)(T-90)(T-100)$$

$$+\tfrac{1150}{15000}(T-80)(T-85)(T-90)(T-95)]$$

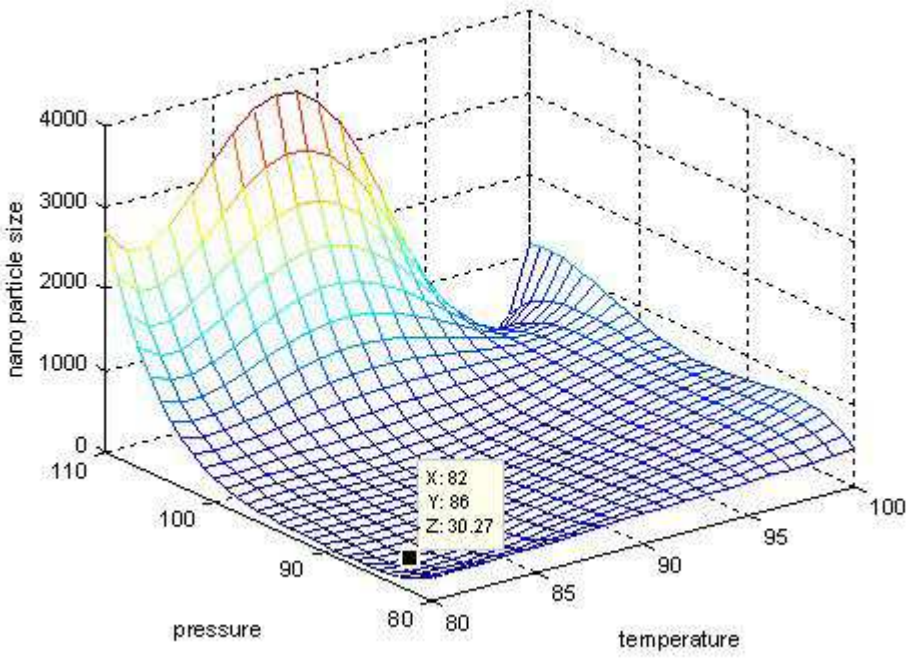

Fig. 3. Calculated size of nanoparticles as a function of nozzle temperature and preexpansion pressure.

4 Optimization

The 2D-Lagrange equation was optimized in order to find the nozzle temperature and preexpansion pressure ,which resulted in the minimum value of mean particle size. The results showed that the minimum particle size (30.27nm) can be gained in the preexpansion pressure of 86 bar and nozzle temperature of 820C. This was proved by running an experiment in the above-mentioned optimal condition. The experimental results showed that the minimum particle size that can be formed in the optimum operational condition is about 40nm (Fig. 4.).

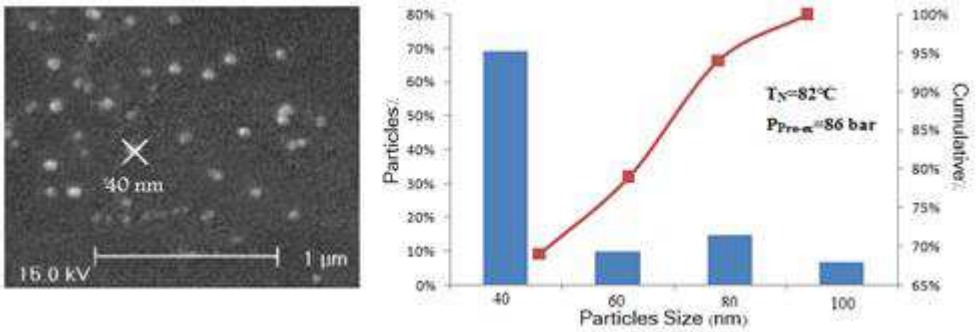

Fig. 4. SEM image and PSA results for the optimal condition.

Fig. 5. shows the results of SEM and particle size analyzing system for another powder that was produced in the preexpansion pressure of 103 bar and nozzle temperature of 860C.

The calculated mean particle size for this sample is 741nm and for the experimental one is 803nm.

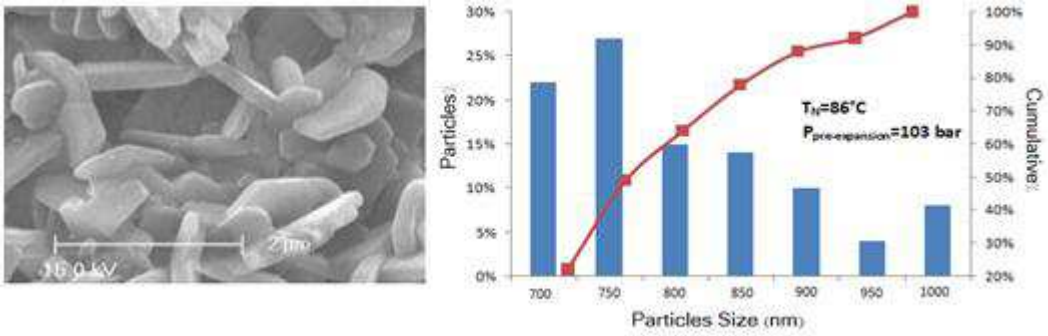

Fig. 5. SEM image and PSA results for the sample processed in P=103 bar and T=86 0C.

5 Conclusion

A mathematical model has been used as a novel method for data prediction. Actually, we matched a 2D-Lagrange interpolation model with our experimental data. The calculated data was checked with the experimental data in the optimal condition and other conditions, and a comparison between them confirms that the interpolated and experimental results are in good agreement with each other.

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