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Study of Supersaturation, Vibration Intensity and Time of Crystallization Variables in the Vibrated Bed Lactose Monohydrate Production Process

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Lactose monohydrate crystallization was carried out in a batch vibrated-bed crystallizer. The crystallization trials were performed to determinate yield and mean crystal size prediction models and effect determination of variables supersaturation level, dimensionless vibration number and process time, using a central composite design. The parameters range adopted were in accordance with a previous study with same component, approximated of industrial processes, and fixed temperature of 50 °C. An optimization was made using response prediction models. It was observed a significant effect of process parameters discussed and the effectiveness of response prediction models, which explained maximum errors below 0.86 % in optimized condition.

1. Introduction

Lactose is industrially produced from whey permeates obtained after the production of cheese and/or whey proteins. The main steps in lactose production are concentration, crystallization and separation. The concentration process involves the evaporation of water in whey permeate to increase lactose concentration, to a saturation/supersaturation condition. Concentrated whey permeate has about 65 - 70 % total solids, with about 80 % of the total solids as lactose. The mixture is cooled to crystallization process temperature (or during the process), in which the lactose is separated as α-lactose monohydrate crystal. Crude/food grade lactose is obtained after two stages of centrifugation, and final drying (Wong et al., 2012).

The solution supersaturation is a relevant variable during batch crystallization processes, which significantly determines the development of nucleation and growth phenomenon (Srisa-nga et al., 2006) and consequently, yield and crystal size (Markande et al., 2012).

Teixeira et al. (2012) have shown that vibrations can be used as a simple and effective controlling tool, to improve the conditions of crystal growth and the quality of the final product.

Seed loading is an important factor controlling the product size distribution. The critical seed concentration can be determined by several batch trials using a small laboratory crystallizer with the help of the seed chart (Kubota et al., 2001). Qamar et al. (2012) observed that supersaturation and mean crystal size of products were reduced on increasing the number of seeds, while productivity, yield and purity were improved.

Process optimization by one-parameter-at-time method (full factorial design) results in a large number of experiments. The use of central composite design guides to a special set of arrays called orthogonal arrays, which stipulate the way of conducting the minimal number of experiments, and gives full information of all the parameters that affect the performance (Bund and Pandit, 2007).

The aim of the present work is to investigate the effect of supersaturation level, dimensionless vibration number and time on crystallization behavior of lactose monohydrate, using vibration as motion mechanism, with high seeding.

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2. Material and methods

2.1 Previous procedures

Reagents used in the experiments are monohydrate lactose (MEGGLE S/A – 99.84 % purity) used as solute component in the preparing of saturated solutions and, with Sauter Mean Diameter of $5.346 \times 10^{-6} \pm 0.206 \times 10^{-6}$ m, as seeds (5.0×10^{-2} kg), absolute ethylic alcohol P.A. (CINÉTICA – 99.5 °GL) used in the washing step of the crystals after the crystallization and separation process, principally due the property of reagent, which the lactose is insoluble. Deionized and distillated water was used to prepare lactose solutions.

Aqueous lactose solutions for crystallization trials were prepared using the adjustment of lactose monohydrate solubility values obtained by Nývlt (1971), Eq(1), at different saturation temperatures. The polynomial adjustment was done using the method *Quasi-Newton*, with a quadratic correlation coefficient (r^2) of 0.9551.

$$C^* = (12.852) + (0.820)T^* + (0.940)T^{*2} + (0.387 \times 10^{-5})T^{*3} \quad (1)$$

where C^* is the saturation concentration (kg lactose monohydrate/100 kg water) and T^* is the equilibrium temperature (°C).

2.2 Experimental setup

The batch vibrated-bed crystallization experiments were performed in a 0.5×10^{-3} m^3 jacketed stainless-steel crystallizer with a trunk-conic shape at an angle of 65° with the horizon. The batch vessel was equipped with vibrated disks agitator. The mixer of 6.0×10^{-2} and 8.0×10^{-2} m diameter perforated disks, fixed on the central oscillating bar, was used at an intercalated height 1.17×10^{-3} m from the bottom of the crystallizer. All experiments were carried out at agitation to ensure all the crystals were maintained in suspension.

The crystallizer temperature was controlled with a thermostatized bath (TECNAL, model TE-184), monitored with a calibrated thermocouple (FULLGAUGE, model TIC17RTG), which was inserted in the crystallizer through a sampling hole. A solution volume of 2.80×10^{-4} m^3 was used in all experiments.

The experimental setup used in this study is presented in Figure 1.

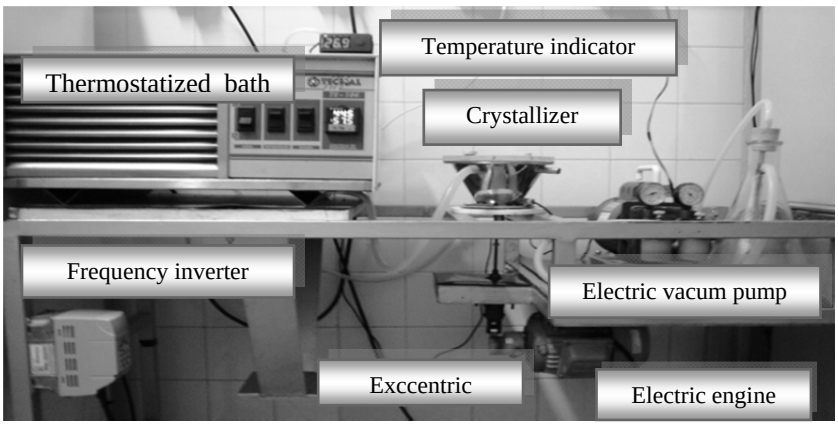

Figure 1: Crystallization unit

Vibration system was composed by an oscillating central axis coupled to an eccentric for transmitting the bed mechanical energy generated by an alternate current engine (WAG, 0.75 CV), controlled by a frequency inverter (WAG, CFW-08).

2.3 Evaluation of yield and produced crystals size

The yield (y) is defined by relation between the produced lactose crystals mass (m_f) and seeded lactose mass (m_s), Eq(2).

$$y = \frac{m_f}{m_s} - 1 \quad (2)$$

Mean crystal size of products (MCSP), which was obtained by Sauter's average diameter, in m, was measured by laser diffraction (MALVERN MASTERSIZER 2000).

2.4 Crystallization trials

Crystallization experiments were carried out adjusting initially the crystallizer at saturation temperature (simultaneously with the solution preparation), in which the solution sample was transferred. The agitator was turned on regulating the vibration frequency by tachometer. The cooling process started, until the solution reached the operation temperature, 50 °C (Shi et al., 2006).

Seeding was performed with commercial monohydrate lactose seeds, 0.5×10^{-1} kg, defined by preliminary tests to reach a dense phase condition, which is commonly assumed in industrial process with fixed temperature operation.

The crystallization products were filtered using vacuum and washed with ethanol. The crystals were dried using an oven (Medicate, MD 1.3), for 24 h at 60 °C, to reach the maximum moisture specification allowed for the commercial lactose.

2.5 Central composite design (CCD)

The experimental design was performed to determine effects of supersaturation level, dimensionless vibration number and time variables to obtain the best operational condition in vibrated bed lactose monohydrate crystallization. The coded variables were: x_1, x_2 and x_3, in terms of original variables: S, Γ and t. Table 1 shows the levels used for the coded independent variables and their respective intervals, where the value of α (orthogonality) is 1.414.

Table 1: Coded levels of the CCD

x_i	–1.414	–1	0	+1	+1.414
S (x_1)	1.14	1.25	1.51	1.77	1.88
Γ (x_2)	0.54	0.62	0.80	0.98	1.05
t (x_3)	0.80	1.00	1.50	2.00	2.20

The experimental levels were chosen by different methods. Supersaturation levels were chosen using metastable experimental limit at 50 °C described by Wong et al. (2012). Figure 2 presents the used range.

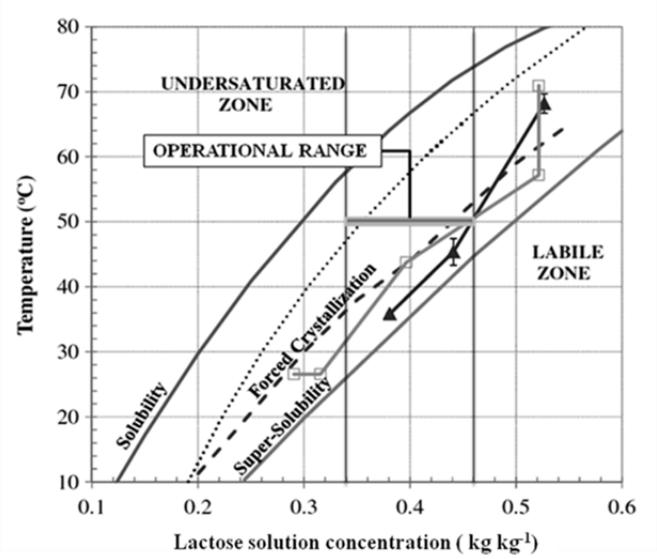

Figure 2: Experimental range of supersaturation (adapted from Wong et al., 2012)

3. Results and discussion

3.1 Experimental design and regression models

The experimental design (CCD) of lactose monohydrate crystallization was performed in eighteen trials. The experiments were conducted with four replications at the central point. Table 2 presents the central composite design with yield (y) and mean crystal size of products (MCSP) responses for each crystallization experiment.

Table 2: Design matrix and experimental responses (A – axial point; C – central point)

Trial	*S*	Γ	*t*	*y* (%)	*MCSP* × 10^6 (m)
1	1.25	0.62	1.00	29.10	4.203
2	1.77	0.62	1.00	34.34	4.604
3	1.25	0.98	1.00	23.10	4.455
4	1.77	0.98	1.00	46.24	4.782
5	1.25	0.62	2.00	95.14	4.277
6	1.77	0.62	2.00	125.98	4.581
7	1.25	0.98	2.00	102.28	4.404
8	1.77	0.98	2.00	119.42	4.132
9 (A)	1.14	0.80	1.50	19.26	4.569
10 (A)	1.88	0.80	1.50	119.42	4.590
11 (A)	1.51	0.54	1.50	79.58	4.574
12 (A)	1.51	1.05	1.50	66.58	4.384
13 (A)	1.51	0.80	0.80	57.56	4.335
14 (A)	1.51	0.80	2.20	77.68	4.476
15 (C)	1.51	0.80	1.50	82.66	4.142
16 (C)	1.51	0.80	1.50	74.62	4.352
17 (C)	1.51	0.80	1.50	73.34	4.411
18 (C)	1.51	0.80	1.50	77.34	4.251

It was notable the MCSP decreasing with yield increase. This inverse trend can be explained by crystallization mechanism and a possible occurrence of nucleation phenomena associated with the operational condition of experiments. The number of crystals and associated total surface area of the crystals have a significant impact on the linear grow rate (Shi et al., 2006).

The experimental design resulted in regression effects, that adjust the experimental isothermal process data for response variables, as a function of supersaturation level, dimensionless vibration number and crystallization time factors. The adjustments for yield and MCSP responses, with coded variables, were represented by Eq(3) and Eq(4), with quadratic correlation coefficients (r^2) of 0.9809 and 0.7009, respectively.

$$\hat{y}_y = 75.78 + 37.64x_1 - 0.99x_2 + 8.73x_3 - 2.01x_1^2 - 0.14x_2^2 \\ -2.88x_3^2 - 0.66x_1x_2 + 2.45x_1x_3 + 0.52x_2x_3 \quad (3)$$

$$\hat{y}_{MCSP} = 4.324 - 0.052x_1 - 0{,}013x_2 - 0.079x_3 + 0.093x_1^2 + 0.042x_2^2 \\ +0.005x_3^2 - 0.094x_1x_2 - 0.087x_1x_3 - 0.081x_2x_3 \quad (4)$$

Predicted values constructed by design in function of observed values, for respective responses, have shown a homogeneous distribution and a system without trends.

3.2 Significant effects in lactose crystallization process

For adopted 15% significance level (p-value), relative with mean crystal size of products response, were significant the linear variable x_3, quadratic x_1 and associated x_1. x_3, x_2. x_3, with a quadratic correlation coefficient (r^2) of 0.6104. For yield, were significant the linear variables x_1 and x_3, reaching a quadratic correlation coefficient (r^2) of 0.9720.

These results indicates that supersaturation level, crystallization time and dimensionless vibration were significant in the lactose monohydrate crystallization process, for yield and MCSP response variables. Associated factors had great interaction among these, not be appropriate analyze them individually, for the experimental range studied.

3.3 Operational optimization of batch lactose crystallization

Many factors including concentration, temperature, viscosity of solution, agitation intensity, etc., have an impact on crystallization process. To an efficiently obtain high quality lactose, crystallization must be optimally controlled (Shi et al., 2006).

The operational optimization was performed using the experimental design adjustments for yield and MCSP, considering the coded independent variables. Optimization of crystallization layout, associated with the response variables, for each coded factor, is presented in Figure 3.

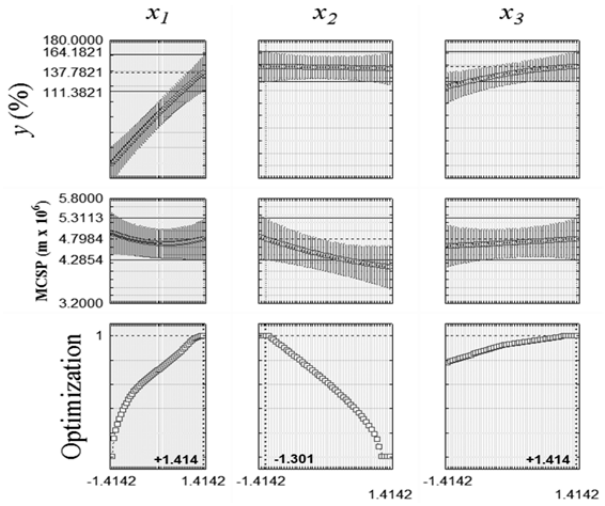

Figure 3: Optimization of crystallization process using Statistica software

The values of the coded variables, resulting in the best associated responses, obtained were +1.414 for supersaturation level, -1.301 for dimensionless vibration number, and +1.414 for crystallization time. The maximum supersaturation level and time associated with mild agitation were required to an efficient lactose crystallization process. Longer batch time and bigger seed size and mass (dense phase) shifts the CSD to the coarser fractions (Sander et al., 2009)

Predicted responses at the optimized condition were 137.78 % for yield and 4.798 × 10^{-6} m for MCSP. Optimized condition tested experimentally, in duplicate tests, presented an average yield of 138.96 ± 3.22 % and MCSP of 4.757 ± 0.092 ×10^{-6} m, which presents an efficient result related than predict (0.85% and 0.86 % errors, respectively) and CCD tests (Figure 4), reaching high yield and MCSP final results.

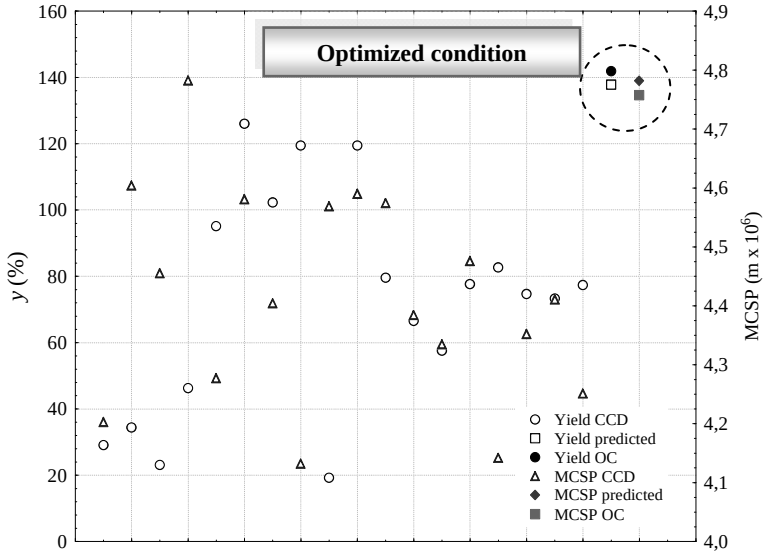

Figure 4: Yield and MCSP responses for CCD and optimized condition (OC) experiments

The conclusion was the effectiveness of design used. Decrease in relation to seeds was caused by the type of commercial lactose used milled. Milled lactose has a great quantity of micro particles that in suspension act as nuclei agent.

4. Conclusions

It was notable the MCSP decreasing with yield increase in design experiments, that can be explained by crystallization mechanism and a possible occurrence of nucleation phenomena associated with the operational condition of experiments. The number of crystals and associated total surface area of the crystals have a significant impact on the linear grow rate.

Predict models presents a quadratic correlation coefficient (r^2) of 0.9809 and 0.7009. Statistical analysis of predicted values in function of observed values have shown a smooth distribution (minimum errors) and a system without trends.

The experimental design allowed the evaluation of the factors - supersaturation level, vibration dimensionless and time that were significant in the lactose monohydrate lactose crystallization process, with high seed number, for the response variables yield and MCSP. Associated factors present relevant interaction among these, not being appropriate analyze them individually for the experimental range studied.

The MCSP response showed a characteristic increase for low supersaturation levels and high crystallization times, characterized by gradual crystal surface grow, avoiding secondary nucleation that is enhanced by high supersaturation levels and intense agitation. Related to yield response, it was notable a significant increase on yield of the design with supersaturation level increase, especially with high crystallization times. However, vibration dimensionless not show significant responses on yield, but the optimized region tends to low conditions.

The optimized condition trials reached an average yield of 138.96 ± 3.22 %, which presents a good result related than predict that was 137.78 %. MCSP obtained under optimized condition trials was 4.757 ± 0.092 × 10^{-6} m, which was an efficient result in relation to MCSP of central composite design and the predicted that was 4.798 × 10^{-6} m. This has shown the search for a balance between high productivity and final size of crystals, which is pronounced in industrial processes, that requires a product commercially favorable and an effective process**.**

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