Petroleum & Coal
ISSN 1337-7027

Available online at www.vurup.sk/pc
Petroleum & Coal 49 (1), 54-60, 2007

STUDY OF PARAMETERS AFFECTING THE CRYSTAL SIZE DISTRIBUTION OF SODIUM CHLORIDE IN BENCH SCALE CRYSTALLIZERS

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Received December 4, 2006, accepted June 25, 2007

Abstract
Sodium Chloride, being consumed by human being from long time ago, is one of the most important existing electrolytes in intercellular liquids of the body. The quality of Sodium Chloride has always been an important issue because of its daily consumption and also its application in food industries. There is variety of methods to increase the quality of sodium chloride. Crystallization is one of the most common industrial methods to prepare food grade sodium chloride. In this method, different operational factors such as heat load transferred to the crystallizer, residence time of magma in crystallizer, mixer speed and feed concentration affect the purity and size of the produced salt. In the current research work an appropriate bench scaled crystallizer with a reservoir volume of 10 lit has been designed and implemented for quantitative investigation of the effects of these factors on sizing of sodium chloride crystals.
In this paper, using Taguchi Analysis method, effects of all parameters on sodium chloride crystallization has been investigated in three different levels by running 9 double repetition tests (Totally 18 tests) and the results have been statistically analyzed using Qualitek-4 software. Test results show that heating load has the highest effect on crystal sizing of the final product while feed concentration and residence time have lower effects. The effect of mixer speed is negligible. Finally, the best operating conditions for production of crystalline salt has been determined to be feed concentration of 37% salt, mixer speed of 900 rpm, residence time of 60 minutes and heating load of 500 Watt.

Key Words: Sodium Chloride, Crystallization, DTB Crystallizer, Nucleation, Growth

1. Introduction

Crystallization is a common method for purification of mineral salt to produce standard food grade sodium chloride. In this method energy consumption is low and impurities of feed are removed during the process resulting in a product with appropriate mesh. Salts that are not purified contain components like Lead, Arsenic, Cadmium, Copper and Mercury which have irreversible adverse effects on human health. Existence of impurities will deteriorate the size of crystals and may lead to formation of other salts in the final product. In addition to impurities, other factors including feed concentration, heating load, residence time, crystallizer form, mixer speed and mixture type affect the mesh of sodium chloride which have been investigated in this paper. In literature, effect of mixer speed on salt crystals size has been investigated [1] but in this research, for the first time, the simultaneous effect of the four mentioned parameters i.e., heating load to crystallizer, magma residence time in crystallizer, mixer speed and feed concentration has been investigated on nucleation and nucleus growth. So control of crystal size distribution in the outlet of crystallizer is dependent on control of nucleation and nucleus growth process. Lots of researches have investigated the nucleation mechanism and have incorporated the results of their studies to simulate the nucleation kinetics as a function of the available operating conditions which can be used in industries. Researches focus on obtaining relations which can be used in industrial scales. In the current

research work, special attention has been given to identification of effective factors in sodium chloride production process in order to optimize the production line and quality of this product.

1. 1. Crystallization Process Assumptions and Selection of Crystallizer Type

As nucleation control and nucleus growth in a crystallizer is very complicated, MSMPR process will be the best possible choice for experiments [2].

MSMPR Process assumptions:

1. This process is stable, i.e. all parameters including temperature, density, viscosity, penetration coefficient, nucleation, nucleus growth rate, solids concentration (percentage), solid sizes, etc. are time invariant.
2. Crystallizer is homogenous, i.e. stability of the process is maintained throughout the whole points of the crystallizer.
3. There are no solid particles in crystallizer feed.
4. Output product has got the same properties as the suspended solid inside the crystallizer.
5. Crystal particles are not broken or combined in the crystallizer.
6. Crystal's Shape Factor is independent from its size [Mullin (1993)] [3].

Continuous process shall not be used in the experiments as in this case, the operating conditions of the crystallizer can't be exactly controlled and hence discrete process will be incorporated. As temperature has got the lowest effect on solubility of the sodium chloride, evaporating crystallizer will be the best choice for crystallization of this material [2]. Considering the above explanation the most important of which is the easy control of crystallization process, DTB evaporating crystallizer has been selected for running of experiments. Selection, implementation and incorporation of draft tube baffle crystallizer is a new approach used for the first time in this research work. In next sections of this paper the operation of this crystallizer will be investigated.
Figure 1 illustrates an evaporative crystallizer which is equipped with an external circulating system to remove small particles and enhance the concentration of liquid. The structure of this crystallizer is similar to that of the condensed DTB with a difference that in this crystallizer the crystalline solids are separated from the soluble phase as the solution evaporates. In DTB evaporative crystallizers, fluid circulation plays an important role in the amount of nucleation and hence a variety of crystallizers, which can internally circulate the fluid have been designed. If circulation is not naturally done in the crystallizer, a pump will be used for this purpose. In DTB crystallizers, the crystalline salt is suspended by means of the impeller and the slurry is directed towards the liquid surface decreasing the rate of super saturation. The cooled slurry moves towards the bottom of vessel and is again circulated by impeller, which causes that to be mixed with the warmed up solution. This crystallizer has some stationary zones causing separation of small and large particles during fluid circulation. Small particles are moved out of the crystallizer via a fluid circulation tube, mixed with a portion of input feed and then entered to a heat exchanger to be warmed and dissolved. Gravity causes the large particles move towards the bottom of crystallizer where they are pumped out [Daudey and De Jong (1984)] [1]. Directing the small particles out of the stationery zone and dissolving them enhances the magma super-saturation which will result in growth of existing crystalline.

1. 2. Determination of Crystallizer Specifications

Considering the achieved researches, the best approach to investigate the effects of the four aforementioned parameters is to use a non-continuous system in which the effects of parameters variation on the crystals sizing can be quantitatively measured. Volume of crystallizer has no effects on crystals sizes and only influences corrosion rate of the crystals and hence a crystallizer with a volume of 10 liters made of Pyrex glass has been implemented for this purpose which is quite suitable for bench scale study. Based on crystallizer volume, growth linear velocity and residence time are calculated form the following equations [3]:

$$G = [\, 30/\alpha\, \rho_s\, K_R\, L_m{}^4\, N^9]^{1/i-1} \quad (1)$$

$$L_m = 3.65\, G.\tau \quad (2)$$

A baffle is used to control the flow pattern and decrease the destroying effect of mixer speed. Also baffle has been incorporated to prevent turbulence flow [4,5].
In the implemented crystallizer, the mixer is of propeller type to minimize the rate of nucleation and also increase the rate of crystal growth (compare to other type of mixer such as blade type).
This kind of mixer has the following advantages:

- Results in a proper flow pattern
- Doesn't destroy the suspending particles
- Cleans itself while spinning
- It is controllable in a wide range of speeds
- It has a reasonable power consumption
- It is possible to provide the required turbulence by inclination of the shaft

Equation (3) is used to calculate the minimum mixer speed to suspend all particles (Njs).
As regards that crystal with average length of 350 micrometer, is standard size for production, so the mixer speed shall be 450 rpm (eq. 3).

$$N_{js} = \frac{su^{0.1} dp^{0.2} \left(\frac{g\Delta\rho}{\rho_1}\right)^{0.45} (x)^{0.13}}{L^{0.85}} \qquad (3)$$

2. Experiments

2. 1. Feed and Equipment

The feed used in the experiments will be unpurified salt. The experiment set consists of crystallizer, condenser, variable speed mixer, heating source, vacuum compressor, output water storage vessel, thermocouple, pressure indicator and connecting tubes as illustrated in Figure 3. 10 litre crystallizer (diam. = 16 cm, height = 50 cm) has been made of glass to avoid corrosion caused by salt. Mixer speed can vary from 0 to 2500 rpm. As the mixer shaft is long, a teflon part has been incorporated in the bottom of the crystallizer which will guard the internal glass body and baffles and will cause the shaft vibrations at the lower part of the crystallizer to be in eliminated.
Four nozzles have been implemented on the crystallizer cap for installation of shaft, pressure indicator, thermometer and vapor outlet . As the system will be under vacuum pressure, special considerations have to be applied to installation of shaft and baffle. The output vapor from the crystallizer is moved through a condenser, which is located beside the crystallizer, where it is condensed by cold water and is then collected in the condensate storage tank. A compressor has been used to provide the necessary vacuum in the crystallizer. An 800-watt heating element installed on the external side of crystallizer provides the required heat for solution evaporation and generation of turbulence. Figure 2 illustrates the DTB crystallizer specifically implemented for the experiments.

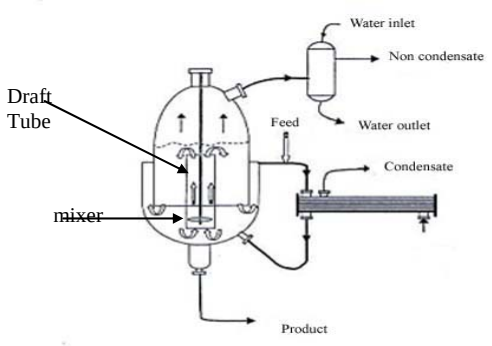

Fig. 1 – Schematic diagram of a DTB crystallizer

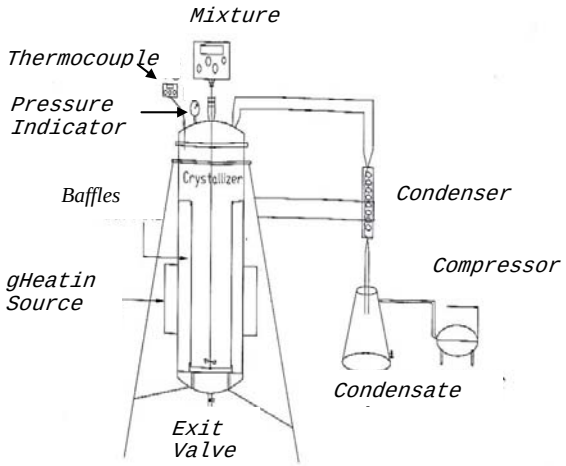

Figure 2: General scheme of crystallizer, which used in the research

2. 2 Arrangements of Tables for running of Experiments Based on Taguchi Analysis [6]

The experiments have been conducted to investigate the effects of the four aforementioned parameters on the crystals size.
All experiments are done at a constant temperature (As temperature and pressure are dependent, it can be concluded that pressure will be constant too). Three different values have been assigned for each parameter as shown in Table 1.
Considering the number of parameters and their values, the total experiments to be carried out are 3^4, i.e. 81. To reduce the number of experiments, Taguchi Analysis Method has been used. Taguchi Analysis also helps to organize the experiments and analyze the results. The required number and condition of experiments are obtained by Qualitek-4 software as shown in table 2. The procedure for running of 9 experiments by which the effects of parameters on crystal sizing can be investigated has been also given in Table 2. To increase precision, each experiment has been done twice and the average quantities have been reported.

3. Procedure of Experiments

First of all, salt solution with under saturated concentration is prepared and transferred to the crystallizer. After installation of pressure indicator and thermometer on the crystallizer, the compressor, mixer and heating element are turned on. At this time, temperature and pressure are recorded. When droplets of condensed water are observed, it is concluded that the system has been stabilized. At this time which is the beginning of experiment, boiling temperature and pressure are also recorded.
Final point of experiments is actually the residence time whose value has been mentioned in Table 2 for each experiment. At the end of the experiment, vacuum is disconnected from the system and the crystals accompanied with magma are directed out of the crystallizer to a 300-mesh sieve through an exit valve where they are washed with distilled water, dried with hot air flow and sorted out using a shaker-sieve system for 6 minutes. The sieves used in the experiments have meshes of 40, 60, 80, 100 and 200.

Table 1: Parameters under Study

No	Parameter	Surface 1	Surface 2	Surface 3
1	Concentration (g of NaCl per g of soln.)	0.24	0.255	0.27
2	Mixer speed, (rpm)	500	700	900
3	Residence time, (minute)	30	45	60
4	Heating power, (watt)	300	500	800

Table 2: Procedure of Experiments

Exp. No	Variable Type			
	Concentration (g NaCl /g soln.)	rpm	Minute	Watt
1	0.255	500	30	300
2	0.255	700	45	800
3	0.255	900	60	500
4	0.240	500	45	500
5	0.240	700	60	300
6	0.240	900	30	800
7	0.270	500	60	800
8	0.270	700	30	500
9	0.270	900	45	300

4. Experiments data

As previously mentioned, the goal of the experiments is to study the effects of four aforementioned parameters, i.e. heating load, mixer speed, residence time and primary solution concentration to yield larger crystals of sodium chloride.

To do this, a base point has to be considered for comparison purposes. Therefore, one of the best pure crystalline salt in the market was selected as a base material for this research. The selected crystalline salt was sorted out by means of a shaker-sieve system. As shown in Table 3, mass percentage of mesh 60 crystals is has the highest value and those of the mesh 40 and mesh 80 are approximately equal. This indicates that the crystalline salt distribution is quite good. According to the results denoted in Table 3, the higher mass percentage of the crystalline salt with mesh 60 was selected as a criterion for identifying of a suitable salt. The results of experiments 1 to 9 were classified based on this criterion. Table 4 presents the results of the experiments.

Table 3: Results of Sizing of the Crystals (Base Point)

Mesh	weight (g)	wt (%)
40	10.8	12.76
60	62.74	74.12
80	10.134	11.97
100	0.6	0.7
200	0.37	0.44
Sum	84.64	100

Table 4 – Results of the Experiments

No	Solubility ($gr_{salt}/gr_{solvent}$)	Mixture Speed (rpm)	Residence Time (minute)	Heating Load (Watt)	Percentage of Sizing on Sieve Mesh 60
1	0.342	500	30	300	0
2	0.342	700	45	800	59.83
3	0.342	900	60	500	79.34
4	0.315	500	45	500	44.63
5	0.315	700	60	300	0
6	0.315	900	30	800	15.4
7	0.370	500	60	800	60
8	0.370	700	30	500	56.22
9	0.370	900	45	300	31.52

5. Discussion and Results

Considering Table 3, mesh 60 is selected as a criterion based on which the Taguchi Analysis shall be carried out. As illustrated in Table 4 effective parameters on crystal sizes will be classified as heating load, feed concentration, residence time and mixer speed respectively.

As indicated in Table 5, mixer speed has no important effect on crystal sizing and hence its effect is ignored. Table 6 shows all the parameters with their respective effect percentage.
Experiments show that increase of heating load enhances the rate of evaporation, which will subsequently lead to increase of super-saturation and occurrence of nucleation, but this does not imply production of large crystals. Heating load must have a proper value to result in suitable growth of crystals. If heating load is considerably increased, it will cause disturbance in the crystallizer, which will subsequently result in crash of the crystals, which had already started growing up. On the other hand, decrease of heating load extends the production time duration. Analysis of experiments shows a specific amount for heating load, which will lead to production of appropriate crystals in the shortest possible time interval.

Table 5 Importance of Operating Parameters in Crystal Growth Process (without error)

Parameter Type	Importance Percentage
Concentration (g_{salt}/g_{soln})	24
Mixer speed (rpm)	1.2
Residence time (Minutes)	15

Table 6 Importance of Operating Parameters in Crystal Growth Process (with error)

Parameter Type	Importance Percentage
Concentration (g_{salt}/g_{soln})	22.8
Mixer speed (rpm)	0
Residence time (Minutes)	13.8
Heating load (watt)	58.6
Error	4.8

Results of the experiments show that increase of feed concentration will cause increase of mesh 60 crystals, on the other hand experiments show that increase of feed concentration is more effective on crystals growth than nucleation rate.
Increase of the residence time, at presence of nucleuses and suitable operating conditions, will result in a better growth process and yield of larger crystals. If the residence time in crystallizer is long, the grown crystals break and are divided into new nucleuses. In other words, growth of crystals is limitedly increasable based on system conditions. As an example if the mixer is of blade type and its speed is high, increase of residence time will have no effects on growth of crystals because when the crystals are a bit grown, they will face collisions with each other, the blade of the mixer and the body of the crystallizer which will cause them to be broken to smaller ones [3].
The results of the experiments confirm this matter. Considering the nucleus equation [3]:

$$B^o = K_B N^h G^i M^j \ (\#/m^3.s) \qquad (4)$$

Increase of mixer speed enhances the rate of nucleation and as a result more nucleuses will be produced in the process environment. Therefore, high mixer speed causes production of crystals with small sizes as in this case the crystals experience more collisions and don't find the opportunity to have growth. This matter is obviously observed when the mixer is of blade type. When the mixer is of impeller type, there are less collisions between crystals and the mixer because of its shape and hence the crystals are less broken. As a result, in identical operating conditions, the product of a crystallizer with impeller mixer has larger size than that with a blade type mixer. Of course in both cases increase of the mixer speed causes production of smaller size product. The result of experiments confirms this matter (increase of crystals with 60 mesh). It is noticeable that the mixer speed shall not be less than a specific quantity called N_{js} obtained from equation (1). Considering the laboratory conditions, N_{js} has been calculated to be 450 rpm.

Table 7 – The Optimum Operating Condition Obtained from Software

Operating Parameter Type	Optimum Condition
Solubility($g_{salt}/g_{solvent}$)	0.37
Mixer Speed (rpm)	900
Residence Time (Minutes)	60
Heating Load (Watt)	500

6. Conclusion

To do the experiments, draft tube baffle crystallizer has been used as control of the crystallization process is more straight forward in it. Selection, implementation and incorporation of this crystallizer for crystallization of sodium chloride has been an innovation in this research work. The experiments show that among the parameters under study, heating load has got the most considerable effect on sizing of crystals while the effect of mixer speed is very low and negligible.
Analysis of the experimental results by means of Qualitek-4 software shows that the conditions denoted in Table 7 are the best ones for production of crystals with proper size. As illustrated in Table 7, high amount of super-saturation, high mixer speed, high residence time and medium heating load yield the proper product. The result of software is completely in compliance with the experimental observations.

Nomenclature

D_p	particle diameter (m)	q	constant
D	crystallizer diameter	S	constant, shape coefficient
g	acceleration gravity (9.81 m/s^2)	X	solid particle partial weight
G	linear velocity of crystal growth (m/s)	ρ_s	solid particle density (kg/m^3)
I	constant	$\Delta\rho$	density difference between solid particles and liquid (kg/m^3)
K_R	constant	ρ_1	liquid density (kg/m^3)
L_m	averaged size of crystal particles (m)	υ	dynamic viscosity (m^2/s)
L	mixer diameter (m)	α	constant
N	run of mixture	#	number
N_{js}	minimum of run of mixture for suspending solid particles		

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