Revista Internacional de Métodos Numéricos para Cálculo y Diseño en Ingeniería

Exploration of the Influence of High Rate Cooling Crystallization Parameters on the Preparation of Ammonium Perchlorate Crystals Using Taguchi's Method

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Abstract

'"Preparation of fine ammonium perchlorate (AP) particles and thermal behaviour of it are known as the most important challenges in the crystallization and propellant sciences. We introduced an effective method of fine AP preparation named high rate cooling crystallization (HRC). This work presents application of Taguchi approach for optimizing the HRC method. In this study a set of experiments was designed according to the Taguchi L9 array. Experimental design was originated from four effective parameters containing; temperature, pressure, solvent type (water and ethanol) and anticaking agent- sodium dodecyl sulfate (SDS). After preparation of fine AP particles average particle size (APS) was measured using laser mass spectrometry (LMS-30). APS, contribution, and the analysis of variance (ANOVA) were used to investigate performance factors affecting the operation condition. According to the data obtained, anti-caking agent and solvent type were represented the main effect on the preparation of micronized AP by using HRC method. Furthermore, the optimum condition in preparation of fine AP particles by using HRC method was ethanol solvent, 10atm pressure, 0.4 % SDS, and 60˚C temperature. Thermal behaviour of the optimized particles was investigated using TG/DSC analysis. Finety of the particles positively shifted the low temperature decomposition (LTD) region of fine optimized sample in comparison with coarse pure AP. Crystallinity of sample was confirmed by SEM and XRD analysis.

Keywords: Ammonium perchlorate.Taguchi's method.Factor contribution. Average particle size. Optimization. High rate cooling method.

Submitted: 28/04/2022

Revised: 20/06/2022

Accepted: 26/06/2022

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1. Introduction

Ammonium perchlorate (NH_4ClO_4, AP) is as an outstanding oxidizer and has been widely used in composite solid propellants. Therefore, the burning behaviour of composite solid propellants is closely relevant to the thermal decomposition of AP particles [1-2].Similarly, the function of oxidant used in the propellant formulation could be improved by enhancing the morphology and sphericity and smoothness of its surface. This is because of the viscosity reduction as the particles slip on each other more easily [3]. Also, the production of fine AP is an interested challenge.Crystallization from solution for the means of production of fine particles is known as complex process considering in chemical engineering and separation science. This process mainly is described in three mechanism; first is the nucleation, the next is aggregation, and the last is the growth [4-6]. Produced particles were originally limited by these three mechanisms [7-9]. Also, clearly production of micronized particles is based on the crystallization principles. Many methods are introduced in solution crystallization containing; cooling crystallization, evaporating crystallization, anti-solvent crystallization, solvent layering, sublimation, and changing the cation or anion. Cooling crystallization is known as the most familiar and lowest cost-consuming method of crystallization in chemical production [10- 12]. In the cooling crystallization method activation energy super-saturation which is decreased by temperature decrement. According to the cooling crystallization theory the cooling rate and the temperature of process are considered as definitive factors [13-16]. High cooling rate make the crystallization procedure tends to the intensive nucleation. Production of superfine energetic materials such as ammonium

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perchlorate (AP) particles with reference to safety concerns, simply scale up, average particle size (APS) and sphericity of crystals is even a challenge to material science scientists. Our literature showed that fine AP particles can be prepared by using some methods including; anti-solvent crystallization, spray drying method, freeze drying method and ball milling method [17-21]. Also, cooling crystallization is used preparing AP particles, by considering that in this method AP particles have great size [22] (up to 500 μm). In this method of crystallization temperature of the saturated solution, cooling rate of the saturated solution and impurities influenced the produced particles. Among high cooling rate the produced particles represented low APS [23]. Also the temperature of the saturated solution mainly determined the solubility of the solvent. Originally, the temperature of the solution directly affect the solubility- it may increase or decrease the solubility according to the solvent features [24]. Solubility of the water for most inorganic compounds is increased by increasing the temperature. According to the crystallization principles the solubility not only increase the solute molecules in the solution, but also decreases the nucleation versus the aggregation [25- 26]. This phenomenon causes the solubility plays ambiguous role in aspect of terminating the particles size. Other way of the prevention in crystal growth and micronization science is application of anti-caking agents in the crystallization process [22, 27-28]. Anti-caking agent limited the humidity of crystals and prevents formation of high mass crystalline [23-30]. These condition to the preparation of the fine particles are so required for optimization. According to the reported researches Taguchi method of optimization was low-cost technique. Also Taguchi is known as a powerful method in experimental design; Because of its possibility to present small amount of experiment number and therefore time and cost savage. The most important property of Taguchi method is that it can strongly decrease the experiments number especially in optimizing the performance of large number of process parameters [31-32]. Also, it is powerful method in determining impact of the factors [33-34]. Promoting to these fundamentals, we introduced a new method for AP crystallization, which temperature fast decrease and nucleation rate overcomes the crystal growth significantly. The introduced method for high cooling rate in this process was possible using the temperature reduction in thermal converter.

Generally, four important factors impact on the cooling crystallization method of preparing micronized particles. Solution temperature, cooling rate of the solution, pressure and anti-caking agent are four key factors considered to be investigated in optimization of the micronization condition. The Taguchi approach of L9 array is considered to examine the impact of these four key factors. New simple and safe method in order to micronization of AP particles is introduced, and the produced particles size is analysed by laser mass spectrometry (LMS-30). Analysis of variance (ANOVA) is evaluated to classify the factor contribution. And the optimum proposed condition of the Taguchi analysis for the AP particles were investigated and validated respectively with SEM. Correspondingly, crystallinity of the produced optimal AP particles is considered by XRD analysis. Finally, the main feature of the products (thermal behaviour of optimized fine AP sample) is investigated by testing in TG/DSC method. Therefore, the key note of this research is to recognize the AP micronization parameters (temperature, solvent type, anti-caking agent and pressure) affecting in the reduction size of AP particles and subsequently the thermal behaviour of it.

2. Experimental

2.1. Materials

Ethanol (99.99%), SDS (99.99) and AP (99.8) were purchased from Merck. The chemicals were used without further purification and the materials in process production were stainless steel. Double distilled water was used in all experiments.

As shown in the Figure 1, the micronization bench setup was contained from 2-liter double-wall tank, heating system, pressure supplier, high rate cooling exchanger, and product tank. High rate cooling exchanger was known as the main section of the setup which is named cooling crystallizer. About 400 g of AP was dissolved in 1000 mL of distilled water at 70 °C in the solution tank then was injected in the cooling exchanger using pressure given with air cylinder. According to the pressure value the time spending to exhaustion was varied about 5-20 seconds. After cooling, the main solution which was containing the crystals was collected in product tank. Finally, the produced AP particles were filtered and were washed three times with 25mL toluene as non-solvent and dried at 70 °C. Obtained AP samples from HRC method were used for further investigation.

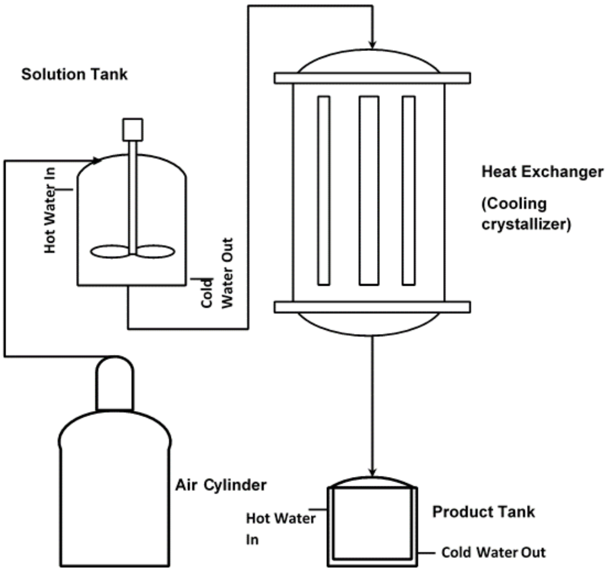

Figure 1. *Schematic representation of HRC system for preparation of fine AP particles.*

2.2. Crystal size determination

In order to investigate the particle size distribution and average crystals size, laser mass spectrometry (LMS-30) was used.

The X-ray diffraction (XRD) was used to investigate the crystalline quality. Confirmation for the crystal structures is completed through XRD. The fine AP particles, making a weighted dispersion on a glass slide, was assessed using an X- ray powder diffractometer (XRD; Siemens D-500 with Cu Kα radiation). The sample was X-rayed using a Cu target tube, and depicted to all lines. A mono-chromator was handled to select the K1 line (θ= 1.54). The terminating angle ranged from 5◦ to 80◦ of the diffraction angle (2θ), and the counting time was considered 1 s/step in steps of 2θ= 0.05◦. The examining rate was 3◦/min. The excitation current and voltage were 40 mA and 30kV, respectively.

2.3. Optimization experiments

According to the effective factors of the micronization process the optimization tests were planned. Four factors were selected effecting on the produced AP particles. Taguchi L9 array was used to design experiments. The parameters and the levels are arranged as the Table 1 and designed experiments in the L9 *array were presented in Table 2.*

2.4. Thermal investigation

Thermal behaviour of AP samples was characterized by thermal gravimetric analysis (TGA) and differential scanning calorimetry (DSC) methods. The TG/DSC curves were performed using Shimadzu DSC-60 by the temperature range from 25 up to 600 ˚C and heating rate was fixed on 10 K/min. For each analysis, the AP powder sample of about 12.0±0.2 mg were taken in crucible. Nitrogen (99.99%) was used as purge gas by flow rate 50 mL/min.

3. Result and Discussion

3.1. Taguchi design

The Taguchi approach in the design of experiment (DOE) has been commonly considered in industries to optimize production condition. Basic of the method is according to comparison between factors affecting the process. By using Taguchi design of L9 arrays main effect of factors can be investigated. Taguchi employs a minimal number of experiments by testing pairs of combinations. This saves both time and resources. Also, in this research Taguchi method is used to identify optimization condition of process in fine AP preparation. According to our experimental efforts in the cooling crystallization, temperature and solvent type affect the operation condition. These parameters play important roles in micronization process: pressure, anti-cake agent and solvent type. After each experiment APS for the samples are analysed. APS for the produced particles are presented in Table 2.

3.2. Particle characterization

As shown in Figure 2, the images of the samples run (7-8) obtained by optical microscopy, represent the smaller particles than all other samples. Also, the largest particles are shown in sample run1. Furthermore, the particles of samples run (4-5) confirm spherical particles. It is shown in Fig. 2 samples (1-3) that the particles tend to form in needle like shape, which, the sample 3 is more needle than the sample 2 and 1. Moreover, this tendency is not observed in other trials of equal solvent type. Clearly it shows force of SDS on the crystal habit of AP particles in the presence of water molecules. As mentioned above, this phenomenon cover two results; the molecular interaction and the AP solubility of solvent. Firstly, Sodium dodecyl sulfate (SDS) originally is known as an orgonosulfate class of chemical compounds. So, in structure the hydrocarbon tail merged with a polar head group perform the compound amphiphilic properties and make it useful as an emulsifier. These properties caused the SDS compound interacting with the polar compounds of H_2O , NH_4ClO_4 and partial-polar compound of CH_3CH_2OH. In the trials which water was used as solvent, interaction between the H_2O molecules and SDS molecules is the polar-polar interaction. In this condition the SDS molecules are in competition with the H_2O molecules interacting with AP molecules. As the polarity difference between H_2O and SDS is small, then, adsorption of each compound on the AP molecules (on crystal) is similar. Additionally, the polarity difference of SDS-H_2O pair is lower than that of SDS-CH_3CH_2OH pair. Beside the polarity difference between SDS and AP molecules are smaller in corporation of them on the crystal surface. Also, the SDS can be considered as the effective crystal habit in water solution than in the ethanol solution. Secondary, the AP is more soluble in water than in the ethanol, so, the amount of AP per water molecules is high than the amount of AP per ethanol molecules in saturation conditions. Subsequently, the SDS per water ratio is more than the SDS per ethanol ratio and SDS is more interactive crystallization inhibitor in water solution than in ethanol solution. Crystallization inhibition can be considered the main cause of the needle like morphology.

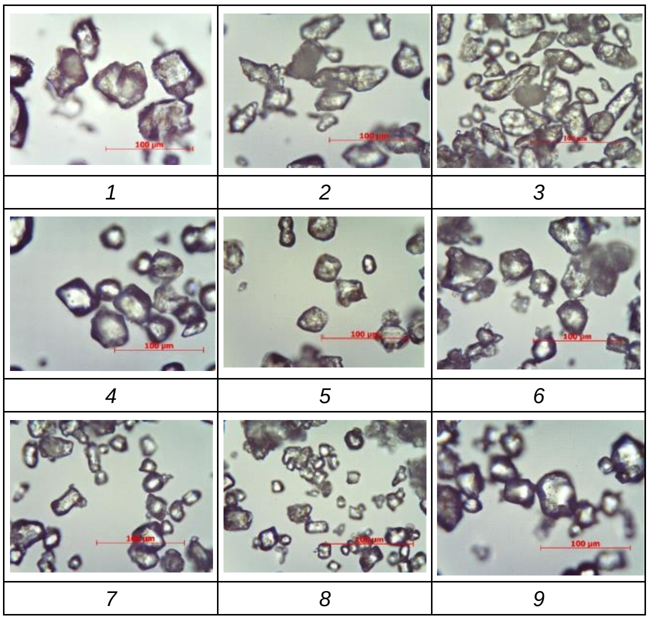

Figure 2. *Optical microscopically images of AP particles produced according to the Taguchi trials.*

3.3. Optimization of micronization process parameters

In this research effect of the several process parameters i.e., saturated solution temperature (T), injection pressure of process (P), sodium dodecyl sulfate (SDS) as anticaking agent and solvent type on the produced AP particle size is investigated. The orthogonal L9 array of Taguchi experiment design is used to investigate the factors effect. Table 1, exhibits the experimental factors and the related levels. Average particle size of produced AP particles as the response are represented in the last column of Table 2.

Table 1. The various parameters selected and their respective levels in the present experimental design.

Process parameter	Symbol	Level 1	Level 2	Level 3
solvent	S	H_2O(W)	H_2O /C_2H_5OH(W/E)	C_2H_5OH(E)
Pressure	P	5	10	15
Sodiom Dodecil Solfate(%)	SDS	0	0.2	0.4
Temperature (ºC)	T	40	50	60

Table 2. Experiments of Taguchi L9 array in the present experimental design and related average particle size.

Trial number	Solvent	P (atm)	SDS (%)	T (c)	APS (µm)
1	W	5	0	40	50.6
2	W	10	0.2	50	32.43
3	W	15	0.4	60	26.92
4	W/E	5	0.2	60	36.82
5	W/E	10	0.4	40	27.53
6	W/E	15	0	50	42.62
7	E	5	0.4	50	24.85

8	E	10	0	60	21.41
9	E	15	0.2	40	34.01

According to the Taguchi method, analysis of all factors are considered by minus considerable interaction. The first order of analysis is investigation of the factors singly effect on the produced results (AP particle size), finding of optimal condition and levels of factors, and then the validation of the identified condition with the real experimental results [35]. Therefore, the influence of each factor on the change of produced AP particles

by variation of each factor levels were investigated and presented in Fig. 3. Solvent type was the well-known factor effecting the crystallization process [36-37]. High solubility of solvent creates high amounts of solute in the solution and can causes more number and large size of crystals.

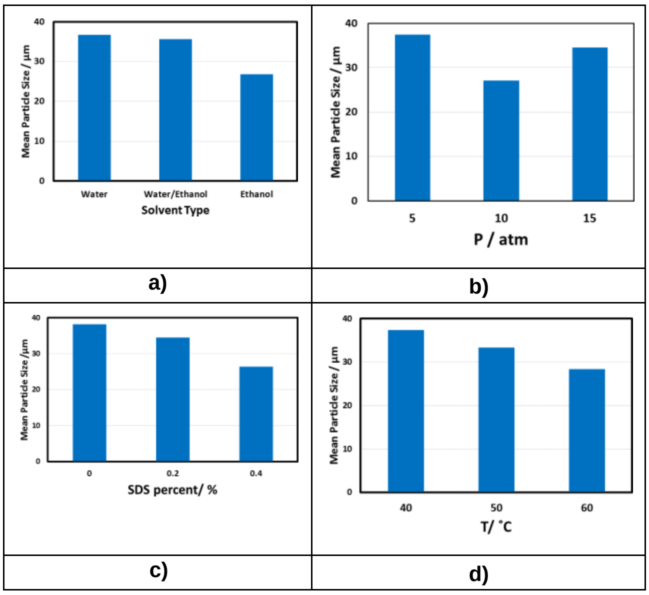

Figure 3. *Average particle size of AP particles obtained via high rate cooling method corresponds to the effect of each level for various factors: Solvent Type; Pressure Value; SDS Percent; Temperature.*

The results in Fig (3-a) represent the influence of solvent quiddity on the produced AP particles. Three types of solvent are used in this analysis; pure water (W), pure ethanol (E), and water/ethanol (W/E). Naturally, the mean size of AP particles relating to the pure water solvent represented the large APS. Similarly, the W/E solvent type represents large APS than that of E solvent. As formerly described, the solvent type according to the solubility limited the APS. High solubility, on the one hand increased the AP molecules ability creating of more nuclei, on the other hand increased growth rate of the produced nuclei. According to the Fig (3-a), it can be concluded that the crystallization of AP particles, unlike ethanol solution, in water solution was founded by creating of large critical nuclei size. As shown in the Fig (3-b) low pressure of injection caused large particle size. The injection pressure affect the retention time, inversely. Low pressure caused high retention time, then, facilitated the crystal to growth. Anti-caking agent (SDS) percent in the HRCmethod was studied to determine its effect on the produced AP particles. Fig (3-c) confirms that increasing the SDS value, decreased the APS. It indicated that SDS played an important role in prevention of particles aggregation to produce small particles. Fig (3-d) gives the influence of temperature levels on the produced particle size. By lowering the saturation temperature the APS increased. Finally, as shown in Fig. 3 all of the factors have considerable effect on the production of AP particle size using the HRCmethod.

3.4. Analysis of variance (ANOVA)

The ANOVA calculated results was given in Table 3. As represented in this table 3, all four selected factors; temperature (T), pressure (P), anticaking agent (SDS), and solvent type (S) played effective role influencing the process of preparing fine AP particles. As shown in Table 3, according to the ANOVA results, the factor SDS has more significant role

(31.6 % contribution) than other factors affecting the production of fine AP particles. On the other hand, the solvent type (S) and the process pressure (P) represented lower importance than the SDS. Also, the same role of these two factors in the HRCprocess can be concluded. The next factor is the temperature that have lower effect (17.7 % contribution) than all other factors in AP micronization process. According to the results obtained by ANOVA, the SDS by preventing the aggregation can be known as the most important factor of the process. Therefore, controlling the aggregation is known as the essential requirement of the HRCprocess.

Table 3. ANOVA results for the preparation process of fine AP particles by HRCmethod using L9 array.

factors	DOF (f)	Sum of Sqrs. (S)	Variance (v)	Pure Sum (S')	PercentP(%)
S	2	178.074	89.037	178.074	25.963
P	2	169.199	84.599	169.199	24.669
SDS	2	216.841	108.42	216.841	31.615
T	2	121.759	60.879	121.759	17.752

4. Conclusion

Thermal activities of AP particles is a notable factor of solid base propellants. Particle size mainly effect on the thermal behavior of AP particles. Also, the method high rate cooling (HRC) was used to prepare fine AP particles. We firstly, tried in this research to understand the main parameters effect on the process and to confirm the essential influence of them on the process by using Taguchi method. Optimized AP sample was investigated using SEM and XRD for the means of crystallinity. According to the data obtained, the solvent type in corporate wit anti-caking agent represent effective impact on the morphological properties of the AP crystals. Interaction between the ammonium, perchlorate, water, and the SDS molecules causes the AP molecules arrange along one dimension. Crystallization is a complex and regular collection of the molecules by chemical and physical interaction, so, the addition of the AP molecules can be developed in direction which the ammonium side of molecules was not blocked. According to the results, anti-caking agent played most important role in optimization of the HRC method. Solvent type is the next important parameter affecting the process production. Complimenting this two important factors concluded that the aggregation of the particles.

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