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Statistical optimization of supercritical carbon dioxide antisolvent process for preparation of HMX nanoparticles

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ABSTRACT

Taguchi robust design was employed as a statistical optimization method to optimize parameters of SAS process in order to prepare HMX nanoparticles. The current application of the Taguchi statistical optimization was highly efficient in optimizing of experimental variables which are effective in HMX particle size produced by the SAS technique. The effect of operation conditions such as: pressure, temperature, HMX concentration, solution flow rate, solvent identification, and flow rate of CO_2 on the size of micronized HMX particles under various levels was investigated. The effects of these variables on the particle size of produced HMX were quantitatively evaluated by the analysis of variance (ANOVA). The results showed that the size of HMX particles prepared by SAS technique could be tuned effectively by controlling the main parameters under their optimum level. Finally, the optimum conditions for preparation of HMX nanoparticles via SAS method were proposed. The results of ANOVA showed that 3.5 mol/l HMX concentration, 3 ml/min solution flow rate, cyclohexanone as solvent, and 70 ml/min flow rate of CO_2 are optimum conditions for producing HMX nanoparticles. Experimental observations showed that under optimum conditions of SAS process, the particle size of produced HMX is about 56 nm.

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1. Introduction

HMX (octahydro-1,3,5,7-tetranitro-1,3,5,7-tetrazocine) is a high explosive which is widely used in various formulations of plastic bonded explosives (PBX), double base propellants and composite propellants due to its high calorific potential, high density and smokeless combustion products [1,2]. HMX could be prepared in four different polymorphs that commonly known as α (orthorhombic), β (monoclinic), γ (monoclinic) and δ (hexagonal) phases. Among these various phases, β-HMX has the most stability one and possesses the highest explosion power which arises from its crystal phase and its high density. Therefore, preparation of HMX in β-phase would be exclusively interested [3].

The size and shape of energetic material particles have a significant influence on the different properties of propellants and explosive formulations [4–6]. Several properties of these compounds such as sensitivity, mechanical properties, content of total solid, density, homogeneity of formulation, and burning rate are dependent to the particle size of energetic materials [7,8]. The particle size reduction of energetic compounds could be lead to the reduction defects, voids and inclusions of crystal and hence reduction the sensitivity of the formulations and enhancing their safety [9,10].

In order to tailoring fine particles, various conventional methods including milling, liquid crystallization and spray drying could be employed [11–13]. But, these techniques are not suitable for treating energetic materials due to some drawbacks such as difficulty of controlling particle size and particle size distribution, high residual solvent, high shear forces, and unexpected problems during their size reduction process containing thermal explosion of energetic materials [14,15]. Supercritical fluids (SCFs) techniques may be useful to overcome these drawbacks of classical micronization processes and have been applied by researchers to size reduction of explosives [16]. The supercritical fluid is characterized with diffusivities considerably higher than those of liquid, which make it suitable for fast supersaturation of solute and then precipitation of small particles of targeting material.

Until today, various techniques for particle formation via supercritical fluid technology including two general processes have been used: supercritical antisolvent (SAS) process [17,18] and rapid expansion from supercritical solutions (RESS) process [19,20]. The SCFs based techniques have many advantages rather than classical micronization processes such as preventing thermal degradation of the target compounds, no mechanical damage, and no residual solvent problem [21]. The main work for an explosive particle formation via SCFs processes such SAS recrystallization and RESS process is preparation of fine particles with various sizes and shapes

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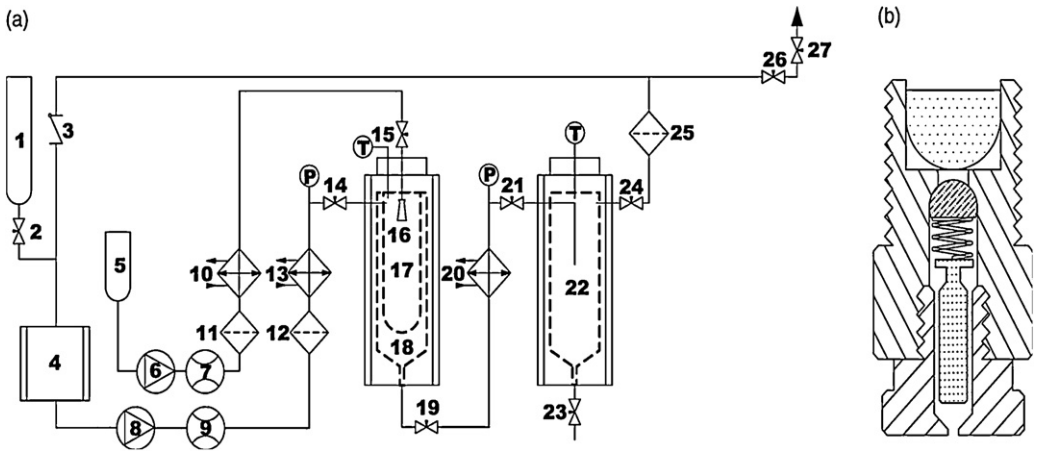

Fig. 1. (a) Schematic of the experimental setup used for SAS process and (b) and the cross section of used nozzle. Taken from Ref. [18] with permission.

by controlling the supersaturation and nucleation rates of the explosive in the SCF media using variation in solvent strength. Especially, the SAS process as a recrystallization technique is a promising process for production of solvent-free explosive particles with fine sizes [22].

The particle size of compounds which prepared by the aid of SAS process is dependent on the various operation parameters and procedure conditions; thus, optimization is an important step in developing SAS technique for size reduction of various materials [23]. Generally, two systematic procedures including simultaneous and sequential methods are used in optimization of the experiments [24]. The main disadvantages of sequential methods are slow convergence on complex response surface and difficulty in dealing with response surface with high dimensionality; while, simultaneous optimization techniques do not suffer from such problems [25]. On the other hand, an obvious disadvantage of the full factorial designs is the number of experimental trials required which increases geometrically by increasing number of variables. Fortunately, this required number of experimental trials is minimized by use of fractional factorial experiments, such as orthogonal array (OA) designs [26].

Since statistical experimental design methods could be carefully explored the experimental space while studying several variables via a small number of observations; Taguchi robust design as a statistical experiment optimization has been widely used in various procedures optimization [27–29]. Therefore, various control factors could be simultaneously investigated and optimized by the aid of Taguchi statistical design. The aim of this study was applying the Taguchi robust design to the optimization of various parameters affecting on particle size of HMX prepared via SAS process and evaluating the effect of these parameters on the size of produced HMX particles. Our goal in this study was optimization of conditions to reduce the size of produced HMX particles and preparation of HMX nanoparticles. To the best of our knowledge, various reports could be found on the micronization of HMX by various techniques [1,30–34]; but, until today there is no report on the preparation of HMX nanoparticles via SAS process. Therefore, optimization of SAS process parameters affecting the size of HMX particles and preparation nanoparticles of this explosive is interested.

2. Experimental

Cyclotetramethylene tetranitramine (Octogen, HMX) was used to investigate the possibility for formation of its nanoparticles using compressed SAS process. It should be noted that HMX is a high explosive. Thus, during its operation much care is required to prevent unexpected hazard such as accidental explosion. Fortunately, in the SAS process which used in this study, the solid of explosive is dissolved in a liquid, and then a supercritical fluid, which is miscible with the liquid but in lower solvent power to the explosive, is added to recrystallize the HMX. This wet operation process is safer for some difficult-to-handle high explosive such as HMX.

Analytical grade acetone, cyclohexanone, and γ-butyrolactone as solvents were used as received from Merck Company (Germany). Carbon dioxide (99.99% purity), contained in a cylinder with an eductor tube, was obtained from Sabalan Co. (Tehran, Iran).

The scheme of the used SAS apparatus is shown in Fig. 1. This SAS system is consisting of a precipitation chamber and a gas–liquid separation chamber. As could be seen in Fig. 1a, the CO_2 is cooled by a cooler (4) before being compressed via a liquid pump (8) and the pressure is controlled with a backpressure-regulating valve. After pre-heating of CO_2 in a heat exchanger (13), CO_2 enters to the precipitation chamber (18). Simultaneously, the solution containing target compound is pumped, heated and fed to the precipitation chamber (1000 ml) through a stainless steel nozzle (16). Meanwhile, Fig. 1b shows a close up with the details of the nozzle, which is a laser-drilled orifice (150 μm inner diameter). This nozzle is placed at the top of the precipitation chamber; which is located in a distinct inlet point from the CO_2. A stainless steel filter (17) with 200 nm pore sizes was put into the precipitation chamber to collect the micronized particles and to let the SC-CO_2/organic solvent mixture pass through. The flow rate of the mixture that leaves the precipitator is controlled via valve (21), which is located between the precipitation chamber and the gas–liquid separation chamber (22), where the mixture suffers a decompression (pressure <5 MPa) to induce separation of the CO_2 from the organic solvent.

The prepared HMX samples via SAS process under various experimental conditions were characterized by scanning electron microscopy (SEM). Scanning electron micrographs were recorded using on a Philips XL30 series instrument using a gold film for loading the dried particles on the instrument. Gold films were prepared by a sputter coater model SCD005 made by BAL-TEC (Switzerland).

To optimize experimental conditions of SAS process, for the micronization of HMX, an experimental design approach was followed. Various operation parameters including pressure, temperature, HMX concentration, solution flow rate, nature of the solvent, and flow rate of CO_2 were investigated in three levels using a L_{18} array proposed by Taguchi robust design as shown in Table 1. As could be seen in this table, each of variables was studied under three different levels. These levels were selected based on the

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