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Research paper

Optimization of the Synthesis Parameters and Analysis of the Impact Sensitivity for Tetrazene Explosive

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Abstract: Tetrazene [1-(5-tetrazolyl)-4-guanyltetrazene hydrate] is widely used in ordnance systems as a sensitizer of primer compositions, for use in both percussion and stab applications. It can be synthesized by reacting aminoguanidinium bicarbonate (AGB) with sodium nitrite (NaNO₂) in the presence of acetic acid. However, little is known about its optimal synthesis parameters in the manufacturing process. In this study, Taguchi's experimental design method was used to further improve the yield of tetrazene. Under the condition that the molar ratio of AGB to NaNO₂ was fixed at 1.0:1.2, a L₉(3³) orthogonal array with three control factors and three levels for each control factor was used to design nine experimental conditions. The experimental data were transformed into a signal-to-noise (S/N) ratio to analyze and evaluate the experimental conditions of the optimal parameter combination for the maximum yield of tetrazene. Verification of the results indicated that the optimal synthesis parameters were as follows: pH value of AGB solution was 5.0, reaction temperature was 35 °C and reaction time was 6 h; the maximum yield of tetrazene could reach 83.7%. Furthermore, the synthesized tetrazene was identified and characterized by elemental analysis (EA), nuclear magnetic resonance spectroscopy (NMR), Fourier transform infrared spectroscopy (FTIR), optical microscopy (OM),

particle size distribution analysis (PSDA), X-ray diffraction (XRD) and differential scanning calorimetry (DSC), and its impact sensitivity was determined using a BAM Fallhammer apparatus and the Bruceton analysis method.

Keywords: tetrazene, yield, optimal synthesis parameters, Taguchi design method

1 Introduction

Tetrazene [1-(5-tetrazolyl)-4-guanyltetrazene hydrate] is a primary explosive that is widely used as a sensitizer in primer compositions for a variety of ammunitions. It is as sensitive to impact as mercury fulminate (MF) and diazodinitrophenol (DDNP). Its power is slightly higher than that of MF, but it does not have sufficient initiating efficiency to permit its use alone as an initiatory explosive. However, tetrazene can act as a good sensitizing agent to other initiating explosives and mixtures, and therefore is extensively used in percussion cap compositions, where as little as 2% in the composition results in an improved uniformity of percussion sensitivity. Although tetrazene is unstable, it is still used as a sensitizer in percussion cup compositions and thus far no other suitable alternative has been identified [1-4].

Tetrazene was first discovered by Roth and Hoffman in 1910 [5] and the molecular structure was determined by Duke in 1971 [6]. It can be prepared from either aminoguanidinium nitrate (AGN) [7-9] or aminoguanidinium bicarbonate (AGB; $\text{CH}_6\text{N}_4 \cdot \text{H}_2\text{CO}_3$) [10]. AGN is a more expensive raw material, but it can produce shorter needle-shaped tetrazene crystals. However, synthesizing tetrazene from AGB is substantially more cost effective, is higher yielding with better purity than that from AGN, and often appears as longer needle-shaped crystals which have approximately twice the length to diameter (L/D) ratio of those produced by using AGN [11]. Several different methods for the synthesis of tetrazene from AGB have been developed. AGB is a water insoluble material, which can react with nitric acid in aqueous solution to form an acidic aqueous solution of AGN. Tetrazene is prepared by reacting an acidic aqueous solution of AGN with aqueous sodium nitrate(III) (NaNO_2) solution in the presence of acetic acid (CH_3COOH). However, this chemical reaction process has a potential runaway hazard at higher temperatures. Therefore, the reaction temperature should be maintained below 10 °C [12]. A better alternative method for preparing tetrazene from AGB is commonly used at present. Firstly, AGB and acetic acid are added to water to form an acidic aqueous solution of AGB,

and then solid sodium nitrite is added to the mixed solution to obtain precipitated tetrazene [13]. This chemical reaction can be processed at room temperature, which is less dangerous, but the yield of tetrazene is less than that from the previously mentioned reaction.

Lao and Sheng [14] have reported the influence of several factors on the synthesis of tetrazene from AGB; these include the molar AGB/ NaNO_2 ratio, the pH value of the AGB solution, the temperature for reaction between AGB and NaNO_2 , and the reaction time. The molar ratio of AGB/ NaNO_2 can affect the reaction rate and the yield of tetrazene. The most favorable molar ratio of AGB to NaNO_2 is 1.0:1.2. The pH value of the AGB solution mainly affects the yield of tetrazene. Tetrazene can only be obtained under neutral or weakly acidic conditions. The pH value of the AGB solution is best in the range of 4.0-6.0. The reaction temperature mainly affects the reaction rate. However, when the reaction temperature is higher than 60 °C, the yield of tetrazene is lowered due to the poor thermal stability of tetrazene. In addition, the reaction time is influenced by the reaction temperature. Our research group [15] has previously studied this subject, and has concluded that several experimental parameters contribute to increasing the yield of tetrazene as follows:

- a molar ratio of AGB to NaNO_2 of 1.0:1.2,
- a pH value of the AGB solution of 4.5-5.5,
- a temperature for reaction between AGB and NaNO_2 of 35-55 °C, and
- a reaction time of 1-6 h.

The maximum yield of tetrazene can reach 70% in the manufacturing process. It is indeed a challenge to further improve the yield of tetrazene in the manufacturing process.

The present study mainly explored the optimal synthesis parameters to further improve the yield of tetrazene. It was synthesized by reacting AGB with NaNO_2 in the presence of acetic acid. Taguchi's experimental design method was used to design the experimental conditions. Three control factors were selected, including the pH value of the AGB solution, the reaction temperature and the reaction time, each at three levels. These three factors were assigned to the $L_9(3^3)$ orthogonal array containing nine experimental sets of conditions. The experimental results were used to analyze and evaluate the experimental conditions of the optimal parameter combination for the maximum yield of tetrazene. The synthesized tetrazene was identified and characterized by EA, NMR, FTIR, OM, PSDA, XRD and DSC, and its impact sensitivity was determined using a BAM Fallhammer apparatus and the Bruceton analysis method.

2 Experimental

2.1 Materials

The standard sample of tetrazene was obtained from the National Chung Shan Institute of Science and Technology (NCSIST) in Taiwan. AGB, NaNO₂ and acetic acid were purchased from Uni-onward Co. All chemicals were of reagent grade and were used without further purification. All solutions were prepared using deionized water.

2.2 Design of the experiment conditions

The Taguchi method is one of the most widely used design methods [16, 17], and uses an orthogonal array from the experimental design to study more variables in a minimal number of experiments. An L₉(3³) orthogonal array with three control factors and three levels has been referred to in the literature [18-20], and was used to design the experimental conditions for this study. Three control factors were selected, the pH value of the AGB solution, the reaction temperature and the reaction time, each at three levels, as shown in Table 1. The above three factors were assigned to the L₉(3³) orthogonal array containing nine experimental formulas as shown in Table 2. Furthermore, the molar ratio of AGB to NaNO₂ was fixed at 1.0:1.2 and acetic acid was used to adjust the pH value of the AGB solution.

Table 1. The control factors and levels of the Taguchi experiments

Control factor	Level		
	1	2	3
A. pH value of AGB solution	4.5	5	5.5
B. Reaction temperature [°C]	35	45	55
C. Reaction time [h]	1	3	6

Table 2. The L₉(3³) orthogonal array of the Taguchi experiments

Exp. No.	pH value of AGB solution	Reaction temperature [°C]	Reaction time [h]
A1	4.5	35	1
A2	4.5	45	3
A3	4.5	55	6
A4	5.0	35	3
A5	5.0	45	6
A6	5.0	55	1
A7	5.5	35	6
A8	5.5	45	1
A9	5.5	55	3

2.3 Apparatus and procedures for the synthesis of tetrazene

AGB (13.6 g, 0.1 mol) and deionized water (200 cm³) were mixed in a double-jacketed glass vessel of about 500 cm³ capacity, which was stirred and maintained at 70 °C by a circulating water bath within an accuracy of ±0.3 °C. A 0.1 N acetic acid solution (200 cm³) was then titrated into the mixture until the pH value reached a set value (4.5, 5.0 or 5.5). This solution was filtered and cooled to room temperature. A NaNO₂ solution was prepared by dissolving NaNO₂ (8.3 g, 0.12 mol) in deionized water (200 cm³) and was poured into another double-jacketed glass vessel of about 500 cm³ capacity, which was stirred and maintained at a set temperature (35, 45 or 55 °C) by a circulating water bath. Subsequently, the NaNO₂ solution was pumped into the double-jacketed glass vessel containing the AGB solution by a peristaltic pump at a constant rate of 20 cm³/min, the reaction mixture was continuously stirred and maintained at a set temperature (35, 45 or 55 °C) for a set time (1, 3 or 6 h). Finally, the precipitated tetrazene was collected by filtration, washed several times with deionized water, dried at 50 °C for 24 h in a vacuum oven and stored in a sealed glass container to keep out moisture. A schematic diagram of the experimental procedure is shown in Figure 1.

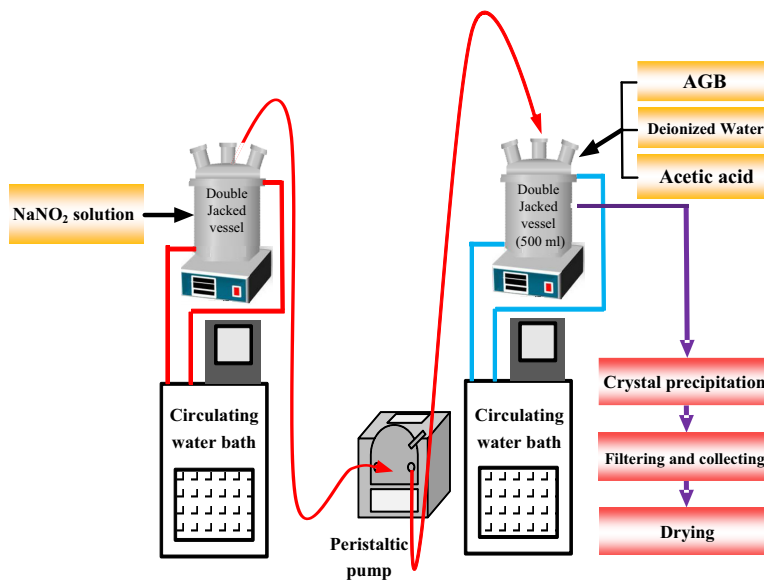


Figure 1 Schematic diagram of the experimental procedure

The synthesized samples were identified and characterized by means of:

- elemental analysis (EA, Elementar vario EL cube),

- nuclear magnetic resonance spectroscopy (NMR, Bruker AVIII 500 MHz),
- Fourier transform infrared spectroscopy (FTIR, PerkinElmer Spectrum 100),
- optical microscopy (OM, Leica DM750),
- particle size distribution analysis (PSDA, Mastersizer 2000),
- X-ray diffraction (XRD, Siemens D5000 & ENRAF-NONIUS 852DX60D), and
- differential scanning calorimetry (DSC, Netzsch Q20).

DSC measurements were used to study the thermal properties of the synthesized product. The sample was placed in a ceramic crucible and the experiments were carried out under a nitrogen atmosphere at a heating rate of $5\text{ }^{\circ}\text{C}\cdot\text{min}^{-1}$ using a sample weight in the range of 0.5-0.7 mg.

2.4 Impact sensitivity test

The impact sensitivity of the synthesized tetrazene crystals was determined with a BAM Fallhammer apparatus (Reichel & Partner, Germany). The test sample was taken with a scoop of 40 mm^3 . The sample was placed in a sample cell consisting of two co-axial steel cylinders, which were already in the locating ring on the intermediate anvil. The upper steel cylinder was set to be 1 mm above the lower cylinder and was held in that position by means of an O ring. When the sample cell was properly positioned, the dropweight was released and the impact result was observed. The Bruceton method [21] was used to evaluate the impact sensitivity of the tetrazene samples, which was based on a statistical analysis by determining the drop height (H_{50}) at which there was 50% probability of obtaining an ignition. Each sample was tested utilizing a 0.375 kg dropweight for 30 times to obtain the H_{50} . The impact energy (E_{50}) was calculated using the Equation 1

$$E_{50} = m \cdot g \cdot H_{50} \quad (1)$$

where m is the dropweight mass [kg], g is acceleration due to gravity [$\text{m}\cdot\text{s}^{-2}$], and H_{50} is the drop height [m].

3 Results and Discussion

3.1 Analysis and verification of the Taguchi experiments

A series of synthesis experiments for tetrazene formation with different operating parameters, such as pH value of the AGB solution, the reaction temperature and the reaction time were carried out, and then the effects of the operating

parameters on the yield of tetrazene were examined. The experimental results of Taguchi's orthogonal array are given in Table 3.

Table 3. Experimental results of Taguchi's orthogonal array

Exp. No.	Yield of tetrazene [%]	Average L/D ratio (-)	H_{50} [mm]	E_{50} [J]
A1	53.4 ±0.5	12.7	18.1	0.067
A2	66.5 ±0.4	7.1	23.6	0.087
A3	63.8 ±0.7	10.0	20.0	0.074
A4	63.8 ±0.4	3.9	25.0	0.092
A5	81.9 ±0.5	3.7	26.3	0.097
A6	73.7 ±0.3	7.6	22.5	0.083
A7	75.7 ±0.7	2.1	27.5	0.101
A8	42.7 ±0.5	7.7	20.8	0.076
A9	47.5 ±0.2	2.2	27.0	0.099

Each experiment was run at least three times and the averaged values were obtained with a relative standard deviation of less than 1.2%. The experimental results obtained were transformed into a signal-to-noise (S/N) ratio using the Taguchi method. The S/N ratio is used as a measure of the effect of noise factors on the target characteristics. In general, there are three categories of the performance characteristics in the study of the S/N ratio: larger-the-better, smaller-the-better and nominal-the-best. The yield of tetrazene was chosen as the quality characteristic in this study. The analysis of yield used larger-the-better type, a larger value represented by a higher yield, as well as a better quality characteristic. Table 4 presents the range and contribution rank of each factor for the yield of tetrazene. In the table, the target values for different levels of each factor are the arithmetic average of the target values corresponding to each level. The range is the difference between maximum and minimum of the target values on each factor and the rank represents the order of effect of each factor on the quality characteristic.

Table 4. Range and contribution rank of each factor for the yield of tetrazene

Level	Control factors		
	pH value of AGB solution	Reaction temperature [°C]	Reaction time [h]
1	61.23%	64.30%	56.60%
2	73.13%	63.70%	59.27%
3	55.30%	61.67%	73.80%
Range	17.83%	2.63%	17.20%
Rank	1	3	2

The trend of parameter influence for the three factors is shown in Figure 2. In the figure, the abscissa and ordinate represent the levels of factors and yield of tetrazene, and all points of corresponding levels for each factor are connected into a polyline, thus the change trend of the objective value for each factor with the increase of level can be obtained. Therefore, levels A2, B1 and C3 have the largest value of yield for the factors pH value of AGB solution, reaction temperature and reaction time, respectively. Based on above study, the maximum value of yield may be found under the conditions that the pH value of the AGB solution is 5.0, the reaction temperature is 35 °C and the reaction time is 6 h. It was also observed that the order of the effect of each factor on this quality characteristic is: pH value of AGB solution > reaction time > reaction temperature. The pH value of the AGB solution (factor A) has the greatest influence.

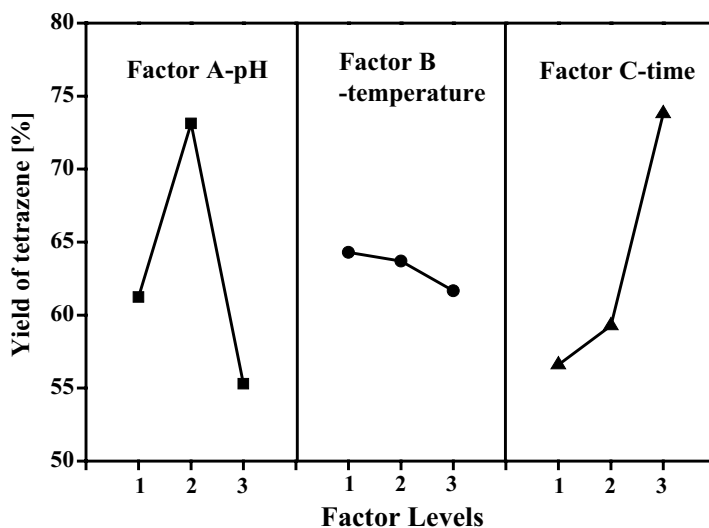


Figure 2. The trend of parameter influence for the three factors on the yield of tetrazene

Additional confirmatory experiments were performed to verify the effectiveness of the optimal parameters identified with the Taguchi method. The optimal design factor was the A2B1C3 parameter combination for the yield of tetrazene. Table 5 gives the results of the confirmatory experiment. This experiment was run five times under the same test conditions and the averaged value was obtained with a relative standard deviation of less than 1.2%. The experimental results indicated that the A2B1C3 parameter

combination produces a larger yield of tetrazene than the other combinations tested. The Taguchi method was successful in predicting this optimal parameter combination in order to obtain the maximum yield. In this way, the maximum yield of tetrazene can reach 83.7%.

Table 5. Results of confirmatory experiments

Parameter	Value
Optimal parameter combinations	A2B1C3
Factor A. pH value of AGB solution	5.0
Factor B. Reaction temperature [°C]	35
Factor C. Reaction time [h]	6
Yield of tetrazene [%]	83.7 ± 1.0
Average <i>L/D</i> ratio (-)	3.2
H_{50} [mm]	26.6
E_{50} [J]	0.098

3.2 Identification of the synthesized products

The elemental composition of the synthesized product was analyzed by EA. The analysis result showed that the synthesized tetrazene contained 12.74 wt.% carbon (C), 4.30 wt.% hydrogen (H), 74.30 wt.% nitrogen (N) and 8.66 wt.% oxygen (O), which are very close to the theoretical values of tetrazene composition (12.77 wt.% C, 4.29 wt.% H, 74.44 wt.% N and 8.50 wt.% O). Of these, the hydrogen and oxygen contents are slightly higher in the synthesized product, possibly due to the presence of moisture from the atmosphere.

The structure of the synthesized product was confirmed by NMR and FTIR. ^{13}C NMR spectrum shows only one signal at 154.5 ppm, which corresponds to the carbon atom in the CN_3 fragments. The FTIR spectrum exhibited characteristic peaks at 3350 cm^{-1} (broad, strong), 3000 cm^{-1} (broad, strong), 1710 cm^{-1} (sharp, strong), 1630 cm^{-1} (sharp, strong), 1490 cm^{-1} (sharp, strong) and 1420 cm^{-1} (sharp, strong), similar to that reported by Bird and Power [22].

The morphology and average length to diameter (*L/D*) ratio of the synthesized tetrazene crystals were observed and analyzed by optical microscopy (OM) and particle size distribution analysis (PSDA). Figure 3 shows the OM images of tetrazene crystals obtained from Exp. Nos. A1 and A7. From Figure 3(a), it can be seen that the synthesized tetrazene of Exp. No. A1 had a needle-shaped morphology with a maximum average *L/D* ratio of about 12.7, but its yield was only about 53.4%. On the other hand, Figure 3(b) illustrates that the synthesized tetrazene of Exp. No. A7 had a smaller particle size and its average *L/D* ratio was only about 2.1; however, its yield can reach

about 75.7%. The average L/D ratios of the synthesized tetrazene crystals for Exp. Nos. A1-A9 are summarized in Table 3. These values are in the range of 2.1-12.7. Through comparison, it seems that there is no direct relation between the average L/D ratio and the yield.

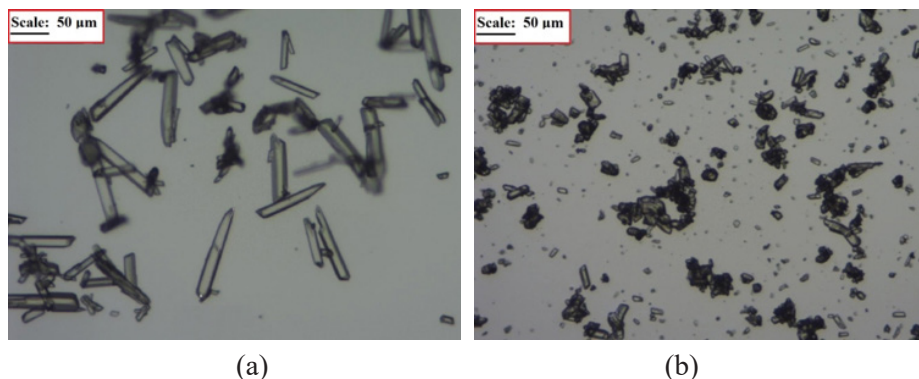


Figure 3. OM image of synthesized tetrazene crystals: (a) Exp. No. A1 (average $L/D = 12.7$), (b) Exp. No. A7 (average $L/D = 2.1$)

Figure 4 shows the XRD patterns of the standard sample and the synthesized samples of Exp. Nos. A1 and A7. It is observed that these three XRD patterns reveal similar peak positions; only relatively slight differences could be seen, which can be ascribed to the effect of a few impurities. This confirms that the synthesized samples were tetrazene crystals. Furthermore, the DSC thermograms of the standard and the synthesized samples of Exp. Nos. A1 and A7 are shown in Figure 5. These three curves show a similar exothermic peak at about 140 °C, which is consistent with the thermal decomposition temperature of tetrazene [3, 4]. Under the operating conditions of the optimal parameter combination (A2B1C3) for the yield, the average L/D ratio of the synthesized tetrazene crystals was 3.2, as shown in Figure 6 and Table 5.

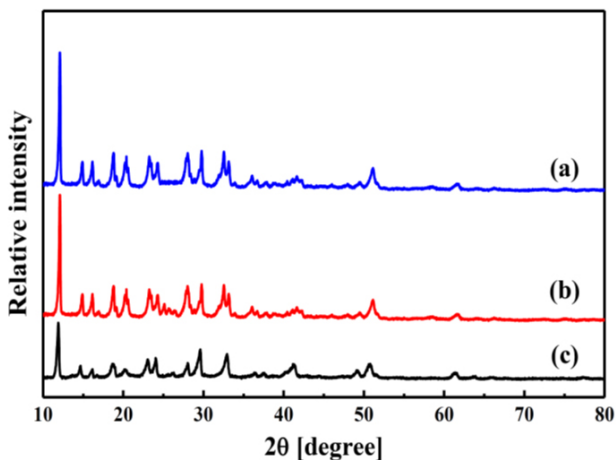


Figure 4. XRD pattern of tetrazene: (a) synthesized sample from Exp. No. A1, (b) synthesized sample from Exp. No. A7, (c) standard sample

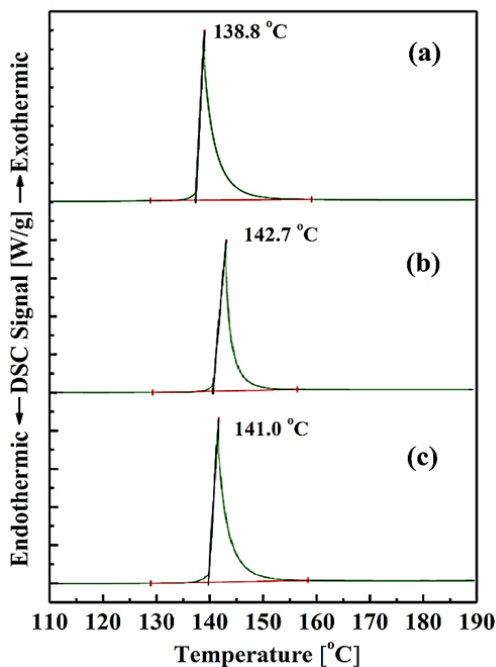


Figure 5. DSC thermograms of tetrazene: (a) synthesized sample from Exp. No. A1, (b) synthesized sample from Exp. No. A7, (c) standard sample

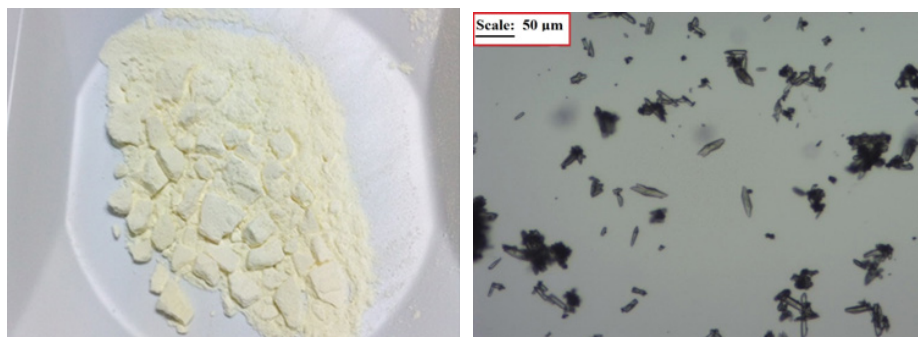


Figure 6. The synthesized tetrazene crystals under the operating conditions of the A2B1C3 parameter combination

3.3 Measurement of impact sensitivity

The impact sensitivities of the synthesized tetrazene crystals were determined using a BAM Fallhammer apparatus and the Bruceton up-and-down method. The experimental and analytical results are also presented in Table 3. The H_{50} and E_{50} are in the ranges of 18.1–27.5 mm and 0.067–0.101 J, respectively. Figure 7 shows the relation between impact sensitivity (impact energy) and average L/D ratio for all of the synthesized tetrazene crystals from Exp. Nos. A1–A9. It was found that the values of H_{50} and E_{50} decreased with increasing average L/D ratio of the tetrazene crystals. In other words, the tetrazene crystals with a larger average L/D ratio are more sensitive than those with a smaller average L/D ratio. Hot spot theory [23] can be employed to explain the above experimental results. In the impact sensitivity tests, while the external force acts on the tetrazene crystals with a larger average L/D ratio, the released heat will dissipate slower and the force acting on unit area of crystal surface becomes higher due to the smaller contacting area among these tetrazene crystals with a larger average L/D ratio. Therefore, “hot spots” are easy to form to enable detonation. Besides, “hot spots” are more likely to be formed at coarser surfaces because of their larger friction coefficient [24]. Under the operating conditions of the optimal parameter combination (A2B1C3) for tetrazene yield, the H_{50} and E_{50} values of synthesized tetrazene crystals were 2.66 mm and 0.098 J, respectively, as presented in Table 5, which has been verified to satisfy the requirements of the detonator functioning test. These operating conditions have also been verified in the actual manufacturing process.

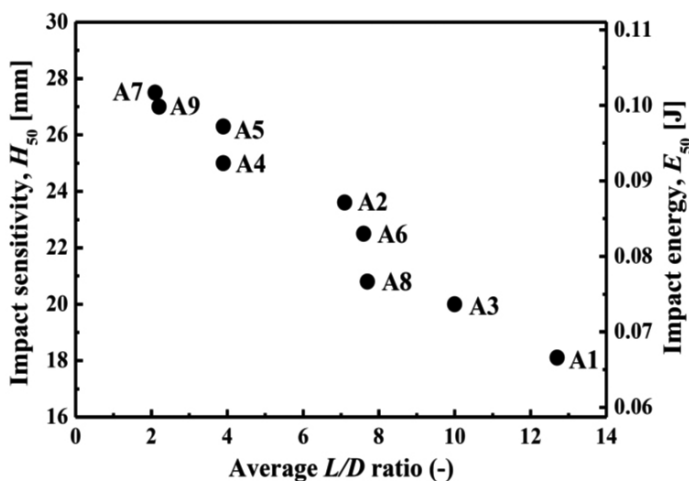


Figure 7. The relation between H_{50} , E_{50} and average L/D ratio

4 Conclusions

This study successfully used Taguchi's experimental design method to obtain the experimental conditions of the optimal parameter combination, in order to further improve the yield of tetrazene. Under the conditions that the molar ratio of AGB to NaNO_2 was fixed at 1.0:1.2, the optimal synthesis parameters were as follows:

- pH value of the AGB solution was 5.0,
- the reaction temperature was 35 °C, and
- the reaction time was 6 h,

then the maximum yield of tetrazene could reach 83.7%. The average length to diameter (L/D) ratios of the synthesized tetrazene crystals were in the range of 2.1-12.7. Through comparison, it seemed that there is not a direct relation between the average L/D ratio and the yield.

Furthermore, the impact sensitivities of the synthesized tetrazene crystals were measured and analyzed using a BAM Fallhammer apparatus and the Bruceton analysis method. The impact sensitivity (H_{50}) and impact energy (E_{50}) were in the ranges of 18.1-27.5 mm and 0.067-0.101 J, respectively. It was found that the values of H_{50} and E_{50} decreased with increasing average L/D ratio of the tetrazene crystals. In other words, tetrazene crystals with a larger average L/D ratio were more sensitive than those with a smaller average L/D ratio. Under the operating conditions of the optimal parameter combination (A2B1C3)

for tetrazene yield, the H_{50} and E_{50} values of the synthesized tetrazene crystals were 26.6 mm and 0.098 J, respectively, which has been verified to satisfy the requirements of the detonator functioning test. In addition, these operating conditions have also been verified in the actual manufacturing process.

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