

***N*-Guanylurea dinitramide: Optimal synthesis process, basic properties and spectroscopic data**

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ABSTRACT

N-Guanylurea dinitramide (GUDN), also known as FOX-12, is indeed a fascinating organic dinitramide salt with high insensitivity and good thermal stability. It shows great promise for use in next-generation insensitive munitions. In this study, GUDN was synthesized through the nitration reaction of ammonium sulfamate salt with an acid mixture (HNO₃/H₂SO₄) and guanylurea salts in water. The optimal conditions for this synthesis have been determined, including reaction temperature at -40 °C, the nitration reaction time of 45 minutes, and specific molar ratios of the components, which include guanylurea sulfate, ammonium sulfamate, and the mixture of sulfuric and nitric acid. Additionally, various properties and spectral data of GUDN, such as FTIR, ¹H-NMR, ¹³C-NMR, DSC-TG, XRD, and UV-Vis, have been thoroughly studied in the referenced article.

Keywords: *N*-Guanylurea dinitramide; Ammonium sulfamate; GUDN; FOX-12; Nitration reaction.

1. INTRODUCTION

It's fascinating to see the advancements in developing high-energy, safe, and environmentally friendly propellants and explosives within the energy materials industry. Dinitramide salts, such as ammonium dinitramide (ADN), potassium dinitramide (KDN), and GUDN, are indeed gaining attention as energetic materials for various applications like propellants, pyrotechnics, and explosives. The versatility of these salts with different cations opens up possibilities for innovative uses in military and space applications [1]. The focus on creating new high-energy materials like dinitramide salts showcases a promising direction for the industry.

ADN is indeed a promising oxidant in solid propellant formulations due to its environmentally friendly nature, high specific impulse, and minimal signature exhaust gases. It is positioned as a potential replacement for ammonium perchlorate. On the other hand, GUDN, a nitrogen-rich dinitramide salt, offers low susceptibility to impact and friction, making it a suitable choice for insensitive munitions. Its thermal stability is comparable to RDX and superior to that of ADN [2]. These characteristics highlight the potential of both ADN and GUDN in various applications within the field of propellants and munitions.

GUDN is indeed a versatile compound with various applications in propellant and explosive formulations. Its properties, such as thermal stability and low water solubility [3], make it a valuable component in these formulations, such as LOVA propellants, melt-cast, and PBX high explosive formulations [4, 5]. The combination of GUDN with other

compounds like ADN can lead to the development of innovative oxidizers for solid composite propellants [6]. Additionally, GUDN plays a crucial role as a precursor for the direct synthesis of ADN or through KDN [7, 8]. Its significance in these processes highlights its importance in the field of propellants and explosives. Moreover, GUDN is indeed a key ingredient in insensitive warhead fillings used in various types of ammunition like bombs, artillery, tanks, and mortars [9].

The synthesis process of GUDN involves a series of carefully controlled steps. First, ammonium sulfamate is nitrated using a mixture of nitric acid and sulfuric acid at a specific temperature range of $-50\text{ }^{\circ}\text{C}$ to $-20\text{ }^{\circ}\text{C}$. This mixture then reacts with guanylurea salts in water. The optimal conditions for this synthesis have been extensively studied, including factors like reaction temperature, component ratios, nitration reaction time, and the use of ion exchange agents.

In this paper, we described the synthesis of GUDN and analyzed the fundamental properties of this compound through various modern techniques like FTIR, $^1\text{H-NMR}$, $^{13}\text{C-NMR}$, DSC-TG, XRD, and UV-Vis.

2. EXPERIMENTAL SECTION

2.1. Materials

The chemicals used include sulfamic acid, ammonia solution, ethanol, sulfuric acid, fuming nitric acid, dicyandiamide, 2-propanol, and acetone. All chemicals used were of AR grade from Sigma Aldrich.

2.2. Equipment and analytical methods

2.2.1. IKA's reactor system

IKA's reactor system consists of a double-walled reactor vessel made from SUS316 stainless steel and connected to a Julabo deep cooling system for temperature regulation, with controlled stirring speed.

2.2.2. UV-Vis spectroscopy method

The UV-Vis spectroscopy method was used to determine the characteristic wavelengths of the GUDN product. The GUDN synthesis product was dissolved in water at a concentration of 10 mg/l and measured on a UV-Vis spectrophotometer Perkin Elmer Lambda 365.

2.2.3. FTIR spectroscopy method

The FTIR spectroscopy method was used to determine the characteristic functional groups of the synthesized products. They were measured on a Perkin Elmer FT-IR Spectrum Two.

2.2.4. NMR spectroscopy method

The NMR spectroscopy method was used to determine the structure of the synthesized compounds. They were measured on Bruker Avance 600 MHz spectrometers. ^1H NMR spectra: δ (H) are given in ppm relative to tetramethylsilane (TMS), using δ (DMSO- d_6) = 2.5 ppm as the internal reference. ^{13}C NMR spectra: δ (C) are given in ppm relative to tetramethylsilane (TMS), using δ (DMSO- d_6) = 39.52 ppm as the internal reference.

2.2.5. Thermogravimetry-differential scanning calorimetry (TG-DSC)

Thermogravimetry-differential scanning calorimetry (TG-DSC) was used to determine

the melting temperature, decomposition, and reduction in mass of the product. DSC-TG was carried out using the Labsys Evo system by SETARAM (France).

2.3. Preparation of precursors for GUDN synthesis

Synthesis of ammonium sulfamate (AS). Sulfamic acid (97.1 g, 1 mol) was suspended in 50 mL of water. Ammonia solution 25% (82 g, 1.2 mol) was added slowly to the beaker containing sulfamic acid and the suspension until pH = 8 was reached. The solution is heated to 100 °C until the volume of the solution is reduced to one-third of its initial quantity. Subsequently, the solution is cooled in an ice bath. A white precipitate appeared and was filtered off with a Buchner funnel, washed with 2-propanol, and dried at 60 °C to obtain a white salt (AS). After drying, the salt was ground and crushed into a fine powder in a porcelain mortar. M.p:131 °C, yield: 95.5%.

Synthesis of Guanylurea sulfate ((GU)₂S). Dicyandiamide (42 g, 0.5 mol) and 50 mL H₂O were introduced into a 500 mL three-necked round-bottom flask equipped with a stirring apparatus and a heating arrangement. Aqueous sulfuric acid (24.5 g, 0.25 mol in 125 mL H₂O) was added dropwise to the flask containing dicyanamide using a dropping funnel. The solution was then heated to 60 °C and stirred for 4 hours. Then, the resulting solution was placed in an ice bath for 2 h and the thus-formed white crystals were filtered off. The remaining filtrate was concentrated in a hot water bath and cooled in ice. The resulting crystals were filtered, washed with 2-propanol, and dried in an oven at 60 °C [10]. Mp: 196 °C, yield: 94.6%.

Synthesis of Guanylurea nitrate (GUN). Dicyandiamide (42 g, 0.5 mol) and 50 mL H₂O were introduced into a 500 mL three-necked round-bottom flask equipped with a stirring apparatus and a heating arrangement. Nitric acid 65% (40.95 g, 1 mol) was added dropwise to the flask containing dicyanamide using a dropping funnel. The solution was stirred for 30 min at room temperature and boiled for 5 min. Then, slow cooling to room temperature resulted first in cloudiness and finally in the formation of an insoluble powder that was filtered, washed with acetone, and dried in an oven at 60 °C [10]. Mp: 203 °C, yield: 96.7%.

2.4. Synthesis of guanylurea dinitramide (GUDN)

Fuming nitric acid (140 mL, 3.29 mol) and sulfuric acid 98% (56 mL, 1.03 mol) were placed into the IKA reactor system reactor vessel. As soon as the reaction vessel temperature reached -40 °C, ammonium sulfamate (57.17 g, 0.5 mol) was placed slowly into the acid mixture for 15-30 minutes. After adding the ammonium sulfamate, stir the mixture vigorously for 45 minutes at a temperature of -40 ± 2 °C. The mixture's viscosity increased as the reaction occurred, and a white precipitate appeared.

Ice cold water (100 mL) was given in an open glass bowl fitted with a mechanical stirrer. Guanylurea sulfate (54.36 g, 0.18 mol) was suspended in ice-cold water (100 mL) and the temperature was maintained at 10 ± 5 °C. The nitration reaction mixture was then poured into an open glass bowl containing a suspension of (GU)₂S under constant stirring (400 ± 50 rpm). The reaction was robust and the color of the mixture turned into a specific yellow-green color. Then, the mixture was cooled at a temperature of 15 °C for 30 min. The formed white precipitate was then filtered off with a Buchner funnel, and washed with cold water. The resulting solid was recrystallized in acetone and was dried at 50 °C to constant weight. Mp: 213 °C, yield: 53.4%.

3. RESULTS AND DISCUSSION

3.1. Effect of nitration reaction temperature and nitration reaction time on GUDN synthesis yield

The initial investigation involved developing the nitration reaction between ammonium sulfamate (AS) and the acid mixture ($\text{HNO}_3/\text{H}_2\text{SO}_4$) at the mass ratio of 1:5.5 as a model reaction. Firstly, the influence of temperature and reaction time should also be seriously taken into consideration for the yield of the nitration reaction. The obtained results are shown in figure 1. It was observed that both temperature and reaction time exhibited a significant effect on the reaction yield.

Based on the data provided (figure 1), it appears that temperature plays a significant role in the efficiency of the reaction. When the temperature decreases from $-20\text{ }^\circ\text{C}$ to $-40\text{ }^\circ\text{C}$, the efficiency of the reaction increases. However, going below $-40\text{ }^\circ\text{C}$ leads to a decline in the reaction yield. Additionally, at a constant temperature of $-30\text{ }^\circ\text{C}$, the efficiency initially increases with reaction time but then decreases after 30 minutes. This peculiar pattern can be elucidated by the exothermic nature of the nitration reaction. Reducing the temperature of the reaction slows down the decomposition of dinitramic acid, thereby elevating the reaction's efficiency. On the contrary, at lower reaction temperatures, the nitration reaction proceeds at a more sedate pace. Consequently, prolonging the reaction duration boosts efficiency until the nitration reaction is fully completed. After this point, the exothermic decomposition of dinitramic acid becomes increasingly conspicuous.

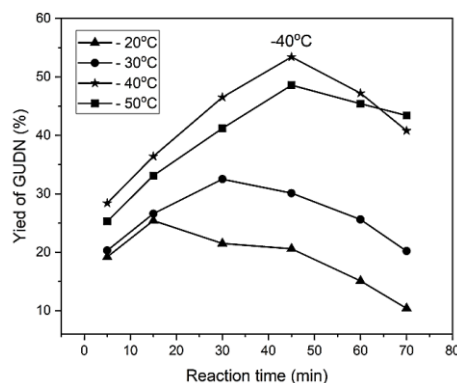


Figure 1. The influence of the reaction temperature and reaction time on GUDN yield.

It observed that the nitration process reaches completion at a temperature of $-40\text{ }^\circ\text{C}$ after stirring the reaction mixture for 45 minutes. Any extension in the reaction time to 60 or 70 minutes results in a reduction of efficiency due to the intensification of dinitramic acid decomposition. This happens because the nitration process has already reached its equilibrium. On the other hand, the nitration process intensifies at $-20\text{ }^\circ\text{C}$, resulting in a surge in the reaction temperature. This promotes a further decomposition of the dinitramic acid. According to the academic article, dinitramic acid undergoes rapid decomposition at temperatures exceeding $-20\text{ }^\circ\text{C}$ [11]. Therefore, the highest yield at $-20\text{ }^\circ\text{C}$ was achieved within 20 min of the reaction time. At this point, a balance is formed between nitrification and dinitramic acid decomposition.

On the contrary, the substance exhibits stability even at $-50\text{ }^\circ\text{C}$. However, the yield

was significantly reduced because the cold temperature resulted in a high viscosity of the reaction mixture. This makes it difficult to dissipate the heat generated during the reaction [12], leading to local overheating of the reaction mixture and rapid decomposition of the resulting products. Therefore, the ideal temperature and time for the nitration reaction are $-40\text{ }^{\circ}\text{C}$ and 45 minutes, respectively.

3.2. Study on the effect of the ratio of the acid mixture and ammonium sulfamate on the synthesis efficiency of GUDN

Furthermore, the effect of the molar ratio of the acid mixture and ammonium sulfamate was also studied. It was observed that the use of an acid mixture ($\text{HNO}_3/\text{H}_2\text{SO}_4$) as the primary nitration agent in the GUDN production process using ammonium sulfamate as a starting material is important. Therefore, the mass ratio between ammonium sulfamate and the acid mixture significantly affects the efficiency of the reaction and the efficiency of the overall synthesis. An experimental investigation was carried out to evaluate this effect, including ammonium sulfamate nitration reactions at $-40\text{ }^{\circ}\text{C}$, maintained for 45 min. This experiment adjusted the mass ratio of the guanyurea sulfate salt to the acid mixture. In this particular scenario, the molar ratio of HNO_3 and H_2SO_4 was kept constant at 3.2:1, a value taken from previous academic research [13]. The results demonstrated the impact of different component ratios on the nitration reaction, which can be seen in figure 2.

The information presented in figure 2 indicates that increasing the mass ratio of the acid mixture and ammonium sulfamate mixture from 3 to 5.5 leads to a significant improvement in reaction efficiency from 26.6% to 53.4%. However, efficiency is still not affected much when the volume of the acid mixture is increased. This implies that excess amounts of the acid mixture do not have a significant impact on effectiveness. However, when the quantity of the acid mixture decreased to 45%, the yield plummeted by 50%. The nitration mechanism depends on the formation of nitronium ions (NO_2^+). Therefore, the acid mixture ratio plays an important role in determining the number of nitronium ions produced. Therefore, a significant decrease in the volume of the acid mixture will result in a decrease in the number of nitronium ions. This leads to incomplete nitration reactions and thus reduces reaction efficiency.

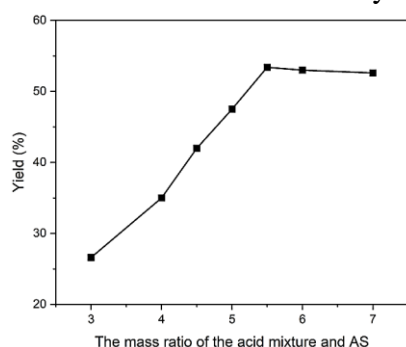


Figure 2. The effect of the acid mixture on GUDN yield.

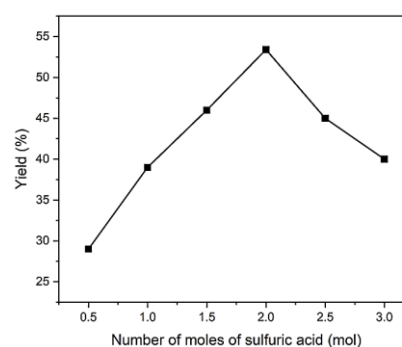


Figure 3. The effect of sulfuric acid quantity on GUDN yield.

Finally, to evaluate the effect of the acid ratio on the efficiency of the nitration reaction, the molar ratio of ammonium sulfamate and nitric acid was kept constant at 1:6.58, while the amount of sulfuric acid was varied in the mixture. The specific results of this effect are

shown in figure 3. Notably, the maximum efficiency observed with one mole of sulfuric acid is 2.06 mol. When the amount of sulfuric acid decreases, the activity of the nitrate mixture also decreases, leading to reduced nitration efficiency. On the other hand, excess sulfuric acid will negatively impact the nitration process because it can increase the viscosity of the mixture at low temperatures. This, in turn, leads to an increase in the local temperature of the reaction mixture. This escalation has caused the product's decomposition to occur rapidly. Therefore, the optimum molar ratio of ammonium sulfamate to sulfuric acid and nitric acid is 1:2.06:6.58.

3.3. Study on the effect of guanylurea salts on the synthesis efficiency of GUDN

The GUDN compound is produced through an ion exchange reaction which occurs between a nitration reaction product mixture (HDN) and an aqueous suspension of guanylurea salt. The quantity of the final product obtained from the nitration reaction was fine-tuned according to the conditions mentioned above. To enhance the efficiency of the GUDN synthesis, an excess of guanylurea salt over the HDN yield during the nitration reaction is required. Therefore, the effect of the amount of guanylurea salt on the efficiency of the GUDN was carefully studied. In particular, the nitration reaction was performed at - 40 °C for 45 minutes, preserving a fixed amount of ammonium sulfamate while only changing the quantity of guanylurea salt ((GU)₂S, GUN) introduced into the exchange reaction. The findings regarding the influence of salt intake on GUDN performance are illustrated in figure 4.

The research indicated that increasing the molar ratio of guanylurea to ammonium sulfamate to more than 0.65 (as shown in figure 4) did not significantly affect the reaction yield. As such, the optimal molar ratio for GUDN generation was determined to be 0.65. Moreover, data from figure 4 indicate that the use of (GU)₂S or GUN did not notably influence the total efficiency of GUDN. Nevertheless, the utilization of (GU)₂S as an ion exchange agent seems to be more advantageous due to its simpler separation process from GUDN compared to GUN. Besides, during the ADN synthesis from GUDN, sulfate salts are commonly employed as ion exchangers. Hence, the application of guanylurea sulfate salt benefits GUDN synthesis by preventing nitrate ion pollution in synthetic ADN and facilitating the recovery of (GU)₂S.

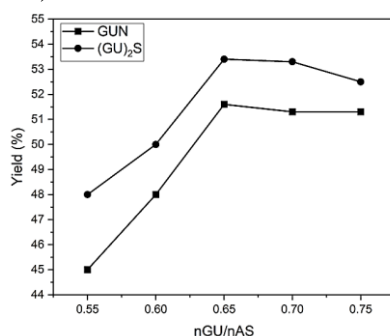


Figure 4. The effect of guanylurea salts on the GUDN yield.

*nGU: number of moles of guanylurea group in (GU)₂S or GUN, nAS: number of moles of ammonium sulfamate.

3.4. Basic properties and spectroscopic data of GUDN

The synthesis product (GUDN) is a white crystal with a melting point of 213 °C (DSC)

and a density of 1.755 g/cm³. GUDN exhibits good solubility in dimethyl sulfoxide (DMSO) and *N*-methyl-2-pyrrolidone (MNP) but has limited solubility in water. The synthesized products were dried and measured by FTIR, UV-Vis, ¹H-NMR, ¹³C-NMR spectra, XRD pattern and DSC-TG.

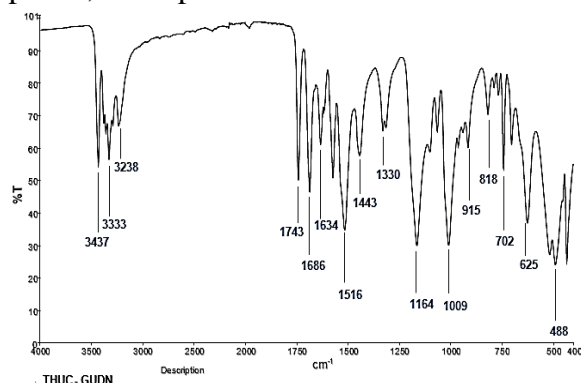


Figure 5. FTIR spectrum of GUDN.

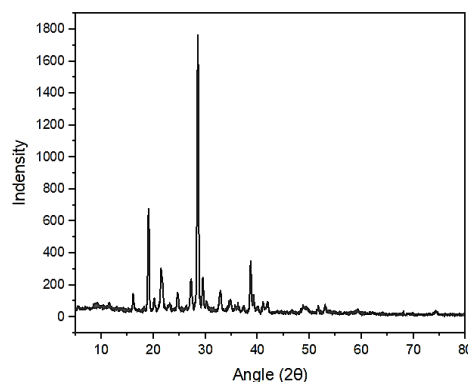


Figure 6. XRD pattern of GUDN.

FTIR spectroscopy of GUDN was carried out to further characterize the GUDN structure. FTIR spectra showed characteristic bands of the –NH₂, –NO₂, –C=O, N₃ functional groups with wavenumber ranges in the 3238-3437 cm⁻¹, 1164-1634 cm⁻¹, 1686-1742 cm⁻¹, 915-1009 cm⁻¹, respectively. Additionally, the XRD pattern of GUDN has characteristic peaks at angles 2θ of 16.115, 18.085, 21.485, 24.635, 27.275, 28.565, 28.525, and 38.675.

The ¹H-NMR spectrum of GUDN displayed a single at δ 7.12 ppm (2H) corresponding to two protons of a group of –NH₂ linked to C2 and a single at δ 8.02 ppm (4H) corresponding to four protons of two groups of –NH₂ linked to C1 (figure 7). The important signal showed at δ 9.58 ppm, which was attributed to –NH group. Furthermore, ¹³C-NMR spectrum showed distinct signals at δ 154.32 ppm and δ 155.38 ppm, corresponding respectively to C1 and C2 carbon atoms (figure 8).

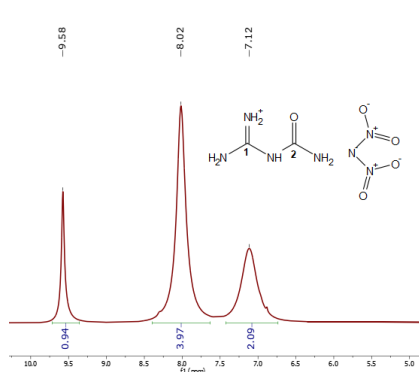


Figure 7. ¹H-NMR spectrum of GUDN.

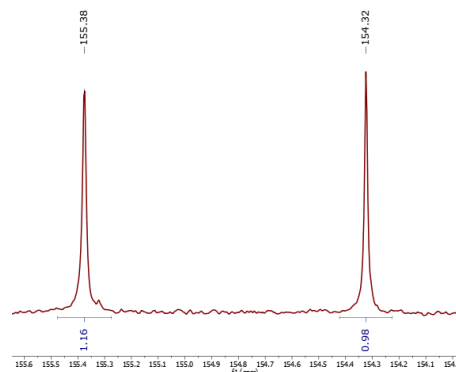


Figure 8. ¹³C-NMR spectrum of GUDN.

From the DSC-TG data of GUDN, it is observed that the melting and decomposition temperature of the product is at 213 °C, and the mass loss of GUDN is 73% at a heating rate of 10 °C/minute.

The UV-Vis spectroscopy of the synthesized product has two peak positions at wavelengths of 213 nm and 285 nm (figure 9). The absorbance at 285 nm is characteristic

of the $(-N(NO_2)_2)^-$ ion caused by low energy $n-\pi^*$ transitions, whereas the absorption maximum at 213 nm is attributed to high energy $\sigma-\sigma^*$ transition. These peaks prove that the product contains dinitramide ions.

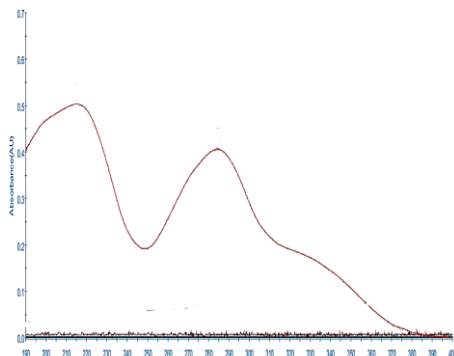


Figure 9. UV-Vis spectrum of GUDN.

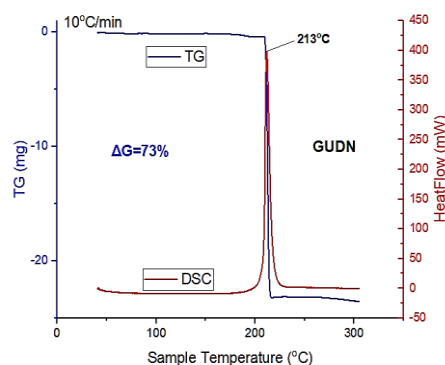


Figure 10. DSC-TG of GUDN.

4. CONCLUSIONS

In summary, GUDN has been successfully synthesized with a notable overall yield of 53.4%, and its identity was confirmed through spectral analyses, including FTIR, $^1\text{H-NMR}$, $^{13}\text{C-NMR}$, UV-Vis, XRD, and DSC-TG data. The white-synthesized GUDN was comprehensively characterized and exhibits a melting and decomposition temperature of 213 °C, along with a density of 1.755 g/cm³. Additionally, the influences of nitration reaction temperature, reaction time, and the ratio of the acid mixture to ammonium sulfamate and guanylurea salts were meticulously assessed to determine optimal conditions. Specifically, the nitration reaction lasts for 45 minutes at a reaction temperature of -40 °C and the ideal molar ratio of $(\text{GU})_2\text{S}$, AS, H_2SO_4 and HNO_3 is 0.325, 1, 2.06, and 6.58, respectively.

REFERENCES

- [1]. Ang How Ghee, G. Santhosh, "Advances in Energetic Dinitramides – An Emerging Class of Inorganic Oxidizers," World Scientific, Singapore, (2008).
- [2]. Bottaro, J. C., "Recent advances in explosives and solid propellants," Chemistry and industry, No.7, pp. 249-252, (1996).
- [3]. Shiota, K., Izato, Y., Miyake, A. "Analysis of guanylurea dinitramide pyrolysis using DSC/Raman spectroscopy". Journal of Analytical and Applied Pyrolysis, Vol.156, 105113, (2021).
- [4]. Östmark, H., Bemm, U., Bergman, H., & Langlet, A, "N-guanylurea-dinitramide: a new energetic material with low sensitivity for propellants and explosives applications," Thermochemica Acta, Vol.384, No.(1-2), pp. 253-259, (2002).
- [5]. Li, C., Ma, F., Sun, J., Sui, H., Yu, Q., Zhao, L., & Yin, Y. "Thermal hazard evaluation of N-guanylurea dinitramide (GUDN) by using kinetic-based simulation approach". Journal of Thermal Analysis and Calorimetry, Vol.141, pp. 905-913, (2020).
- [6]. LIJun-qiang, P. Q., WANGKe, X. Q., & XUHui-xiang, F. Z. (2019), "Effect of ADN/GUDN dual oxidizers on the combustion features of nitrate ester plasticized polyether solid propellants," Chinese Journal of Energetic Materials, Vol.27, No.4, pp.297-303, (2019).
- [7]. Stefan, E. K., & Johansson, J, "U.S. Patent Application No. 17/276,513" (2022).
- [8]. Johansson, J., Ek, S., & Skarstind, M, "One-step synthesis of ADN from FOX-12," In 47th International Annual Conference of the Fraunhofer ICT. Karlsruhe, Germany, Vol. 33, (2016).

- [9]. Charles, D, "New low-sensitivity modular charge propellant based on GUDN," In Insensitive munitions and energetic materials technology symposium, Bristol, (2006).
- [10]. Klapötke, T. M., & Sabaté, C. M., "The preparation and characterization of guanylurea nitrate and perchlorate salts". *Heteroatom Chemistry: An International Journal of Main Group Elements*, Vol.19, No.3, pp. 301-306, (2008).
- [11]. Kumar, Pratim, "An overview on properties, thermal decomposition, and combustion behavior of ADN and ADN based solid propellants," *Defence technology*, Vol. 14, No. 6, pp. 661-673, (2018).
- [12]. Nazeri, Gh H., Nia M. Fayaz, And Ghobadi P. Key, "Synthesis of ammonium dinitramide by nitration of potassium and ammonium sulfamate. The effect of sulfamate counterion on ADN purity," *Iran.J.Chem.Chem. Eng*, Vol.27, No.1, pp. 85-89, (2008).
- [13]. Mandal, A. K., Kunjir, G. M., Singh, J., Adhav, S. S., Singh, S. K., Pandey, R. K., "Optimization of ammonium sulfamate nitration for the preparation of ammonium dinitramide," *Central European Journal of Energetic Materials*, Vol.11, No.1, pp. 83-97, (2014).

TÓM TẮT

N-Guanylurea dinitramide: Tối ưu hoá quá trình tổng hợp, đặc trưng phổ và tính chất cơ bản

N-Guanylurea dinitramide (GUDN), còn được gọi là FOX-12, là muối dinitramide hữu cơ hấp dẫn với độ nhạy thấp, độ ổn định nhiệt tốt. Nó hứa hẹn là một lựa chọn xuất sắc cho việc sử dụng trong các loại đạn không nhạy thế hệ tiếp theo. Trong nghiên cứu này, quá trình tổng hợp GUDN bao gồm phản ứng nitrat hóa của muối amoni sulfamat với hỗn hợp axit và muối guanylurea trong nước. Điều kiện tối ưu cho quá trình tổng hợp này đã được xác định, bao gồm nhiệt độ phản ứng ở -40 °C, thời gian phản ứng nitrat hóa là 45 phút, và tỷ lệ mol cụ thể của các thành phần tham gia gồm guanylurea sulfate, amoni sulfamat và hỗn hợp axit sulfuric với axit nitric. Ngoài ra, các tính chất và dữ liệu phổ của GUDN, như FTIR, ¹H-NMR, ¹³C-NMR, XRD, DSC-TG và UV-Vis đã được nghiên cứu kỹ trong bài báo.

Từ khóa: N-Guanylurea dinitramide; Ammonium sulfamate; GUDN; FOX-12; Phản ứng nitro hóa.