Influence of Nano-Reduced Graphene Oxide on the Thermal Characteristics of 1,3,5-Trinitro-1,3,5-Triazinane (RDX)

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Abstract: It is well known that the improvement in the energetic materials field needs to rally the efficiency by increasing the performance of the materials. Optimizing the sensitivity and the performance is considered a challenge that obstacle the developer to getting the required high-energy materials. Nanoparticles with reduced graphene oxide (RGO) rely on a new series of energy materials that can be produced chemically or thermally to reduce the oxygen content by exploiting the interlaying spacing and functionalization of the graphite basal planes. The RGO could easily disperse in an organic solvent due to its hydrophobic material and achieve a large surface area. In this paper, The production of RGO was discussed, and the solvent-nonsolvent method was applied to disperse the RGO on 1,3,5-trinitro-1,3,5-triazinane (RDX). Scanning electron microscopy (SEM) was used to check the morphology of the tested composition. The deflagration experiment was applied to determine the ignition temperature and delay. The thermal behavior was determined by TGA and DSC techniques. The results showed that the presence of RGO layers accumulated the heat release inside the crystals during the heating process and decreased the ignition temperature of the prepared composite RDX-RGO. The decomposition peak of RDX-RGO decreased dramatically compared with the individual RDX. The RGO acts as a catalyst and increases the reactivity of RDX.

Keywords: reduced graphene oxide; RDX; scanning electron microscopy; thermal study.

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1. Introduction

There is no argument that increasing the performance of high-energy materials and decreasing their sensitivity to different stimuli is the objective of all the advanced research [1]. Recently several researchers have studied energetic systems based on carbon nanomaterials [2]. The combustion characteristics of different metalized propellants have been enhanced using carbon nanomaterials [3,4]. On the other side, interesting researchers have studied the influence of nano-carbons on the sensitivity characteristics of primary explosives. Yan et al. studied the influence of the carbon hybrid system on the ignition characteristics of lead azide [5]. While the preparation of carbon-based copper azide has been discussed, the characteristics of copper azide in the presence of graphene and graphene oxides have been reported deeply in recent research [6-11]. In addition, the influences of graphene oxide on the characteristics of several secondary explosives have been reported recently. Liu et al. studied the ignition behavior of a composite based on graphene oxide/RDX [12]. Other composites' thermal stability and safety based on graphene oxide and energetic materials have been studied [13-
The influence of reduced graphene oxide (RGO) on the sensitivity and the thermal behavior of secondary explosives has not been studied yet in literature. The reduced graphene oxide (RGO) might preserve the high energy when used in energetic materials. It can be used in a very tiny amount, but its effect should be effective. RGO has a two-dimensional (planar) material that can be extracted by oxidizing and reducing the graphite powder [16]. RGO has already been used extensively in materials research due to its large theoretical specific surface area, which might affect the characteristics of the materials [17,18]. Preparation of graphene sheet [19] divided into a two-part method, the first one is up to down (chemical vapor deposition) (CVD) [20] and the second one is divided into two different processes (Mechanical – Chemical) [21]; the mechanical by (Cleavage of Graphite crystals) [sonication of exfoliation graphite] and [double sided adhesive] and the chemical by (oxidation process) and (graphite intercalation compounds) [22]. The oxidation process was done by 4 different methods 1- [Brodie (KClO3-KNO3)], 2-[Staudenmaier use (KClO3, NaClO3, H2SO4)], 3- [Hummer method by using (NaNO3, KMNO4, H2SO4)], 4- [Modified Hummer method by using (NaNO3, KMNO4, H2SO4)] [23]. Novel and simple methods for preparing RGO have been published recently in literature [24-26] which could encourage the application of RGO in the field of energetic materials.

In the area of materials for energy use, there is a general rule that advanced explosives perform high performance with high sensitivities [27-31]. The challenge in the explosive field is to obtain modern techniques to preserve the high performance of explosives and ensure safety while storage and processing [32-34]. The explosive sensitivities might be modified by regulation about the forms and proportions of crystals [35,36]. However, these techniques are broad; they are nevertheless uncontrollable and not monetary. A well interesting widespread used explosive is 1,3,5-trinitro-1,3,5-triazinane (RDX). It is an essential explosive in energy materials that have many uses [30, 31]. RDX is recorded as relatively sensitive and cannot be pressed as a pure explosive. Traditionally, RDX is pressed in the form of plastic bonded explosive to reduce the sensitivity [37] but also the performance is reduced [38] due to the addition of inert materials (4-20 wt%). The influence of nano-Aluminum on the sensitivity of waxed RDX was studied by Dong et al. [39] and proved the enhancement of the sensitivity concerning the pure RDX. In this study, a modern carbon additive (RGO) was prepared, and its effect on the characteristics of the RDX explosive was discussed. A method for preparing RGO-RDX composite by 2 wt% of RGO is presented. The characterization of the prepared RDX composite was investigated by SEM. The thermal behavior of the studied composite and the pure RDX was examined. The effect of RGO on the ignition temperature and activation energy of RDX also were studied.

2. Materials and Methods

2.1. Materials.

Hydrochloric acid 5% (HCl), Phosphoric acid (H3PO4), and Sulfuric acid (H2SO4) was obtained from Aldrich. Potassium permanganate (KMnO4) and graphite were obtained from WINLAB, UK. Hydrogen Peroxide (H2O2) was obtained from Merck (Germany). Acetone was obtained from Alfa Company, India. Pure RDX crystals were purchased from Eurenco (France).
2.2. Preparation of Reduced Graphene Oxide (RGO).

The modified Hummers method has been used to prepare RGO. 100 ml of H₂SO₄ was poured on 70 ml of H₃PO₄ in a 400 ml beaker. Then, 3.5 g of graphite was added to the acids under the stirring in the presence of an ice bath. Once the beaker temperature reached 0°C, 22 g of KMnO₄ was added. After 35 min, the ice bath was removed and the beaker left at room temperature to warm up. The mixture was subjected to mechanical stirring for 1 hour afterward; the beaker was placed on a hotplate to keep its temperature at 65°C for 22 hours. Cold deionized water of 500 ml was added to the mixture. Then 35 ml of H₂O₂ was added, which turned the brownish color of the mixture into yellow. The mixture was then sonicated for 50 min. at 50 GHz. The content of the beaker was washed by deionized water and diluted HCL until its pH reached 6. The precipitation was dried for 14 hr under vacuum at 60°C.

2.3. Preparation of RDX/RGO composites.

The prepared RGO was used to prepare RDX/RGO composite. 50 ml of acetone was added to 15 mg of the prepared RGO in a 400 ml beaker. The mixture was sonicated for 1 hr. Then, 5 g of RDX has added to 150 ml of another acetone portion. The mixture was under continuous sonication for 100 min. at 45°C in 1L beaker. The suspension was added to anti-solvent (cold water). At last, the RDX/RGO composite was separated by a centrifuge and washed with distilled water. The prepared RDX/RGO was dried at 60°C for 14 hr. under a vacuum.

2.4. Morphology of the crystals.

Morphology of the crystals of RDX and the RDX/RGO crystals was studied using a scanning electron microscope (SEM, JOEL JSM-6010LA). Figure 1 presents the SEM micrograph for the prepared RGO-RDX composite.

2.5. Ignition temperature measurements.

The ignition temperature was determined by using the Chilworth deflagration test. Samples (0.1 g) were added in three test tubes and placed vertically into the heating block. The increase in the heating rate was 10 °C min⁻¹. The ignition temperature was reported at the moment of ignition of the samples. The mean value of three measurements was reported in Table 3. Developers can obtain the kinetic parameters by the ignition delay method analysis. The Arrhenius equation was used to plot a linear relationship to obtain the activation energies. Each sample with both time to ignition (τ) and 1000/temperature (T/K) is presented in Fig. 4. The kinetic parameters were determined and reported from the curve slope and table 3.

2.6. Thermal behavior of RDX/RGO composites.

The pure RDX and RDX/RGO composition were studied by thermogravimetric analysis (TGA55, TA instruments, USA, open high-temperature crucible) under 10 K min⁻¹ heating rate. The TG was tested at 40-350°C, and the mass of samples was 2.5 mg under a nitrogen atmosphere of 40 ml min⁻¹. The heat flow properties were also determined by the differential scanning calorimetry technique (Q20 by TA Instruments) with a closed pan. The sample mass was 2 mg under a heating rate of 10 K min⁻¹ and a temperature range of 40-350°C.
3. Results and Discussion

3.1. SEM characterization.

The morphology of the crystals of RDX-RGO and the individual RDX is presented in Figure 1. RDX-RGO crystals have nearly the same crystal size as the RDX crystals. This indicates that the coating of RDX by RGO does not influence the crystal size of RDX. The formulated RDX/RGO crystals have a nature that looks like that of individual RDX. This result confirms the neutral effect of RGO on the surface of the individual RDX. RDX crystals’ defects have disappeared in the presence of RGO; it seems that the RGO covers the RDX crystals and causes smooth crystal surfaces of RDX-RGO. This result might have an improvement in the sensitivity of the pure RDX. Also, a decrease in the sharp corners of the RDX crystals has been caused by the RGO; as a result, it may be another explanation for RDX sensitivity. It was concluded that the precipitation of RGO of the surface of RDX decreased the sharp edges of RDX crystals without influence on the crystal sizes. So it is predicted to decrease the sensitivity of the individual RDX.

3.2. DSC study.

The thermograms obtained by DSC for the thermal action of RDX coated by RGO2 % and pure RDX under 10°C min-1 heating rate are presented in (Fig.2 a) and (Fig.2b), respectively. At the same time, the DSC parameters for the studied RDX composite and the pure RDX are listed in Table 1. The DSC curve of pure RDX shows endothermic decomposition at a maximum peak temperature of 203.9 0C and a well-formed decomposition peak at 235.8 0C. In the case of the composite RDX with RGO, The exothermic decomposition peak has decreased due to the presence of RGO, which causes heat accumulation and shifts the decomposition peak. At the same time, a dramatic decrease in the endothermic peak of the RDX (melting peak) was observed. The maximum endothermic peak of RDX/RGO was 142 0C which has been decreased from 203.9 0C for pure RDX. The high heat conductivity of RGO resulted in the high accumulation of the heat and speed up the melting process of crystals of RDX. Also, it is obvious here that the presence of the RGO with the RDX reduces the...
composite’s decomposed heat compared to the individual RDX. This could be a result of the effect of RGO on the decomposition mechanism of the pure RDX. This result required more investigation to study the kinetics of the composite RDX/RGO to clarify the influence of RGO on the RDX crystals.

| Sample        | Heating rate °C/min | Endothermic Peak | Exothermic peak |
|---------------|---------------------|------------------|-----------------|
| RDX-RGO2%     | 10                  | 140.3/142.0      | 123             |
| RDX           | 10                  | 201.4/203.9      | 83              |

Table 1. DSC data of RDX and RDX-RGO 2% at 10°C/min heating rate.

Figure 2. Thermogram of DSC for the studied explosive samples.

3.3. Thermogravimetric analysis.

The weight loss of both the pure RDX and RDX/RGO was studied by thermogravimetric analysis (TGA) at a 10 °C min⁻¹ heating rate. The TG/DTG curves for RDX/RGO and the pure RDX are presented in Fig.3. at the same time, the TG/DTG data are reported in Table 2.

Table 2. TGA data of RDX and RDX-RGO 2% at 10°C/min heating rate.

| type          | # /°C/min⁻¹ | TG curve | DTG curve |
|---------------|-------------|----------|-----------|
| RDX-RGO2%     | 10          | 161.7    | -98.5     |
| RDX           | 10          | 188.4    | -99.8     |

The DTG thermogram clarifies the influence of the RGO on the thermal action of RDX. It is also obvious that RGO shifted the temperature of decomposition of RDX. The decomposition temperature shifted from 188.4°C to 161.7°C by an approximately 26.7 °C decrease in the decomposition temperature. Also, the shift in the maximum peak decomposition is 39.8 °C. Also, a sharp decomposition peak was observed in the case of RDX/RGO, which might be due to the heat accumulation due to the presence of the RGO in the studied composition, which leads to catalysis the decomposition compared with the pure RDX. It was published in ref. [40] that RGO has a quick exothermic decomposition peak due to oxygenic functional groups on its structure where the RGO absorbs heat to trigger the deoxygenation process.
Figure 3. TGA/DTG for a. pure RDX b. Composite RDX/RGO at the heating rate 10 °C min⁻¹.

3.4. Ignition temperature and ignition delay measurements.

For modeling and estimating safety, time-to-ignition measurement can sometimes be assumed. The moment the ignition is postponed, the sample generates energy that inevitably contributes to the sample getting thermally ignited. The kinetic parameters placement by the explosion delay process was stated as an efficient method of understanding the heat stability of explosives [40]. The method has also been applied to detect the influences of the materials added to the thermal action of explosives [41,42]. The prepared RGO-RDX was tested at several temperatures ranging from a temperature that exceeds the ignition temperature. The duration required to ignite the material was reported in Table 3. The data were plotted in a relationship presented in Fig.4. The activation energies were determined based on the slope of the curve and presented in Table 3. The determined activation energies of the individual RDX and RDX-RGO 2%wt were 45.2 and 88.4 kJmol⁻¹, respectively. The activation energy of RDX-RGO is more than double that of individual RDX. The RGO coating increased the ignition temperature of the individual RDX and increased its activation energy.

Table 3. Ignition time delay (τ) and the activation energy (Ea) of the tested materials.

| Sample      | Ignition temp . | τ/s at temperatures / °C | Ea/kJ mol⁻¹ | R²    |
|-------------|-----------------|---------------------------|-------------|-------|
|             | Tₐ/℃            | 250 260 270 280 290 300 310 320 |             |       |
| RDX         | 201.4           | 34 29 26 21 18 15 12 10     | 45.5        | 0.9890|
| RDX/RGO2%   | 193.2           | 32 25 18 12 9 6 4 -         | 88.4        | 0.9912|

Figure 4. For all the samples tested, a linear relation for both ln (τ) and 1000 T⁻¹.
4. Conclusions

By adding 2 wt% of RGO, a composite explosive based on RDX-RGO was successfully prepared. RDX-RGO crystals have the same shape and size as the pure crystals of RDX. For RDX-RGO, smooth crystal surfaces have been observed, and the cracks of the pure RDX crystals have been disappeared, which could decrease the RDX sensitivity to mechanical stimuli. The thermal reactivity of RDX-RGO is somewhat different from RDX; the decomposition peak of RDX-RGO is lower than pure RDX. Besides, a dramatic decrease in the melting temperature of RDX/RGO was observed due to the large heat accumulation in the presence of RGO, which caused a speed up of the melting process of the RDX crystals. Also, from the DTG results, it was observed that the presence of oxygenic functional groups on the RGO structure resulted in the absorption of the heat to trigger the deoxygenation process and caused the sharp decomposition of RDX/RGO. On the other hand, the activation energy of RDX-RGO is near twice the pure RDX activation energy. RDX-RGO is a promising composition and needs more investigation.

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Conflicts of Interest

The authors declare no conflict of interest.

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