

Preparation and Characterization of Cellulose/RDX Composite Aerogel Spheres

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Abstract: As a high-energy explosive, 1,3,5-Trinitroperhydro-1,3,5-triazine (RDX) has been widely used in military and civilian applications such as explosives, rocket fuels and gun propellants due to its good comprehensive properties. However, at present, the application of RDX has been encumbered by its high sensitivity and crystal defects. Herein, inspired by the unique structure and eco-friendliness of cellulose aerogel, for the first time, cellulose aerogel was introduced into RDX to prepare cellulose/RDX composite aerogel spheres by the solution impregnation method. The cellulose/RDX composite aerogel spheres were characterized by scanning electron microscopy (SEM), elemental analysis, thermogravimetric and differential scanning calorimetry (TG-DSC), X-ray diffractometer (XRD), and a vertical impact sensitivity tester. The material detonation speed

was calculated using EXPLO5. The results showed that the surface of cellulose/RDX composite aerogel spheres was smooth, and the internal three-dimensional cross-linked void of the cellulose aerogel spheres was well filled with RDX particles. Compared with RDX raw materials, the particle size of RDX in the cellulose/RDX composite aerogel spheres was greatly reduced. The N content in the cellulose/RDX composite aerogel spheres was as high as 32.24%. Thermal analysis results showed that the activation energy of cellulose/RDX composite aerogel spheres increased by 31.28 kJ/mol. The crystal structure of RDX in the cellulose/RDX composite aerogel spheres remained unchanged. Moreover, the impact sensitivity H50 of the cellulose/RDX composite aerogel spheres increased by more than 65 cm.

Keywords: Cellulose · RDX · Aerogel spheres · Thermal stability · Sensitivity

1 Introduction

RDX is a high-energy explosive with good comprehensive properties and most widely used in high-energy explosive at present, but there are various shortcomings in RDX raw materials, such as high impurity content, high sensitivity, crystal defects, and their internal quality (density, purity, shape, dispersion, etc.). Due to the defects of RDX raw materials, their application in weapons have been limited. They also cause potential hazards during storage and transportation. [1–7]. Compounding or nanocrystallization of RDX raw materials is an effective way to improve its performance [8–9]. Some alternative materials obtained from natural resources have recently garnered considerable attention to be combined with energetic materials, which is an innovative solution to make some unprecedented advances. Among the various eco-friendly materials reported to date, cellulose has been highlighted as a promising building block because of its natural abundance, easy modification, physicochemical robustness, biocompatibility, lightweight, and recyclability [10]. Enormous efforts have been undertaken to use cellulose derivatives as an alternative natural material for a wide variety of energetic materials [11].

In 2003, Tappan B C et al. [12] used the cellulose derivative NC as the skeleton to prepare CL-20/NC composite energetic materials. The composite energetic materials con-

tain CL-20 with a mass fraction of 50%. Compared with CL-20 raw materials, the particle size of CL-20 in the CL-20/NC composite energetic materials decreased greatly and the peak temperature of thermal decomposition also decreased. In 2014, Jin et al. [13] prepared NC/RDX nanocomposite energetic materials. The NC/RDX nanocomposite energetic materials contain RDX with a mass fraction of 30% and an average size below 100 nm. Compared with RDX raw materials, the thermal decomposition temperature decreased by 30 °C. In 2016, Wang et al. [14] used the cellulose derivative NC as the skeleton to prepare HMX/NC nanocomposite energetic materials. The mechanical strength of HMX/NC nanocomposite energetic materials has been improved greatly with a reduction of the thermal stability. Pivkina A, Spitzer D [15–16] and Song X L [17] also prepared ultrafine RDX particles by different methods. Results show that compared with RDX raw materials, after the

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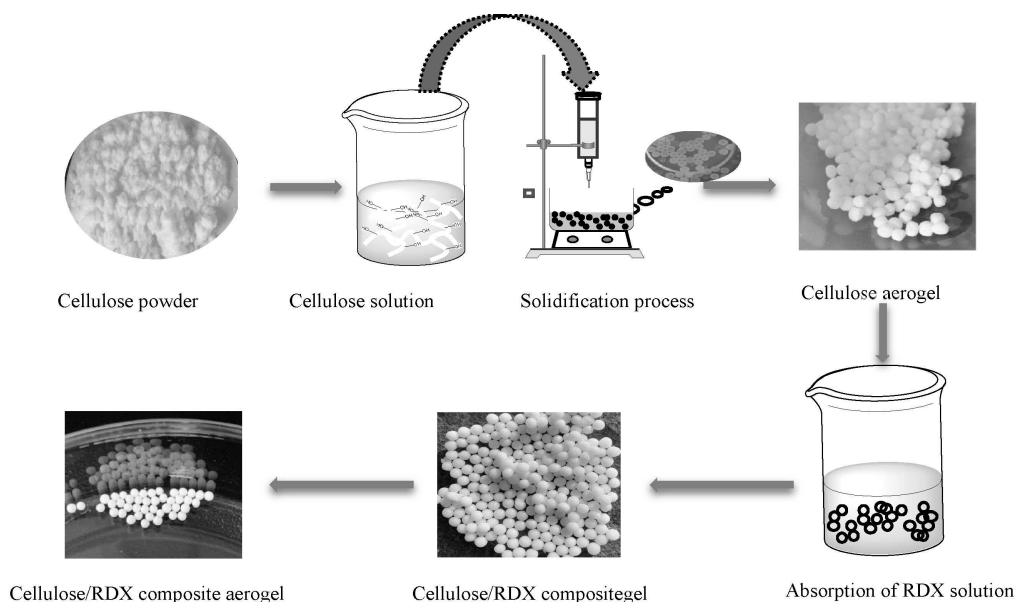


Figure 1. Experimental flow chart.

RDX particles nanocrystallization, the mechanical sensitivity decreases significantly, and the peak temperature of thermal decomposition decreases by more than 10 °C.

The above studies show that the RDX/cellulose derivative composites contain energetic materials with a mass fraction below 60%. The friction sensitivity of RDX cannot be improved effectively. The thermal decomposition stability reduced with the decreased particle size of RDX. To solve these problems, in this paper, cellulose aerogel spheres were used for the first time as a carrier for high-energy explosives and cellulose/RDX composite aerogel spheres were prepared by the solution impregnation method. Cellulose spheres show excellent three-dimensional cross-linking structure; more interestingly, RDX particles can be effectively protected within the three-dimensional cross-linking holes.

The experimental process is simple and easy to realize in industrial production. The cellulose/RDX composite aerogel spheres have the advantages of high explosive content, large output power, excellent thermal stability, good mechanical properties and low mechanical sensitivity. More interestingly, with the reduction of the particle size, the thermal stability increased.

2 Experimental Section

2.1 Materials

RDX raw materials were provided by Gansu Yinguang Chemical Industry Ltd. size: 150–450 μm. High purity cellulose powder was provided by North Century (Jiangsu) Cellulose Material Co., Ltd. Acetone and Trichloromethane were

purchased from Damao Chemical Reagent Factory; Acetic Acid, Tert-Butyl alcohol, Ethyl Acetate, NaOH and Urea were purchased from Beijing Tong Guang Fine Chemicals Company; Deionized water was self-made.

2.2 Experimental Process

Cellulose gel spheres were prepared by the droplet free drop method. After washing and drying, the spheres were placed in a supersaturated RDX solution in acetone. After completion of the impregnation absorption, the samples were dried again to obtain cellulose/RDX composite aerogel spheres. The process flow is shown in Figure 1.

2.3 Preparation of Cellulose/RDX Composite Aerogel Spheres

The preparation of cellulose/RDX composite aerogel spheres was carried out as follows: (1) 2 g cellulose powder was added into an aqueous NaOH/urea/water (7/12/79) solution and stored in the fridge at −20 °C. Then the cellulose solution with 2% cellulose content was obtained [18]. (2) 100 mL ethyl acetate, 100 mL trichloromethane and 30 mL acetic acid were mixed to obtain the acid coagulation [19]. The coagulation bath was placed on a blender, adjusting the stirring rate to 300 r/min. (3) The cellulose solution was transferred into the dispensing syringe with a 0.4 mm diameter needle. Under the influence of gravity, the cellulose solution dropped into the coagulation bath at a rate of 5 drops per second to form cellulose gel spheres. In order to achieve the uniformity of the cellulose spheres, the cellu-

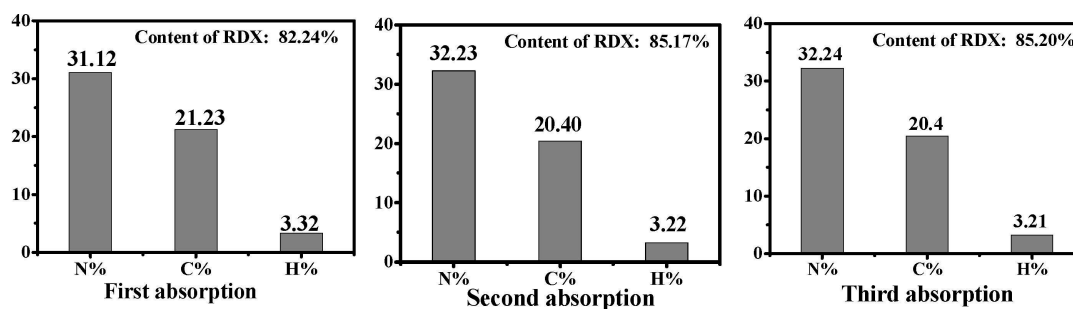


Figure 2. Histogram of nitrogen content of cellulose/RDX composite aerogel spheres.

lose solution was added to the dispensing syringe at the same speed as the dripping to make the droplets drip under the same pressure. (4) The cellulose gel spheres were washed with 50% and 75% concentrations of tert-Butanol, 4 hours each time, thus cellulose alcohol gel spheres were obtained. (5) Cellulose aerogel spheres were obtained by quickly freezing the spheres in liquid nitrogen and freeze-drying them, followed by immersion in a supersaturated acetone solution of RDX to achieve the absorption equilibrium. After the acetone was volatilized, the cellulose spheres were incubated again in the supersaturated acetone solution of RDX to achieve the absorption equilibrium. (6) Step 5 was repeated until the RDX in the cellulose aerogel spheres was saturated. Then the cellulose/RDX composite aerogel spheres were obtained by quick freezing in liquid nitrogen and freeze-drying of the cellulose composite spheres.

2.4 Characterization and Tests

The morphology of cellulose aerogel spheres, the internal structure of cellulose aerogel spheres, RDX particles, cellulose/RDX composite aerogel spheres and the internal structure of cellulose/RDX composite aerogel spheres were characterized by SEM. (S-4800; Hitachi; Japan). All samples were sputter coated with gold to provide adequate conductivity.

The shrinkage rate of the cellulose/RDX composite aerogel spheres after freeze drying was calculated according to the following formula (1).

$$S = (L_0 - L_C) / L_0 \times 100\% \quad (1)$$

Where S is the shrinkage, L_0 is the diameter before freeze drying and L_C is the diameter after freeze drying [20].

The thermal decomposition analysis of RDX raw materials and cellulose/RDX composite aerogel spheres was measured by using a thermogravimetric analyzer (STA 449 F3 Jupiter, Netzsch, Germany)

X-ray diffraction patterns of cellulose/RDX composite aerogel spheres, cellulose powders and RDX raw materials were determined by using an X-ray diffractometer (Bruker

D8 Discover, Bruker Co. Germany) equipped with Cu K α radiation in the 2 θ range of 5~60° in steps of 0.02°.

The impact sensitivity of the cellulose/RDX composite aerogel spheres, the RDX materials and the mechanical mixture of RDX and cellulose was tested by an HGZ-1 impact instrument. Test method: GJB772A-1997 standard method [21]; Mass of samples: 35 ± 1 mg, using 5.0 kg drop hammer; Temperature: room temperature; Representation of test results: H50 (critical drop height).

The detonation speed of cellulose/RDX composite aerogel spheres was calculated with the EXPLO5 software. Density: 90% of theoretical density and 95% of theoretical density.

3 Results and Discussion

3.1 Elemental Analysis

To analyze whether or not the cellulose aerogel spheres and RDX were combined, the nitrogen content of the cellulose/RDX composite aerogel spheres was analyzed. There is no nitrogen in pure cellulose spheres. After compounding cellulose with RDX, the nitrogen content test results of the composite aerogel are shown in Figure 2.

After the impregnation with RDX in a supersaturated acetone solution for three times, the nitrogen content of the cellulose aerogel spheres almost reached the saturation equilibrium state. When the cellulose aerogel spheres were impregnated with RDX supersaturated acetone solution for the first time, the nitrogen content was 31.12%, the RDX content was calculated as 82.24%. To optimize the RDX content, the cellulose/RDX composite aerogel spheres were impregnated to the RDX supersaturated acetone solution for a second time. The result shows that the nitrogen content can reach up to 32.23%, and the content of RDX is 85.17%. The nitrogen content in three times impregnated cellulose aerogel spheres can reach 32.24%. Compared with the two-times-impregnated sample, the difference in nitrogen content is small, which indicates that the content of RDX in the cellulose aerogel spheres reached a maximum value. The RDX content in the cellulose/RDX composite aer-

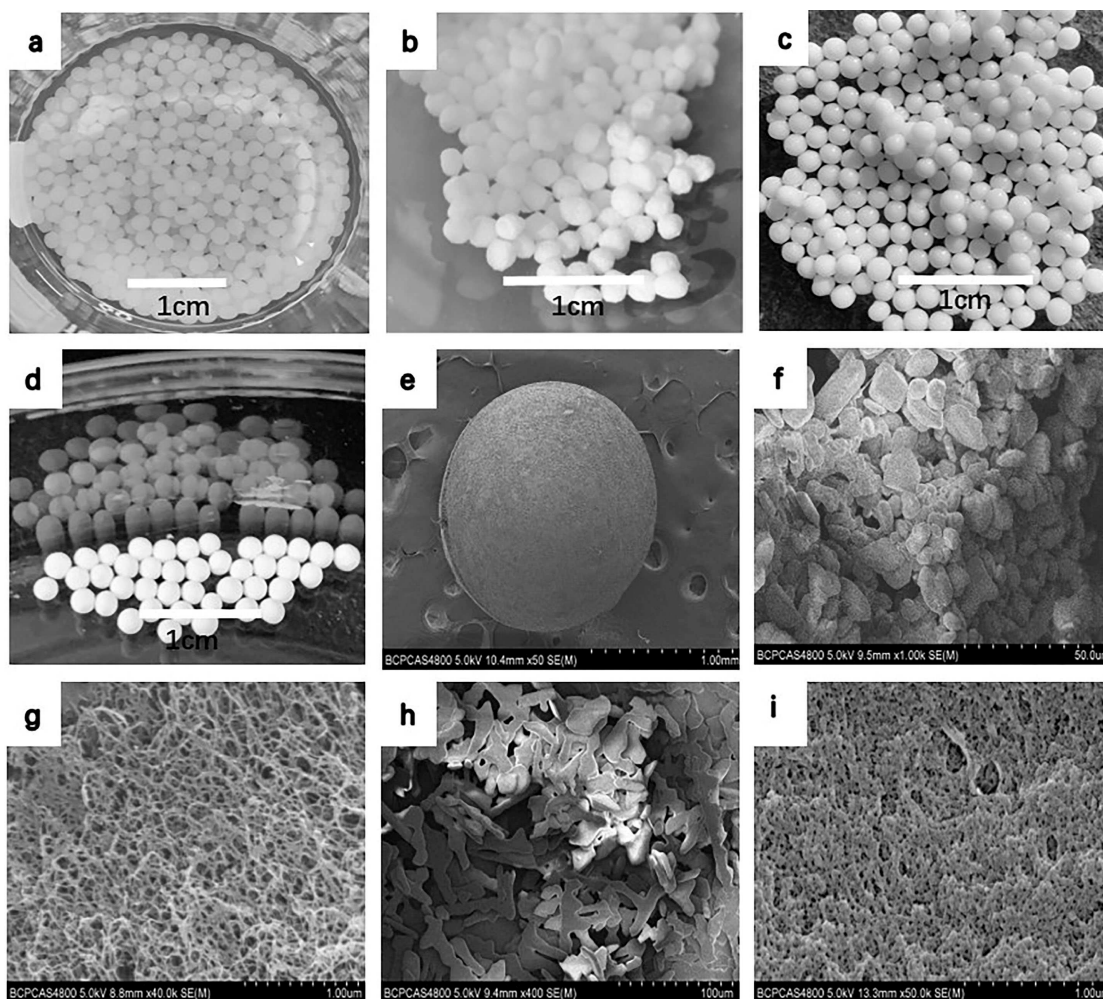


Figure 3. Appearance and SEM image of cellulose/RDX composite aerogel.

ogel spheres is as high as 85.2% based on the calculation, which is much higher than the content of high-energy explosives in aerogel materials prepared by co-solution of cellulose derivative and energetic materials.

3.2 SEM Analysis

In order to observe the morphology of the cellulose aerogel spheres, the cellulose/RDX composite aerogel spheres and RDX in the composite aerogel spheres more directly, the surface of the cellulose aerogel spheres, the cellulose/RDX composite aerogel spheres, and the interior were characterized by SEM. The characterization results are shown in Figure 3

It is shown in Figure 3-a that uniform spheres with regular shape are formed by cellulose solution drops in a coagulation bath under the same pressure. After the cellulose spheres were washed and dried, the cellulose aerogel spheres were obtained and maintained a good spherical

shape (Figure 3-b). Figure 3-c is the image of cellulose/RDX composite gel spheres; the surface of the composite gel spheres is smooth. Compared with the cellulose gel spheres, the size did not change significantly, but the color changed to opaque white. It can be seen from Figure 3-d and Figure 3-e that cellulose/RDX composite aerogel spheres maintain a good spherical shape. Since the interior of the cellulose aerogel spheres is filled with RDX particles, cellulose/RDX composite aerogel spheres become much denser. Compared with pure cellulose aerogel spheres, the compressive strength of cellulose/RDX composite aerogel spheres increased significantly. Figure 3-f shows the non-uniform size and irregular shape of RDX particles, which were obtained by dissolving RDX materials in acetone and re-precipitating them. The diameters of the particles range from 10 μm to 40 μm . Figure 3-g shows the internal image of the cellulose aerogel spheres, which shows a three-dimensional cross-linked network structure. Figure 3-h shows the internal image of the cellulose/RDX composite aerogel spheres. As shown in Figure 3-h, the internal three-dimen-

sional network structure is maintained, since the interior of the cellulose aerogel spheres is filled with RDX particles. Compared with the cellulose fibers in the pure cellulose spheres, the diameter of the fibers in the cellulose/RDX composite aerogel spheres is much bigger, which can be due to a large amount of RDX particles being attached to the surface of the cellulose fibers, thereby linking multiple cellulose fibers together and wrapping the cellulose fibers inside, thus covering the original cross-linked structure and forming a new cross-linked structure. The magnified SEM images in Figure 3-i indicate that a large amount of RDX particles are deposited on the surface of the cellulose cross-linked network structure, and filled in the three-dimensional intersection network pores. In addition, the cellulose aerogel skeleton also helps to maintain the small diameter of RDX, because the fine pore structure of aerogels can prevent particles from overgrowth and reuniting.

3.3 Shrinkage Analysis

The shrinkage rate of the cellulose aerogel spheres and cellulose/RDX composite aerogel spheres was analyzed using the shrinkage calculation formula. 50–100 gel spheres were selected before drying, to measure their diameter and to calculate the average value L0. The selected samples were freeze dried separately. After drying, the diameter was measured again, and the average value LC was calculated. L0 and LC were substituted into formula (1). The results are shown in table 1.

Table 1. Shrinkage rate of cellulose aerogel spheres and cellulose /RDX composite aerogel spheres.

samples	Cellulose aerogel spheres/mm	Cellulose /RDX composite aerogel spheres/mm
Before drying	1.519	1.537
After drying	1.495	1.535
Shrinkage rate	1.58%	0.13%

As can be seen from Table 1, the diameter of the cellulose aerogel sphere decreases, and the shrinkage rate is 1.58%. Because the cellulose aerogel sphere has a rich cross-linked pore inside, there are many internal voids. After being dried, the internal cross-linked pores have a certain degree of collapse. The diameter of the cellulose/RDX composite aerogel sphere remains almost unchanged after being dried, and the shrinkage rate is only 0.13%. This is because the cross-linked pores inside the cellulose/RDX composite aerogel sphere are filled with RDX particles, the internal voids are reduced, and the RDX particles filled in

the cross-linked pores prevent the collapse of the cross-linked pores.

3.4 DSC Analysis

The thermal properties of RDX raw materials and cellulose/RDX composite aerogel spheres were characterized by DSC-TG at heating rates of 5, 10, 20 Kmin⁻¹ respectively. The results are shown in Figure 4 and table 2.

Table 2. Thermal decomposition temperatures of cellulose/RDX composite aerogel spheres and RDX materials at different heat rates.

Sample	$\beta/(^{\circ}\text{C min}^{-1})$		
	5	10	20
RDX raw materials	234.95	239.77	246.11
Cellulose/RDX composite aerogel sphere	233.50	238.17	243.47

Table 2 demonstrates that relative to the RDX raw materials, the thermal decomposition peak temperature of cellulose/RDX composite aerogel spheres did not decrease significantly at different heating rates, which is almost the same as that of RDX raw materials. This indicates that after combining RDX with cellulose aerogel spheres, the particle size decreased greatly, but the thermal decomposition temperature remained almost unchanged. This is due to the good thermal insulation properties of cellulose. Another reason is that heat transfer can be blocked by the complex internal cross-linked structure. We can see from Figure 4-a, b, and c that there is exotherm after the exothermic peak temperature of RDX, which is the heat release caused by the decomposition of cellulose decomposition since the exothermic temperature of cellulose is higher than RDX.

Research shows that: $\ln\left(\frac{\beta}{T_{pi}^2}\right)$ is a linear function of $(1/T_{pi})$. By analyzing the thermal decomposition peak temperature of cellulose/RDX composite aerogel spheres and RDX raw materials at different heating rates, the Kissinger formula (formula 2) [22] to fit the straight line is applied to calculate the apparent activation energy (Ea) and the pre-factor(A) of the cellulose /RDX composite aerogel spheres and RDX raw materials (see Figure 4-e, f). The results are shown in Table 3.

Table 3. Thermal decomposition properties of cellulose/RDX composite aerogel spheres and RDX materials.

samples	Ea(kJ/mol)	A(min ⁻¹)	Te([°] C)	Tb([°] C)
RDX raw materials	262.51	6.15*1026	229.03	230.72
Cellulose/RDX composite aerogel spheres	293.79	1.36*1030	227.48	229.15

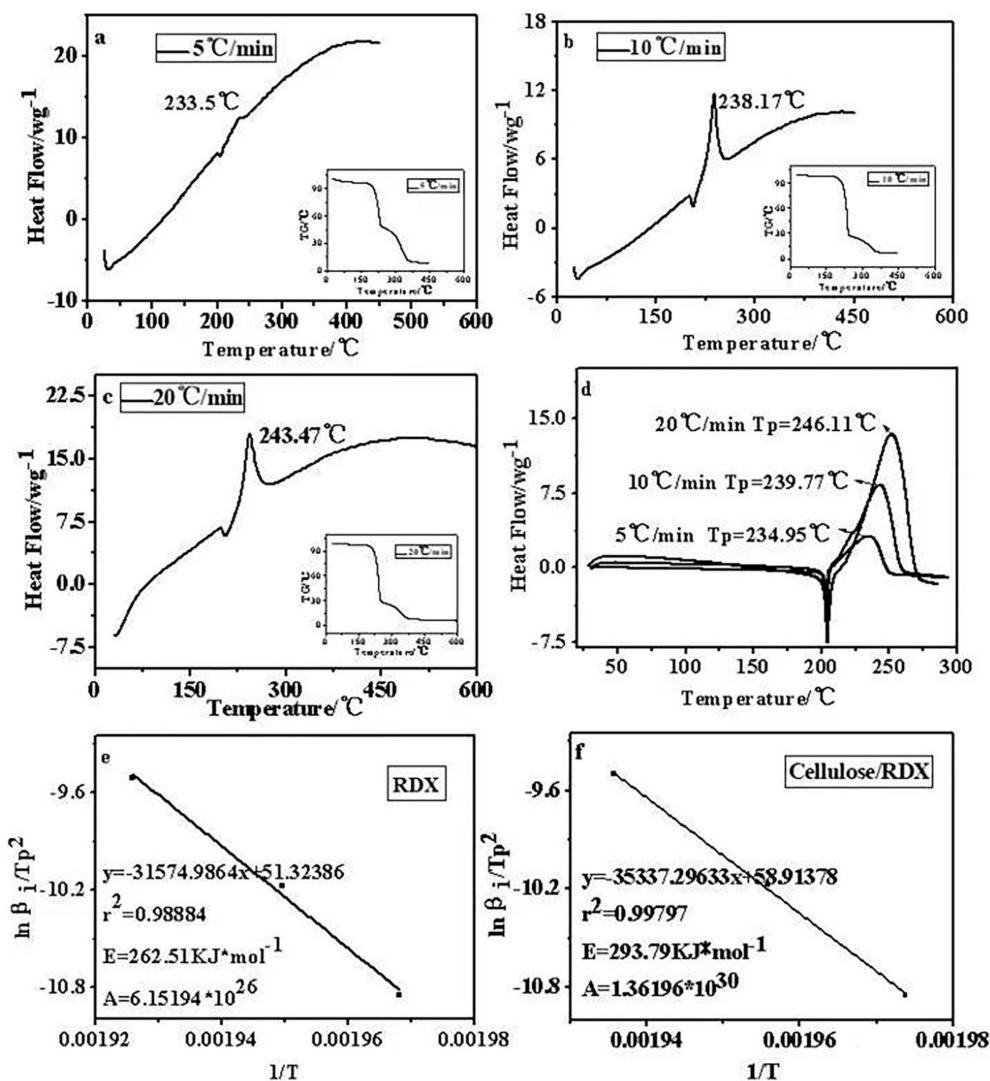


Figure 4. DSC-TG diagram of cellulose/RDX composite aerogel and RDX material at different heating rates and fitting line of activation energy calculated by Kissinger method.

$$\ln \frac{\beta_i}{T_{pi}^2} = \ln \frac{AR}{E_a} - \frac{E_a}{RT_{pi}} \quad (2)$$

β_i is the heating rate, $Kmin^{-1}$; T_{pi} is the decomposition peak temperature of the explosive at the heating rate β_i , K; A is the pre-factor, min^{-1} ; R is the gas constant, $8.314 J mol^{-1} K^{-1}$; E_a is the apparent activation energy, $J mol^{-1}$.

Based on the results of the apparent activation energy (E_a) and the pre-factor (A), formula 3 and formula 14 [23] can be used to calculate critical temperature of thermal explosion. The calculation results are shown in Table 3.

$$T_{pi} = T_{p0} + b\beta_i + c\beta_i^2 \quad (3)$$

$$T_b = \frac{E_a - \sqrt{E_a^2 - 4RE_aT_{p0}}}{2R} \quad (4)$$

We can see from Table 3 that the apparent activation energy of cellulose/RDX composite aerogel spheres is 31.28 kJ/mol, which is higher when compared with that of RDX raw materials and means that the activation of RDX in cellulose/RDX composite aerogel spheres requires more energy and its stability is significantly improved, the increase of pre-exponential factor A also illustrates this point. This is because the three-dimensional cross-linked network structure and the excellent heat insulating properties of the cellulose aerogel blocks the transfer of heat effectively. There is no significant difference between the thermal explosion

critical temperature of cellulose/RDX composite aerogel spheres and RDX raw materials, which indicates the thermal explosion critical temperature of RDX has not been affected by the compounding of cellulose and RDX.

3.5 XRD Analysis

In order to investigate the change of crystal form after the combination of RDX and cellulose, the cellulose/RDX composite aerogel spheres, cellulose powder and RDX raw materials were characterized by X-ray diffraction, which is shown in Figure 5.

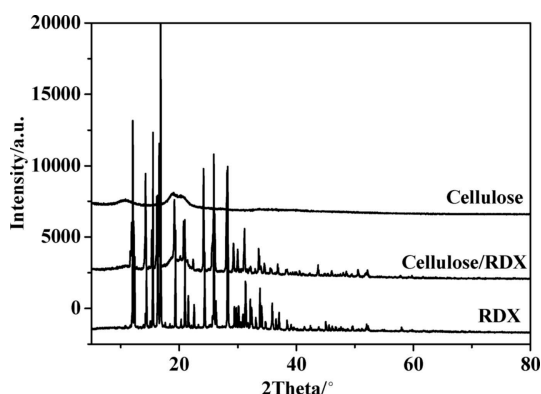


Figure 5. X-ray diffraction pattern of composite aerogel spheres, cellulose and RDX raw material.

As shown in Figure 5, the X-ray diffraction patterns of cellulose/RDX composite aerogel spheres contain almost all the characteristic peaks of RDX raw materials and cellulose powder, and the position of the characteristic peak does not change obviously. It means that the combination of RDX and cellulose aerogel did not affect the original crystal form and properties of cellulose and RDX. It can be seen that the intensity of the X-ray diffraction peak of the cellulose/RDX composite aerogel spheres is significantly weaker than that of the RDX raw materials, such as at $2\theta = 12^\circ$, 15.5° , 17° , 26° , 28° , and so on. It is considered that cellulose aerogel has obvious amorphous characteristics, after RDX is combined with the cellulose aerogel spheres, a large number of RDX particles are attached to the cross-linking network of the cellulose matrix, thus there are very few RDX particles on the surface of the sphere. The protective effect of cellulose aerogel spheres on RDX particles weakens the diffraction peak intensity of RDX.

3.6 Impact Sensitivity Analysis

An HGZ-1 impact instrument was used to characterize the mechanical sensitivity of the RDX raw materials, cellulose/

RDX composite aerogel spheres and mechanical mixture of RDX and cellulose. In order to mix cellulose and RDX raw materials evenly, RDX particles were obtained by the method used to prepare cellulose/RDX composite particles. RDX particles were dissolved in acetone and precipitated rapidly at lower temperature. Then the RDX was added into the cellulose suspension according to the mass ratio of RDX:cellulose = 85.2:14.8. The mixture was stirred quickly for 20 minutes, then frozen in liquid nitrogen and put it in the freeze dryer to freeze-dry. The results of the impact sensitivity test are shown in Table 4.

Table 4. Impact sensitivity test results of cellulose/RDX composite aerogel spheres, RDX materials and mechanical mixture of RDX and cellulose.

samples	Impact sensitivity H50/cm (S)				
	Test 1	Test 2	Test 3	Test 4	average
RDX raw materials	17.3	20.1	22.6	19.4	19.85
mechanical mixture of RDX and cellulose	60.2	58.4	62.3	63.7	61.15
Cellulose/RDX composite aerogel spheres	> 85	> 85	> 85	> 85	> 85

After compounding RDX with cellulose aerogel spheres, the impact sensitivities of the cellulose/RDX composite aerogel spheres decreased greatly compared with RDX raw materials, and H50 increased from 19.4 cm to more than 85 cm. Compared with the mechanical mixture of RDX and cellulose under the same conditions, the H50 increased by more than 23 cm.

This is because the cellulose/RDX composite aerogel sphere maintains the same three-dimensional cross-linked network structure as the pure cellulose aerogel, most of the RDX particles are present inside the cellulose/RDX composite aerogel sphere, few RDX particles remain on the surface of the composite aerogel sphere and the cellulose aerogels as coating layer has a protective effect on the internal RDX particles. The stable cross-linked structure not only has a certain compressive property but also has a buffering effect on the impact. The impact of the RDX particles is greatly weakened, and it is difficult to form the hot spot, so that the impact sensitivity is significantly reduced.

3.7 Detonation Velocity Calculation

The detonation speed of cellulose/RDX composite aerogel spheres was calculated with the computer software EXPLO5. The detonation speed of the cellulose/RDX composite aerogel spheres was calculated under the conditions of density of 90% and 95%. The calculation result shows that the detonation speed at 90% pressure density is 7832 m/s,

and the detonation speed calculation result is 8112 m/s when the pressure density is 95%.

4 Conclusion

Cellulose spheres successfully combined with RDX and cellulose/RDX composite aerogel spheres were prepared successfully. The main conclusions are as follows:

- (1) The diameter of the sphere is about 1.5 mm, and the interior is three-dimensional cross-linked network structure. RDX is attached to the cellulose fibers inside the cellulose aerogel sphere. The nitrogen content is as high as 32.24%, and the RDX content is 85.2%.
- (2) Compared with RDX raw materials, the size of RDX particles in cellulose/RDX composite aerogel spheres is reduced greatly. The apparent activation energy is increased by 31.28 kJ/mol, and the thermal stability is improved. The X-ray diffraction characteristic peak position do not change, maintaining the original crystal form characteristics.
- (3) Compared with the raw material RDX, the impact sensitivity of the cellulose/RDX composite aerogel sphere is greatly reduced. The results obtained in four experiments all show that H50 > 85 cm.

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