Application of Supercritical CO_2 Foaming Technology for Waste Double-Base Propellants

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ABSTRACT: This paper used a supercritical CO_2 batch foaming process to treat a waste SP double-base propellant, which is a type of double-base propellants containing 58.6% nitrocellulose, 40.0% nitroglycerin, 0.8% centralite, and 0.5% vaseline, to solve a problem of poor stability of industrial explosives directly prepared by the propellant. Experiments show that this process can produce dense pores inside the SP double-base propellant. With the increase of the pressure of supercritical CO_2, the number of pores inside the foamed SP double-base propellant increased, and these pores served as hotspots in the detonation reaction. An increased number of hotspots improved the detonation stability of the perfusion explosive. During the explosion, the energy of the perfusion explosive with the foamed SP double-base propellant was released more completely, so the shock wave energy and bubble energy of the explosive gradually increased with the increase of pressure. Therefore, the supercritical CO_2 foaming process can promote the treatment technology of waste double-base propellants and can optimize the detonation performance of perfusion explosives by increasing the pressure of supercritical CO_2.
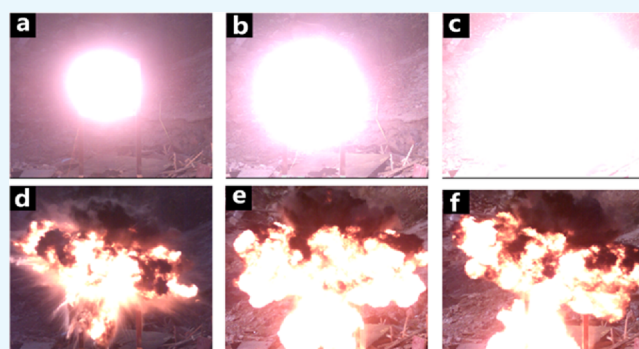
1. INTRODUCTION

Mortar is the main land warfare weapon for countries around the world, and the commonly used launch energy is generated from SP double-base propellants.[1] The SP double-base propellant is a nitrate-ester-base propellant containing 58% nitrocellulose, 40% nitroglycerin, and some centralite and vaseline. The life of the propellant is generally 20−30 years. After the expiration date,[2] the performance of propellants cannot meet the requirements of use and must be processed in time.[3] Since the double-base propellant contains some nitroglycerin and the compatibility between nitrocellulose and nitroglycerin results in poor performance,[4,5] the direct preparation of this double-base propellant into an industrial explosive leads to a disadvantage of incomplete detonation.[6] Its detonation stability is inferior to industrial explosives prepared with a single-base propellant, and sensitization is one of the ways to improve the detonation stability. At present, a large number of waste double-base propellants like SP double-base propellants have been decommissioned in various military powers, and thus, it is urgently desirable to study formulations and technologies[7−9] for preparing industrial explosives[10,11] from these double-base propellants.

Supercritical CO_2 foaming technology has advantages of easy realization of critical conditions, low cost, and easy operation. The technology is widely used in the preparation of porous materials. Through a foaming process, a large number of pores are generated inside the material to increase the inner surface area, which improves mechanical performance,[12] density[13] and thermal conductivity.[14] In the field of energetic materials, gun propellants should have high burning rates and better impact strength.[15] The foaming process can increase the inner surface of the propellant, which can increase the burning area and improve the linear burning rate of gun propellants.[16,17] Therefore, the supercritical CO_2 foaming technology has a good application prospect in the field of propellants.[18] It is of great significance to study the influence of a foaming process on the combustion performance of propellants. The effect of increasing the inner surface area by the foaming process can not only be used in the combustion of a propellant but also in the explosion of explosives[19,20] requiring a microporous structure as a detonation hotspot. These pores can provide numerous hotspots for detonation and improve the detonation stability of explosives.[21]

This paper used a supercritical CO_2 batch foaming technology to sensitize an SP double-base propellant and studied the CO_2 absorption, internal structure, and explosion

heat of the foamed SP double-base propellant. The foamed SP double-base propellant was used to prepare a perfusion explosive. Detonation performance such as the shock wave energy and bubble energy of the underwater explosion of the perfusion explosive as well as the fireball shape of the explosion process were studied. To prove the effect of the foaming process on the SP double-base propellant, the experiment here mainly investigated the foaming effect of different reaction pressures[22] of supercritical CO_2. The effect of pressure on the performance of the foamed SP double-base propellant and the detonation performance of the perfusion explosive was studied.

2. EXPERIMENT

2.1. Preparation of Foamed SP Double-Base Propellant. An SP double-base propellant used here was an $80 \times 20 \times 2$ mm rectangular propellant composed 58.6% of nitrocellulose, 40.0% nitroglycerin, 0.8% of centralite, and 0.5% vaseline. In this experiment, a supercritical batch foaming process was used to prepare a foamed SP double-base propellant. The experimental scheme is shown in Figure 1. A
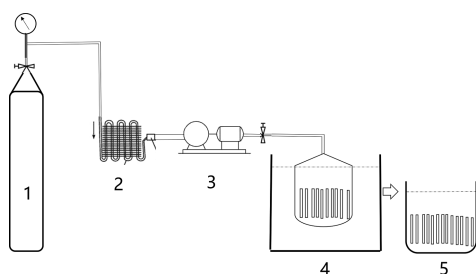

Figure 1. Scheme for preparation of foamed SP double-base propellant (1, CO_2 cylinder; 2, condenser; 3, pump; 4, high-pressure vessel; 5, thermostat water bath).

sample of the SP double-base propellant was placed into a high-pressure vessel to absorb CO_2 so as to form a SP-supercritical CO_2 system after 1 h. Then, the sample was taken out and weighed immediately, and the amount of CO_2 absorbed was calculated. Then, the sample was kept in a hot bath at a temperature of 70 °C for foaming treatment for 3 min. After the escape of CO_2 in the system, the sample was placed in an oven to drive away water. Other experimental conditions are shown in Table 1.

Table 1. Conditions of the CO_2 Reaction Foaming Process

process	condition	value
CO_2 reaction	pressure (MPa)	8, 9, 10, 11
	temperature (°C)	40
	time (h)	1
foaming	pressure (MPa)	0.1
	temperature (°C)	70

2.2. Preparation of Perfusion Explosive with Foamed Propellant. The sheet-shaped foamed SP double-base propellant was bundled as a 2−3 cm thick cuboid and then placed into a mold for later use. Since the foamed SP double-base propellant was originally in the form of a sheet, it was not easy for the perfusion liquid to enter the mold containing the scattered SP double-base propellant, which causes nonuniform density of the perfusion explosive. Bundling as a cuboid allowed the perfusion liquid to better penetrate to the bottom

of the mold to provide a perfusion explosive with uniform density. In the preparation of the perfusion liquid, a cross-linking agent was put into a beaker and melted in a 60 °C water bath. Ammonium nitrate and sodium nitrate were then added in certain proportions into the molten liquid, and then the mixture was stirred uniformly. Before pouring into the mold, an initiator, a retarder, and a catalyst were added to the mixed perfusion solution. Finally, the perfusion liquid was poured into the mold and left to cool for 5 h. The specific operation process is shown in Figure 2.
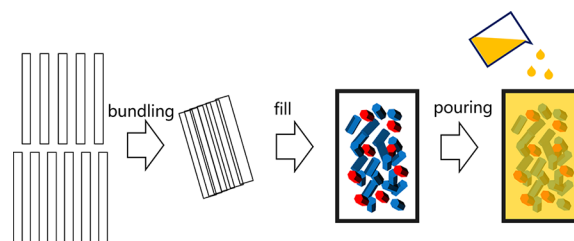

Figure 2. Scheme for preparation of perfusion explosive.

2.3. Characterization of Foamed Propellant. The foamed SP double-base propellant was put into low-temperature liquid nitrogen, kept for 30 min, and then subjected to brittle failure. After spraying with gold on the section, the internal structure of the foamed SF-3 propellant was then observed by a QUANTA FEG 250 scanning electron microscope (FEI USA) at an accelerating voltage of 5 kV so the effect of the pressure of supercritical CO_2 on the internal structure of the SP double-base propellant can be studied.

The section of the foamed SP double-base propellant was observed by a RENISHAW Raman spectrum scanner, and the pore distribution was analyzed by collecting characteristic peaks of nitrocellulose and nitroglycerin of the SP double-base propellant. The wavelength of laser light was 532 nm. The scanning area was 50×500 μm.

Explosion heat of the foamed SP double-base propellant was tested by a ZDHW-6 W computer calorimeter from Hebi Auvai Dimension Electronic Technology Co., Ltd. (Hebi, China) in order for us to analyze the relationship between the pressure and the explosion heat of foamed SP double-base propellants. The gas in the oxygen bomb was high-purity nitrogen at a pressure of 2 MPa. A nickel−chromium wire with a length of 10 cm and a mass of 1.0000 ± 0.0002 g was used as an ignition wire.

2.4. Testing of the Performance of the Perfusion Explosive. The destructive ability of the perfusion explosives to the steel plate can be compared through the detonation test, and the difference in the functional power of the explosives can be obtained. A sample of the perfusion explosive was placed on a steel plate with a size of $200 \times 200 \times 12$ mm and detonated. After the explosion, the hole diameter of the steel plate was measured. A booster explosive used in this experiment was passivated RDX (cyclotrimethylene trinitramine), which was made by using a hydraulic machine to press 40 g of passivated RDX into a cylinder with a diameter of 40 mm. The density of the booster explosive was 1.6 g/cm³. The detonation velocity was measured by an ion probe method. Explosive samples for underwater explosion and high-speed photography experiments were also prepared by the method described above.

The underwater explosion experiment can better reflect the energy of the perfusion explosive with the foamed SP double-

base propellant, where the shock wave energy and the bubble energy of the explosive can be obtained from the underwater explosion experiment. The device for the underwater explosion experiment is shown in Figure 3. The underwater explosion
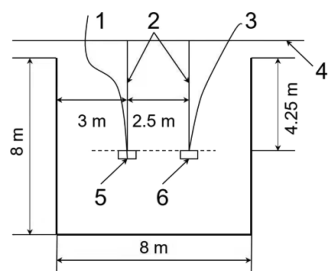

Figure 3. Device for underwater explosion experiment. (1, ignition cable; 2, load-bearing rope; 3, transmission cable; 4, experimental holder; 5, test device; 6, sensors).

pool was a cylindrical pool with a diameter of 8 m and a depth of 8 m. The explosive sample was placed at a depth of 4.25 m below the water surface. The center of the sensor was flush with the center of the explosive. The experimental data were recorded by a sensor (PCB138M), a signal disposal instrument (PCB482A16), and a data collector (JOVIAN 5200), respectively.[20]

The sample for the underwater explosion experiment should be reinforced with a layer of waterproof film and sealed with a tape to prevent the sample from entering water. In order to reduce errors resulted from buoyancy, it was necessary to tie a stone with a mass of about 1 kg to the bottom of the explosive.

The detonation process was analyzed with pictures taken by a high-speed camera. The brightness and fireball size were compared during different time periods of the detonation. A PCO.Dimax (COOK USA) high-speed camera was used in the experiment, and the frame rate was 13,000 frames/min.

3. RESULTS AND DISCUSSION

3.1. Morphology and Energy of Foamed Double-Base Propellant.
The solubility of CO_2 in the double-base propellant is shown in Figure 4. According to the calculation
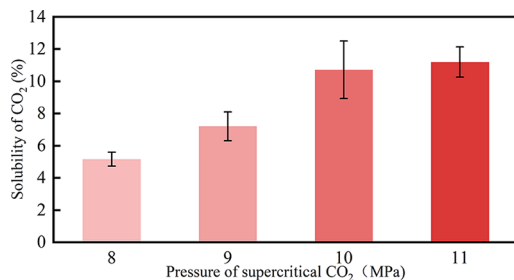

Figure 4. Solubility of supercritical CO_2 at different pressures in SP double-base propellant.

results, the effect of the pressure of supercritical CO_2 on its solubility in the double-base propellant was compared.[23] It can be seen from Figure 4 that with the increase of pressure, the content of supercritical CO_2 in the double-base propellant gradually increased, and thus, it is assumed that the solubility of supercritical CO_2 gradually increased as well.[24] However, after the pressure exceeded 10 MPa, the speed of increase in the amount of CO_2 dissolved decreased. The SP-supercritical

CO_2 system tended to be saturated within the same saturation time, so the dissolution of CO_2 slowed down.

The color of the SP double-base propellant turned lighter after foaming. The supercritical foaming process produced many pores inside the sample, which achieved the effect of sensitization. During the experiment, the foaming effect in some samples was not good as a small number of small foaming dots could be seen on the surface of the sample, while the well-foamed sample had a uniform color on the surface and more pores inside. The reason for the defective products is that the high-pressure reactor is not well sealed during the reaction, making the pressure fail to reach the reaction condition.

A microstructure of the foamed SP double-base propellant under a scanning electron microscope is shown Figure 5. It can be seen from Figure 5 that compared with the unfoamed sample, there were many pores in the foamed SP double-base propellant after the gas escaped.[25] The sample foamed at a pressure of 8 MPa only had a small number of cracks. Pores began to appear around the cracks at a pressure of 9 MPa. As the pressure increased, the number of pores around the cracks gradually increased. The pores inside the SP double-base propellant foamed at a supercritical CO_2 pressure of 11 MPa had a small size and a high-density distribution. The reason is that as the pressure increased, more supercritical CO_2 was dissolved into the sample uniformly. After heating and foaming, the gas escaped from the layered structure and caused dense pores in the layered structure.[26] Therefore, as the pressure increased, the pores inside the foamed SP double-base propellant increased and the structure became loose.

Results of the Raman test conducted on the double-base propellant are shown in Figure 6. Contents of active ingredients were obtained by comparing collected characteristic peaks of the foamed propellant with those of the unfoamed sample. The active ingredients of the unfoamed double-base propellant were uniformly distributed on the cross section of the testing area, and the color was uniform. When pores were formed inside the sample, no characteristic peak appeared in the pore part within the same scanning area, so the contents of active ingredients were low in the sample. In the scanning area, the shaded area of sample 2# was small and distributed outside the test surface. This is because CO_2 penetrated into the SP propellant from the outside to the inside. In the same time period, the CO_2 permeation was smaller when the pressure was lower, and thus CO_2 was distributed outside the sample. After the foaming process, this CO_2 escaped the sample to form pores. Therefore, when the reaction pressure was 8 MPa, the pores were mainly distributed on the outside. The 4# sample foamed at a pressure of 10 MPa had uniformly distributed pores and a large shadow area, proving that the foaming process provided the sample with a better pore structure. The shadow area in the middle of the section of sample 5# foamed at a pressure of 11 MPa was increased, and more shadows were distributed on the outer part. It can be seen that as the pressure of supercritical CO_2 increased, the area of the black-colored part in the middle gradually increased. Combining with the dissolution of supercritical CO_2 in the double-base propellant, it can be seen that as the pressure increases, the solubility of carbon dioxide gradually increases; thus, the number of pores formed inside also gradually increases and the content of active ingredients per unit area decreases. This is consistent with the SEM results. When the pressure was 8 MPa, the small shaded area in the middle of the sample may result from that the SP-
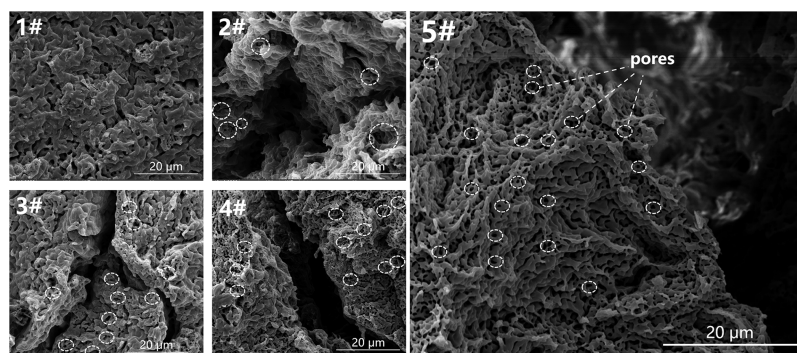
Figure 5. SEM images of double-base propellants foamed at different pressures (reaction pressure: 1#, unfoamed; 2#, 8 MPa; 3#, 9 MPa; 4#, 10 MPa; 5#, 11 MPa).
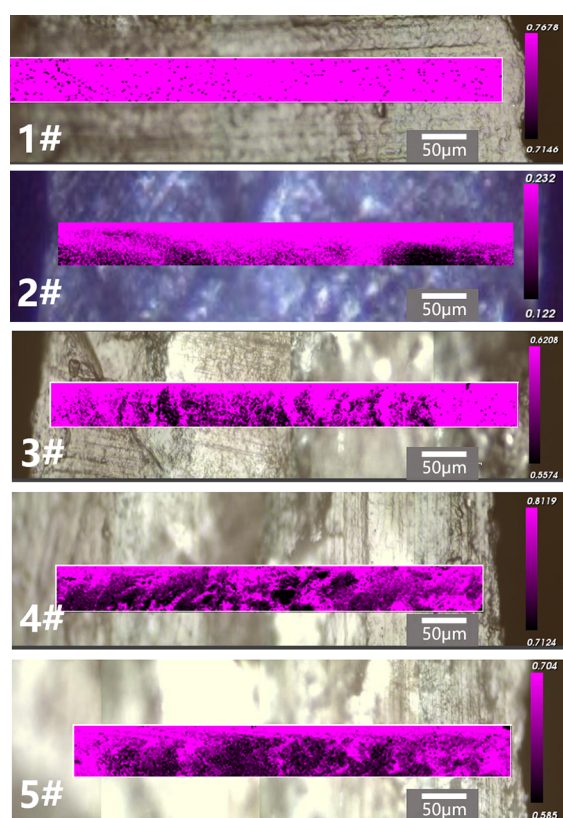

Figure 6. Raman images of double-base propellants foamed at different pressures (reaction pressure: 1#, unfoamed; 2#, 8 MPa; 3#, 9 MPa; 4#, 10 MPa; 5#,11 MPa).

Figure 7. Explosion heat of foamed propellants.

increase of the pressure of supercritical CO_2, the change of the explosion heat value of the foamed SP double-base propellant was small. Thus, it is proven that the pressure from 8 to 11 MPa has a small effect on the energy of the foamed SP double-base propellants, and the foaming process can be effectively applied to the treatment of waste SP double-base propellants.

3.2. Performance of Perfusion Explosive. Results of detonation test of perfusion explosives with SP double-base propellants foamed at different pressures are shown in Table 2.

Table 2. Diameter of Explosion Hole and Detonation Velocity of Perfusion Explosives

sample	reaction pressure (MPa)	diameter of blast hole (cm)	detonation velocity (m/s)
1		indentations	0
2	8	5.2	6403
3	9	5.1	6501
4	10	5.2	6625
5	11	5.2	6734

Table 2 shows that the perfusion explosive with the unfoamed SP double-base propellant had no detonation performance. During the test, an explosion sound could be heard when the perfusion explosive containing the unfoamed sample was detonated, but there was no hole caused in the steel plate. After the explosion, a large amount of black smoke was found to rise, accompanied by a strong smell of gunpowder. The sound of the explosion was caused by the electric detonator and the booster explosive, and the booster explosive failed to make the perfusion explosive explode. The black smoke and smell produced in the experiment were caused by the high temperature, which resulted in the combustion of the SP

supercritical CO_2 system had not reached equilibrium. When the pressure was 11 MPa, the shaded parts were uniformly distributed on the test surface, proving that the pores were uniformly distributed on the cross section of the foamed SP propellants. This indicates that the system is close to an equilibrium state. Therefore, a higher pressure is likely to form a saturation SP-supercritical CO_2 system. On the other hand, the reaction time may be too short for samples formed at a lower reaction pressure, making the system fail to reach the equilibrium.[21]

The explosion heat value of the foamed SP double-base propellant is shown in Figure 7. The unfoamed SP double-base propellant had an explosion heat value of 5042 J/g, and the SP double-base propellant foamed at a pressure of 8 MPa had an explosion heat value of 4649 J/g. It can be seen that with the
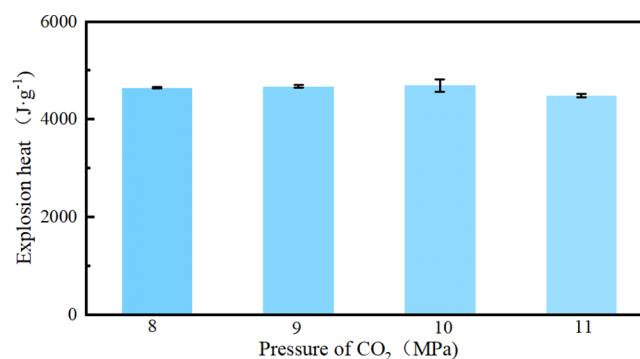
double-base propellant in the perfusion explosive. However, the perfusion explosive with the foamed SP double-base propellant produced an effective detonation, causing the steel plate to break. When the pressure of supercritical CO_2 was 8 MPa, the diameter of the blast hole in the steel plate was 5.2 cm. For other perfusion explosives with foamed SP double-base propellant, the diameter of the blast hole in the steel plate was around 5.2 cm, proving that the explosive made from foamed SP double-base propellants can detonate.

The detonation velocity of the perfusion explosive with the SP double-base propellant is shown in Table 2. It can be seen from Table 2 that the perfusion explosive with the unfoamed SP double-base propellant failed to detonate, and thus, the detonation velocity was 0 m/s. When the SP double-base propellant was treated by 8 MPa supercritical CO_2, the detonation velocity of the perfusion explosive was 6403 m/s. As the pressure of supercritical CO_2 increased, the detonation velocity of the perfusion explosive gradually increased. When the pressure was 11 MPa, the detonation velocity of the perfusion explosive reached 6734 m/s.

Figure 8 shows results of shock wave energy and bubble energy in underwater explosion experiments of perfusion
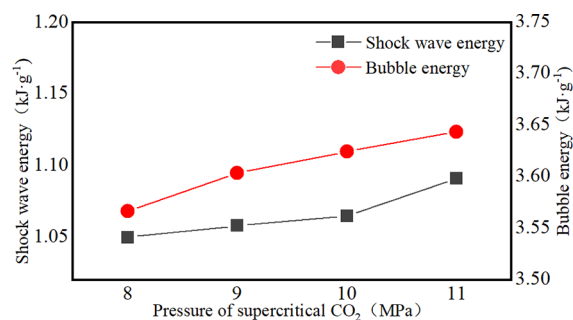

Figure 8. Variation of shock wave energy and bubble energy of perfusion explosive as a function of pressure.

explosives with SP double-base propellants foamed at different pressures. Figure 8 also shows that the shock wave energy and the bubble energy were 0.29 and 1.45 kJ/g, respectively, for the perfusion explosive with the unfoamed propellant and 1.050 and 3.567 kJ/g, respectively, for the perfusion explosive with the propellant foamed at 8 MPa. This proves that the perfusion explosives with the foamed SP double-base propellant could be effectively detonated. When the explosive exploded, the collapse of a large number of pores in the foamed SP double-base propellant caused a large amount of heat to be accumulated locally.[27,28] If the heat accumulation efficiency is too high and the heat transfer rate is too low, the temperature will rise rapidly to form a hot spot.[29,30] If the hot spot has a temperature higher than the reaction temperature, a violent chemical reaction will occur. When the heat accumulates to a certain degree, deflagration or detonation will occur.[31] The bubble of the perfusion explosive resulted from the secondary reaction of the explosive in the explosion. With the increase in the pressure of supercritical CO_2, the bubble energy gradually increased, reaching a maximum of 3.644 kJ/g in this experiment.

The shock wave energy and bubble energy of the perfusion explosive with the SP double-base propellant foamed at a pressure of 9 MPa were 1.058 and 3.604 kJ/g, respectively. It can be seen that the energy of this explosive was higher than that of the sample foamed at a pressure of 8 MPa. When the

pressure reached 11 MPa, the shock wave energy and bubble energy of the underwater explosion of the perfusion explosive reached 1.091 and 3.644 kJ/g, which were 3.91 and 2.16% higher than the sample foamed at 8 MPa. With the increase of pressure, the shock wave energy and bubble energy of the perfusion explosive gradually increased. According to the analysis of the solubility of CO_2, as the pressure increases, the per unit mass of the SP double-base propellant absorbs more supercritical CO_2. The number of pores inside the foamed SP double-base propellant will increase, so the hotspots of the prepared perfusion explosive also increase. Therefore, when the detonation of the perfusion explosive occurs, the detonation stability is improved, and the release of energy from the detonation becomes relatively complete.[32]

The explosives released huge amounts of energy during the detonation, and the released energy generated detonation waves. Fireballs of different shapes appeared during the detonation process. From Figure 9, it can be seen that the
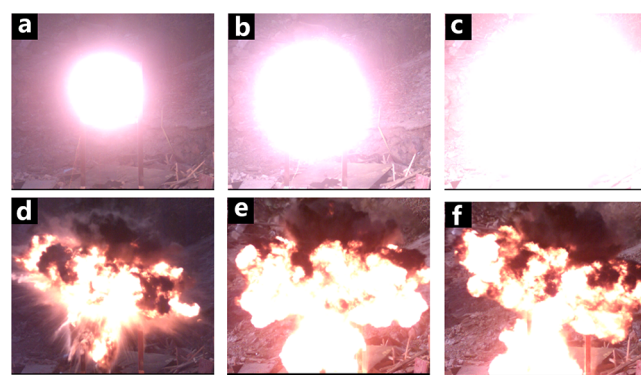

Figure 9. Change of fireball in explosion process (detonation time: a−c, 100 μs; d−f, 1 ms; reaction pressure: a and d, unfoamed; b and e, 8 MPa; c and f, 11 MPa).

perfusion explosive with the unfoamed SP double-base propellant produced a small fireball and a large amount of black smoke during the detonation process. The black smoke over the test field caused a pungent burning smell of gun propellant. At 100 μs of detonation, the unfoamed sample provided a small diameter of the fireball, which was smaller than that of the perfusion explosive with the foamed SP double-base propellant. At 1 ms of the detonation, the firework-like smoke balls were sprayed around. This was because the shock wave of the booster explosive scattered the sample during the detonation process, and the high temperature caused the unfoamed propellant to burn,[33] resulting in the jet of black smoke.

During the explosion process, the fireball of the perfusion explosive with the foamed SP double-base propellant gradually increased in size, with high brightness. When the SP double-base propellant was one foamed at a pressure of 8 MPa, the fireball was larger than that in the case of the unfoamed sample. No jet of black smoke appeared during the detonation process, proving that the perfusion explosive had undergone a complete detonation. At 1 ms of the detonation, the explosive with the SP double-base propellant exploded as there were many pores generated by the foaming inside the explosive. These pores formed hotspots for the detonation process and the hotspots were compressed,[34,35] thus improving the stability of the explosion.

4. CONCLUSIONS

The supercritical CO_2 foaming process produced dense pores in the SP double-base propellant, thereby promoting the treatment technology of waste SP double-base propellants. As the pressure of supercritical CO_2 increased, the solubility of CO_2 in the SP double-base propellant gradually increased. Then, dissolved CO_2 became gas and escaped from the sample under the action of the high temperature water bath, resulting in many pores in the foamed SP double-base propellant. When the pressure was 9 MPa, cracks appeared in the middle of the cross section of the SP double-base propellant. More CO_2 was dissolved in the SP double-base propellant at higher pressure. During the foaming process, the gas concentratedly escaped from some pores and formed cracks. When the pressure was 11 MPa, the number of pores inside the foamed SP double-base propellant reached the maximum and the pore size became smaller. The explosion heat experiment showed that the explosion heat value of the foamed SP double-base propellant was marginally affected by the pressure of supercritical CO_2, proving that the performance of the sample can be adjusted by changing the pressure of the process.

In performance testing of the perfusion explosive, the perfusion explosive with the unfoamed SP double-base propellant failed to detonate effectively. The perfusion explosive with the foamed SP double-base propellant had good detonation performance, as it caused holes in a 12 mm thick steel plate in the detonation test and generated shock waves and bubble energy in the underwater explosion experiment. The pores produced by supercritical foaming were sharply compressed to form hotspots during the detonation, thereby promoting the release of detonation energy of the perfusion explosive. When the pressure of supercritical CO_2 was 8 MPa, the shock wave energy of the perfusion explosive was 1.050 kJ/g, and the bubble energy was 3.567 kJ/g. With the increase of pressure, the shock wave energy and the bubble energy of the explosive with foamed SP double-base propellant gradually increased. When the pressure was 11 MPa, the shock wave energy and the bubble energy of the perfusion explosive were increased by 3.91 and 2.16%, respectively, compared with those at the pressure of 8 MPa. It is proven that increasing the pressure of supercritical CO_2 helps to improve the detonation performance of a perfusion explosive with a foamed propellant.

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The manuscript was written through the contributions of all authors.

Notes

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■ REFERENCES

(1) Trache, D.; Tarchoun, A. F. Analytical Methods for Stability Assessment of Nitrate Esters-Based Propellants. *Crit. Rev. Anal. Chem.* **2019**, *49*, 415−438.

(2) Mehilal; Dhabbe, K. I.; Kumari, A.; Manoj, V.; Singh, P. P.; Bhattacharya, B. Development of an Eco-Friendly Method to Convert Life Expired Composite Propellant into Liquid Fertilizer. *J. Hazard. Mater.* **2012**, *205-206*, 89−93.

(3) Boulkadid, M. K.; Lefebvre, M. H.; Jeunieau, L.; Dejeaifve, A. Burning Rate of Artificially Aged Solid Double-Base Gun Propellants. *J. Energy Mater.* **2020**, *38*, 1−19.

(4) Wu, W.; Yang, Y.; Chen, J.; Li, Y.; Cheng, Y. Nitroglycerine Migration to Polyurethane Used for Inhibition of Double-Base Propellants through Thermal Analysis Techniques. *J. Therm. Anal. Calorim.* **2016**, *125*, 881−886.

(5) Agrawal, J. P.; Singh, H. Qualitative Assessment of Nitroglycerin Migration from Double-Base and Composite Modified Double-Base Rocket Propellants: Concepts and Methods of Prevention. *Propellants, Explos., Pyrotech.* **1993**, *18*, 106−110.

(6) Reese, D. A.; Groven, L. J.; Son, S. F. Formulation and Characterization of a New Nitroglycerin-Free Double Base Propellant. *Propellants, Explos., Pyrotech.* **2014**, *39*, 205−210.

(7) Shim, Y.-H.; Kim, J.-T. Successful Plant Scale Production of Solid Propellant Recycling from Obsolete Ammunition. *J. Mater. Cycles Waste Manage.* **2017**, *19*, 898−905.

(8) Sa, A. A.; Dhabbe, K.; Kulkarni, P.; Mehilal. Studies on Conversion of Waste Nitramine and Fuel-Rich-Based Propellants into Liquid Fertilizer. *Environ. Technol.* **2019**, *40*, 1035−1042.

(9) Wang, M.; Du, S.; Yan, J. Study on the Extraction of AP Components in Htpb Solid Propellant by Water/Ethanol Recovery. *AIP Conf. Proc.* **2017**, *1890*, 040006-1−040006-7.

(10) Wang, P.; Wei, X. A.; He, W. D.; Shi, Y. L. Applications and Technologies of Reusing Waste Gun Propellants. *Advanced Materials Research* **2013**, *781-784*, 1998−2002.

(11) Ravi, P.; Badgujar, D. M.; Gore, G. M.; Tewari, S. P.; Sikder, A. K. Review on Melt Cast Explosives. *Propellants, Explos., Pyrotech.* **2011**, *36*, 393−403.

(12) Li, J.; Liao, X.; Jiang, Q.; Wang, W.; Li, G. Creating Orientated Cellular Structure in Thermoplastic Polyurethane through Strong Interfacial Shear Interaction and Supercritical Carbon Dioxide Foaming for Largely Improving the Foam Compression Performance. *J. Supercrit. Fluids* **2019**, *153*, 104577.

(13) Li, M. J.; Xiang, K.; Luo, Q. G.; Shen, Q.; Zhang, L. M. Preparation and Density Control of Pmma Microcellular Foams via

Supercritical Carbon Dioxide Foaming. *Key Eng. Mater.* **2014**, *616*, 242−246.

(14) Wang, J.; Chai, J.; Wang, G.; Zhao, J.; Zhang, D.; Li, B.; Zhao, H.; Zhao, G. Strong and Thermally Insulating Polylactic Acid/Glass Fiber Composite Foam Fabricated by Supercritical Carbon Dioxide Foaming. *Int. J. Biol. Macromol.* **2019**, *138*, 144−155.

(15) Wang, Y.; Jiang, L.; Dong, J.; Li, B.; Shen, J.; Chen, L.; Fu, Y.; He, W. Three-Dimensional Network Structure Nitramine Gun Propellant with Nitrated Bacterial Cellulose. *J. Mater. Res. Technol.* **2020**, *9*, 15094−15101.

(16) Yang, W.; Li, Y.; Ying, S. Fabrication of Graded Porous and Skin-Core Structure RDX-Based Propellants via Supercritical CO_2 Concentration Profile. *J. Energ. Mater.* **2015**, *33*, 91−101.

(17) Li, Y.; Yang, W.; Ying, S. Preparation and Characteristics of Foamed NC-Based Propellants. *Propellants, Explos., Pyrotech.* **2014**, *39*, 677−683.

(18) Li, Y.; Yang, W.; Ying, S. The Effects of Porous Structure on the Burning Characteristics of Foamed NC-Based Gun Propellants. *Propellants, Explos., Pyrotech.* **2014**, *39*, 852−858.

(19) Wei, X.; Zhang, D.; Chen, F.; Wang, Z. Research on Perfusion Explosives from SF-3 Double-Base Propellants Using Supercritical CO_2. *Propellants, Explos., Pyrotech.* **2016**, *41*, 740−745.

(20) Zhang, D.; Wei, X.; He, W.; Chen, F.; Wang, Z. Research on Preparation of Perfusion Explosive Using Foamed SF-3 Double-Base Propellant. *Propellants, Explos., Pyrotech.* **2017**, *42*, 1179−1184.

(21) Tan, D.; Wei, X.; Zhang, D.; Chen, F.; He, W.; Wang, Z. Process Optimization of Supercritical CO_2 Foamed SF-3 Double-Base Propellant. *Propellants, Explos., Pyrotech.* **2020**, *45*, 20−25.

(22) Azimi, H.; Jahani, D. The Experimental and Numerical Relation Between the Solubility, Diffusivity and Bubble Nucleation of Supercritical CO_2 in Polystyrene Via Visual Observation Apparatus. *J. Supercrit. Fluids* **2018**, *139*, 30−37.

(23) Roberta, C.; Paola, F.; Ernesto, R.; Iolanda, D. M. Polycaprolactone/Nimesulide Patches Obtained by a One-Step Supercritical Foaming + Impregnation Process. *J. Supercrit. Fluids* **2019**, *146*, 47−54.

(24) Zhang, M.; Dou, M.; Wang, M.; Yu, Y. Study on the Solubility Parameter of Supercritical Carbon Dioxide System by Molecular Dynamics Simulation. *J. Mol. Liq.* **2017**, *248*, 322−329.

(25) Wong, A.; Guo, Y.; Park, C. B. Fundamental Mechanisms of Cell Nucleation in Polypropylene Foaming with Supercritical Carbon Dioxide-Effects of Extensional Stresses and Crystals. *J. Supercrit. Fluids* **2013**, *79*, 142−151.

(26) Sarikhani, K.; Jeddi, K.; Thompson, R. B.; Park, C. B.; Chen, P. Effect of Pressure and Temperature on Interfacial Tension of Poly Lactic Acid Melt in Supercritical Carbon Dioxide. *Thermochim. Acta* **2015**, *609*, 1−6.

(27) Yi, J.; Zhao, F.; Xu, S.; Zhang, L.; Gao, H.; Hu, R. Effects of Pressure and Tegdn Content on Decomposition Reaction Mechanism and Kinetics of Db Gun Propellant Containing the Mixed Ester of Tegdn and Ng. *J. Hazard. Mater.* **2009**, *165*, 853−859.

(28) Pan, J.; Dong, S.; Wei, H.; Li, T.; Shu, G.; Zhou, L. Temperature Gradient Induced Detonation Development Inside and Outside a Hotspot for Different Fuels. *Combust. Flame* **2019**, *205*, 269−277.

(29) Chaudhri, M. M.; Field, J. E. The Role of Rapidly Compressed Gas Pockets in the Initiation of Condensed Explosives. *Proc. R. Soc. A* **1974**, *340*, 113−128.

(30) Towery, C. A. Z.; Poludnenko, A. Y.; Hamlington, P. E. Detonation Initiation by Compressible Turbulence Thermodynamic Fluctuations. *Combust. Flame* **2020**, *213*, 172−183.

(31) Hu, Y.; Brenner, D. W.; Shi, Y. Detonation Initiation from Spontaneous Hotspots Formed During Cook-Off Observed in Molecular Dynamics Simulations. *J. Phys. Chem. C* **2010**, *115*, 2416−2422.

(32) Wang, Z.; Qi, Y.; He, X.; Wang, J.; Shuai, S.; Law, C. K. Analysis of Pre-Ignition to Super-Knock: Hotspot-Induced Deflagration to Detonation. *Fuel* **2015**, *144*, 222−227.

(33) Bernecker, R. R.; Price, D. Burning to Detonation Transition in Porous Beds of a High-Energy Propellant. *Combust. Flame* **1982**, *48*, 219−231.

(34) Khokhlov, A. M.; Oran, E. S. Numerical Simulation of Detonation Initiation in a Flame Brush: The Role of Hot Spots. *Combust. Flame* **1999**, *119*, 400−416.

(35) Field, J. E.; Bourne, N. K.; Palmer, S. J. P.; Walley, S. M.; Sharma, J.; Beard, B. C. Hot-Spot Ignition Mechanisms for Explosives and Propellants. *Philos. Trans. R. Soc., A* **1992**, *339*, 269−283.