Short Communication

Propellants, Explosives, Pyrotechnics

DOI: 10.1002/prep.201900106

Effects of Crystal Quality and Morphology on the Mechanical Performance of LLM-105 Based PBXs

Zhijian Yang,*[a, b] Congmei Lin,[a] Feiyan Gong,[a] Chengcheng Zeng,[a] Jianhu Zhang,[a] and Fenglei Huang*[b]

Abstract: Mechanical performance is significant for the application of pressed polymer bonded explosives (PBXs) materials. In this research, several types of 2,6-diamino-3,5-dinitropyrazine-1-oxide (LLM-105) were selected to study the effects of crystal quality and morphology on the mechanical performance of LLM-105 based PBXs. The surface bonding status of the LLM-105 crystal and molding powder was characterized by scanning electron microscopy (SEM) and contact angle analysis, and the moldability was evaluated by determining the relative density of pressed cylinders of corresponding PBXs. Besides, static and dynamic mechanical properties were studied and compared, presenting the compressive, tensile and creep curves. For LLM-105 based PBXs, spherical morphology and rough surface could result improved mechanical strength and creep resistance, facilitating the structural and dimensional stability of charges. In this case, it can bring benefits for their reliability and safety performances during application.

Keywords: LLM-105 · polymer bonded explosives · mechanical performance · crystal quality · morphology

1 Introduction

During the past decades, PBXs have been extensively used for military and civilian purposes in view of their high energy and nice comprehensive performance [1–3]. Generally, PBXs are composed of high-loading explosives (*e.g.* 85–97 wt%) and a polymer as the matrix. For PBXs, it is widely accepted the detonation, safety, mechanical, environmental adaptation and storage performances are the most important characteristics as concerned for their application [4, 5]. Among them, the mechanical performance, including the mechanical strength, modulus and creep resistance, has attracted great attentions because it plays a key role to maintain the structural strength of explosive charges, facilitating the safety, reliability and long storage performances [6]. Considerable efforts have been devoted to improving the mechanical performance of PBXs, especially for the pressed ones because the content of polymer binder is fairly low. Interfacial modification, regulation of binder structure and particle filling are the most common and effective strategies to enhance the strength and toughness of PBXs as reported recently [7–10].

Apart from the adjustment in polymer matrix, their pristine state of the explosive particles, including the crystal quality and morphology, have obvious influence on the forming density and mechanical properties of PBXs. Specifically, polyhedral or spherical morphology, and the grain composition of proper particle size are usually profitable for molding and gaining high strength. In addition, the energetic crystals with high quality (compact internal structure, few impurities or defects) as prepared by recrystallization can not only reduce the shock wave sensitivity markedly, but also improve the crystal strength [11–13]. Nevertheless, the smooth crystal surface is definitely difficult to be bonded by the polymers, due to the lack of anchor points for connecting. In this case, weak interface interaction and poor tensile strength will be obtained. In other words, high-quality crystal is a double-edged sword for the mechanical performance, and sometimes the surface of explosive crystal needs to be roughened to create more anchor points at the interface.

LLM-105, as synthesized by Lawrence Livermore National Laboratory in 1995, is known as a potential insensitive high explosive (IHE) which has good prospects for application in insensitive munitions (IM) or boosters [14–17]. It has a nice safety performance and 15% higher energy than 1, 3, 5-triamino-2, 4, 6-trinitrobenzene (TATB) [18]. However, experimental results show that the mechanical performance, particularly the tensile strength of LLM-105 based PBXs is relatively poor due to the weak interface between LLM-105 and commonly used polymers, as compared with octahydro-1,3,5,7-tetranitro-1,3,5,7-tetrazocine (HMX), hexahydro-1,3,5-trinitro-1,3,5-triazine (RDX) and

[a] *Z. Yang, C. Lin, F. Gong, C. Zeng, J. Zhang*
Institute of Chemical Materials, China Academy of Engineering Physics,
Mianyang, 621900, P.R. China
**e-mail: zhijianyang@caep.cn*
[b] *Z. Yang, F. Huang*
Department of Engineering Mechanics, Beijing Institute of Technology,
Beijing 100081, P.R. China
**e-mail: huangfl@bit.edu.cn*

TATB based PBXs. Despite its high importance, up to now, few works have been reported on how to adjust the mechanical performance of LLM-105 based PBXs.

In this work, the effects of crystal quality by recrystallization and spherical morphology on the mechanical performance of various LLM-105 based PBX are studied. Structure characterizations, static and dynamic mechanical property tests were carried out. It is found that the surface acid treatment can largely enhance the tensile strength of LLM-105 with high crystal quality, and spherical morphology with rough surface is beneficial to gain apparently improved strength and creep resistance.

2 Experimental Section

2.1 Materials

LLM-105 with $>99\%$ purity and polyhedron morphology was synthesized by taking iminodiacetonitrile (IDAN) as the starting material and performing a three-step reaction including nitrosation, cyclization and nitration in our institute. Crystals with high quality were prepared by recrystallization using dimethyl sulfoxide (DMSO) as the solvent and ethyl acetate (EA) as the anti-solvent, surface acid treatment of the high-quality LLM-105 was performed in 5 wt% nitric acid solution under stirring for 4 h, followed by filtration, washing with excessive hot water for several times and drying in vacuum oven at 100 °C for 48 h. Spherical LLM-105 crystals with smooth and rough surface were prepared according to the literature [19]. The fluoropolymer binder F2314, copolymerized by vinylidene fluoride and chlorotrifluoroethylene with mole ratio of 1:4, were provided by Zhonghao Chenguang Chemical Industry Co., Ltd. China. All the other reagents of analytical grade were commercially obtained and used without further purification.

2.2 Preparation of LLM-105 Based PBXs

LLM-105 based PBXs were prepared by a water suspension granulation method according to the formulation of LLM-105/binder = 95/5 in weight. The fluoropolymer was dissolved in ethyl acetate and added dropwise into the explosive suspension, molding powder products were obtained as the polymer coated on the LLM-105 grains. The pressed specimens with different size were prepared under 120 °C preheating and pressed in a stainless steel mould at 380 MPa, the relative density of the specimens was calculated by measured/ theoretical density.

2.3 Characterization

The morphology of samples was tested by scanning electron microscopy (SEM) measurements with a LE0438VP instrument at an operating voltage of 25 kV. The static contact angle test was conducted by a Krüss DSA100 instrument, a molding cylinder was prepared by pure LLM-105, tested with F2314/ethyl acetate droplets, then the contact angle was obtained. A Laser Particle Sizeron Coulter LS230 was adopted to conduct the particle size distribution (PSD) measurements. Differential scanning calorimeter (DSC) test was recorded on a Mettler Toledo instrument from 50 °C to 450 °C under nitrogen (40 ml/min) atmosphere with ramp of 10 °C/min. Static mechanical tests were performed with a universal testing machine (INSTRON 5582, USA) at room temperature, with pressed cylindrical specimens of $\Phi 20\ mm \times 20\ mm$ and $\Phi 20\ mm \times 6\ mm$ for the compression and Brazilian test, respectively. Dynamic mechanical analysis (DMA) was carried out by an apparatus (DMA 242C, Netzsch, Germany) with the sample size of $30\ mm \times 10\ mm \times 2\ mm$.

3 Results and Discussion

3.1 Effects of Crystal Quality on the Mechanical Performance

Improved crystal quality by recrystallization is useful to reduce the sensitivity of energetic materials, but smooth surface will also cause weak interfacial bonding. To investigate the effects of crystal quality on the mechanical performance, raw LLM-105, recrystallized sample with high crystal quality before (LLM-105-H) and after acid treatment (LLM-105-HA) were adopted.

Figure 1 shows typical SEM images for these three LLM-105 crystals and PBXs. The morphology and grain size of the raw LLM-105 are poor proportioned with some twinned crystals (Figure 1a). After solvent/anti-solvent recrystallization, both the morphology and size distribution become uniform. More specifically, the LLM-105-H displays regular polyhedron structure, smooth surface with the size of about 80 μm (Figure 1b). Obviously, it is difficult for the LLM-105-H based PBX to form molding powders at millimeter scale as compared with the raw LLM-105, as shown in Figure 1e and 1 h, attributing to the poor interaction between the smooth surface and the polymer binder. For the LLM-105-H based PBX, loose crystals and negligible coating effects were observed, and few binders were connected between the LLM-105 particles, which might cause poor moldability and safety performance. Therefore, it indicates that improving the crystal quality of explosives by recrystallization might be unfavorable for the mechanical strength due to the absence of anchor points on the smooth surface. To verify this deduction, recrystallized LLM-105 after acid treatment (LLM-105-HA) was prepared and tested (Figure 1c). Since the surface of LLM-105-HA became rough by the etching of HNO_3, molding powders could be successfully prepared, which was similar to the raw LLM-105 based PBX. Based on these observations, it is indicated that acid treat-

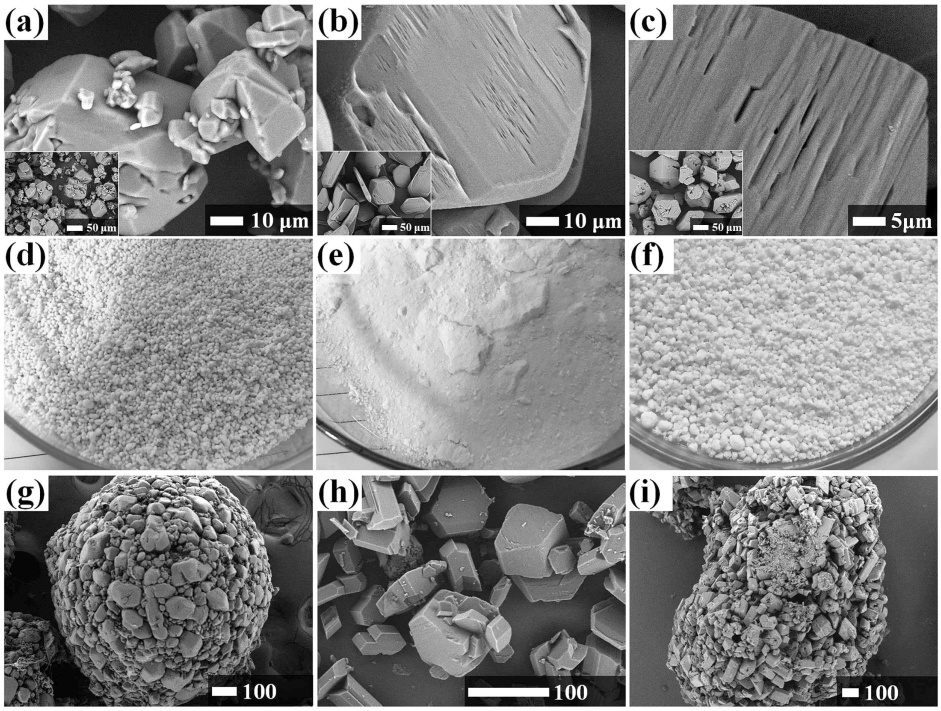

Figure 1. SEM images of raw LLM-105 (a), LLM-105-H (b), LLM-105-HA (c) (corresponding images with low magnification are inserted), optical photos and SEM images of raw LLM-105 (d, g), LLM-105-H (e, h), LLM-105-HA (f, i) based PBXs.

ment on high-quality explosive crystals can help to create extra anchor points on the surface with the high-quality inside structure maintained, facilitating to gain both nice mechanical and safety performances. Herein, not only the disadvantage of high-quality energetic crystal was revealed, how to solve this problem was also raised and proved.

Before the mechanical tests, the moldability of different LLM-105 based PBXs was evaluated, and the relative density of the Φ20 mm × 20 mm and Φ20 mm × 6 mm specimens was shown in Figure 2a. Clearly, the raw LLM-105 based PBX (Φ20 mm × 20 mm specimen) exhibited high relative density as 97.8%, while the value for LLM-105-H was decreased to 96.2%, indicating poor moldability for the high-quality crystals. For the LLM-105-HA sample after acid treatment, the relative density was increased to 97.3%, which was close to that of raw LLM-105. Similar results were found for the Φ20 mm × 6 mm samples. The reason for this phenomenon is that more polymer binder was coated around the surface of LLM-105-HA as compared with the high-quality crystals, corresponding to the SEM results in Figure 1.

Figure 2b and 2c shows the compressive and tensile curves of different LLM-105 based PBXs. Similar compressive strength was found for the three kinds of LLM-105, but the compressive strain was slightly decreased for high-quality crystals of LLM-105-H and LLM-105-HA. Tensile results were displayed by Brazilian disc tests, it was seen that the tensile strength was dramatically decreased from 3.90 MPa to 1.32 MPa as the crystal quality was improved by recrystallization, probably due to the weak bonding effect on the interface. Fortunately, the tensile strength could be recovered to 3.53 MPa after acid treatment, contributing to the anchor points created on the crystal surface.

Additionally, creep resistance behaviour represents the dimensional stability of PBXs under long-term force loading, and it is of great importance for the application of explosive in weapon systems. Herein, the three-point bending creep performance by DMA tests was exhibited in Figure 2d. The raw LLM-105 based PBX showed a steady creep with the strain of 3.23×10^{-4} under 2 MPa at 60 °C after 3600s, when the stress was increased to 4 MPa, the specimen tested was quickly fractured after 420s. For the LLM-105-H based PBX, the creep resistance became worse as expected, due to the weak bonding of fluoropolymer F2314. Specifically, the steady creep was only found under 1 MPa, with the strain 8.18×10^{-4} after 3600s. When the stress was increased to 2 MPa, the cylinder was fractured at 375 s. As to the LLM-105-HA sample, the creep resistance could be visibly improved. Steady creep was found even the stress was increased to 4 MPa, and the creep strain was 6.20×10^{-4} after 3600s. It was proved that the rough surface generated by acid treatment could evidently improve the creep resistance performance under the same condition.

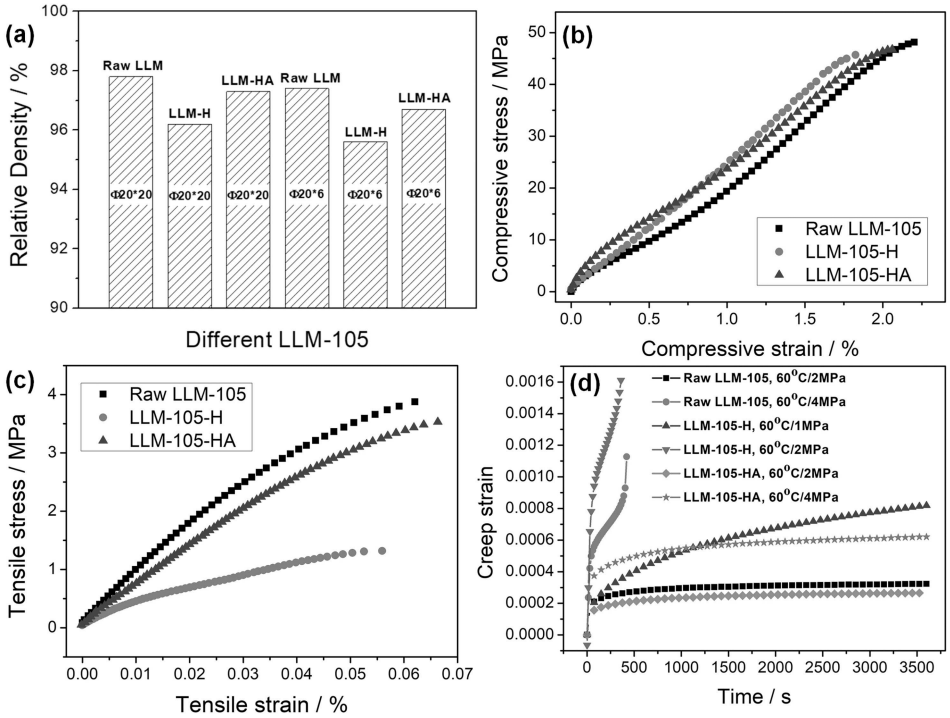

Figure 2. Mechanical performances of raw LLM-105, LLM-105-H and LLM-105-HA based PBXs: relative molding density (a); compressive (b), tensile (c) and creep (d) strain curves.

3.2 Effects of Spherical Morphology on the Mechanical Performance

Generally, spherical morphology of explosive was considered to be profitable to enhance the charge density and reduce the sensitivity, due to the reduction of crystal edges and corners. Recently, highly spherical LLM-105 was successfully fabricated with the assistance of surfactant additives [19]. In order to test the application performance of this novel products, the mechanical performance of their PBXs was studied in this work, including two kinds of spherical LLM-105 samples with smooth (LLM-105-SS) and rough (LLM-105-SR) surface, as shown in Figure 3a and 3b. Both these two spherical LLM-105 showed well proportioned particle distribution as displayed in the images with low magnification, with the average size of about 60 μm. For the LLM-105-SR sample, apparent burrs were observed on the crystal surface, as compared with LLM-105-SS.

For the PBXs, molding powders at millimeter scale were obtained using both these two spherical samples (Figure 3c and 3d). However, the LLM-105-SS based PBX showed more independent crystals, small amount of polymer binder was found, while the molding powders of LLM-105-SR exhibited more compact structure, indicating a better coating effect by the polymer matrix.

Contact angle tests were performed to further study the interface interaction between LLM-105 crystals and polymer matrix. F2314/ethyl acetate solution with concentration of 6 wt% was selected and dropped on the cylinders of different LLM-105, the results are shown in Figure 4. For the raw LLM-105, the contact angle was 25.9°, while the value was increased to 46.5° for the high-quality sample LLM-105-H, indicating a poor spreading and interaction on the surface. After acid treatment, the contact angle was 27.1°, close to that of raw LLM-105, implying a recovered interaction caused by rough surface. In addition, the angle value for

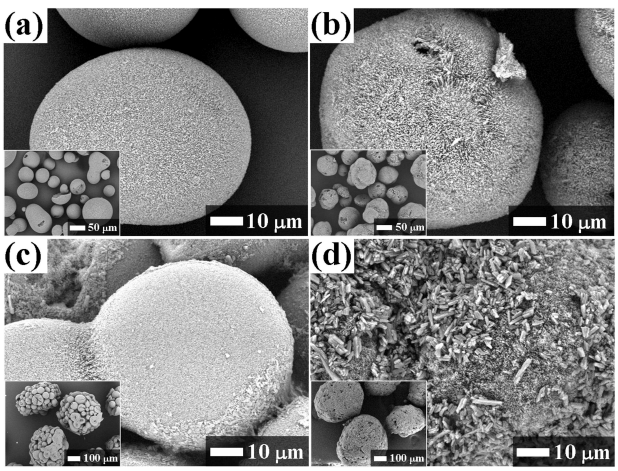

Figure 3. SEM images of LLM-105-SS (a), LLM-105-SR (b), LLM-105-SS based PBX (c) and LLM-105-SR based PBX (d), corresponding images with low magnification are inserted.

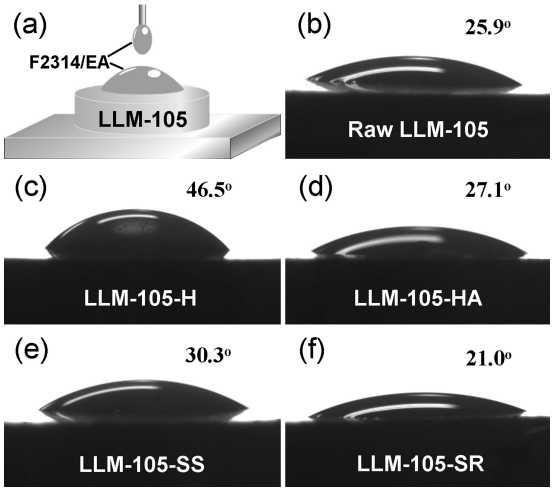

Figure 4. Static contact angle results by dropping F2314/ ethyl acetate solution on cylinders of different LLM-105 crystals: sketch (a), results (b-f).

LLM-105-SS and LLM-105-SR was 30.3° and 21.0°, respectively. LLM-105-SR showed the best wettability and might be beneficial for surface coating. In a word, the contact angle results were corresponding well with the SEM observations.

As mentioned above, rough surface and nice coating structure can result positive effects on the molding density and mechanical strength of PBXs. Therefore, cylinder samples were prepared, and the mechanical properties were studied, the results are shown in Figure 5. Reasonably, the relative molding density of LLM-105-SS based PBX was only 93.0 %, which was much lower than that of raw LLM-105 (97.8 %), ascribing to the poor coating structure. As for the LLM-105-SR, nice moldability was returned, providing a relative density of 97.1 % close to the polyhedral raw sample. Formability property for cylinders with different size showed the same trend (Figure 5a). Then, static mechanical tests were carried out, the compression and Brazilian test results are presented in Figure 5b and Figure 5c, respectively. For the compression test, the LLM-105-SS and raw LLM-105 based PBXs showed similar curves, both the compressive strength (48.0 MPa) and strain values were approximately equal. However, the compressive strength of LLM-105-SR was noticeably improved to 77.8 MPa, displaying a 62.5 % increase. Such fairly high compressive strength is not commonly observed for current pressed PBXs [6–8, 20], indicating that the LLM-105-SR with regular spherical morphology and rough surface can result outstanding compressive behaviors. As for the tensile tests, it is gratifying to note that both the LLM-105-SS and LLM-105-SR witnessed an evident increase of the tensile strength and strain. Specifically, the tensile strength of LLM-105-SS based PBX was 5.29 MPa, which is 35.6 % higher than that of raw LLM-105 (3.90 MPa). In addition, the tensile strain of LLM-105-SS also

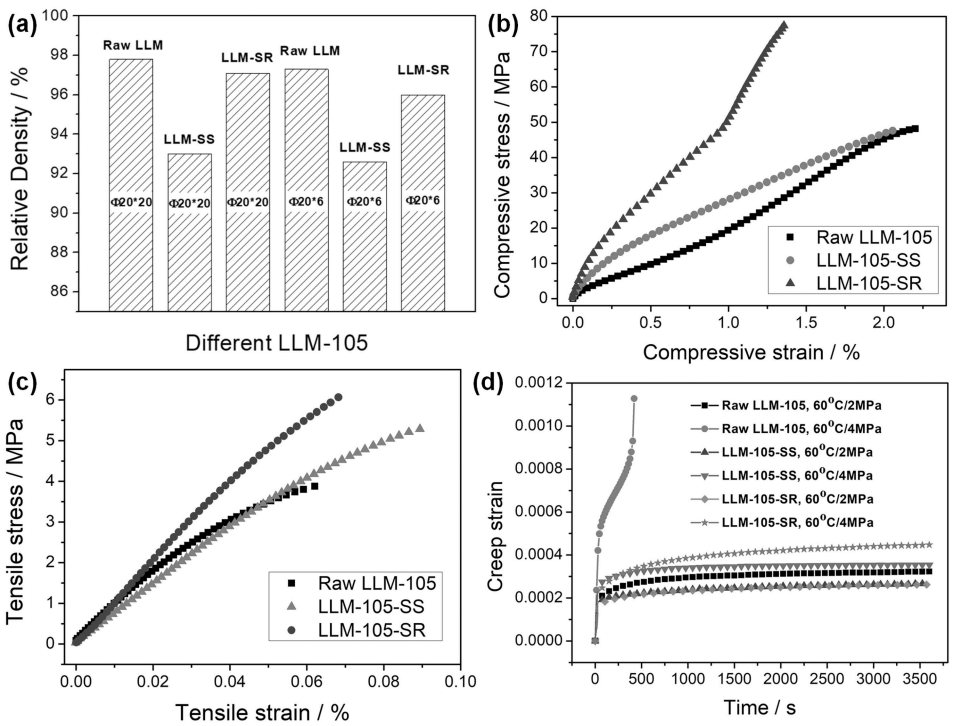

Figure 5. Mechanical performances of raw LLM-105, LLM-105-SS and LLM-105-SR based PBXs: relative molding density (a); compressive (b), tensile (c) and creep (d) strain curves.

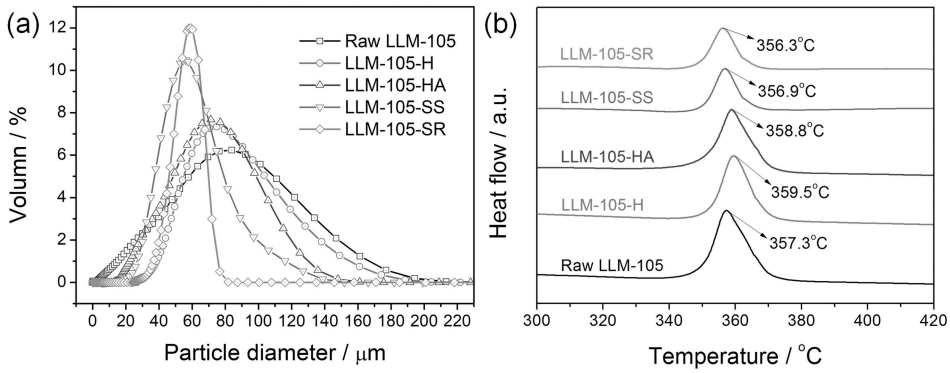

Figure 6. Particle size distribution (a) and DSC (b) results of the five LLM-105 materials used in this work.

showed an increase of 36.4%, demonstrating a promising mechanical performance.

It is reasonable that rough surface can further improve the interfacial interaction because more anchor points are exposed to the polymer binder. Just as expected, the tensile strength of LLM-105-SR based PBX reached 6.40 MPa, 64.1% higher than that of raw LLM-105. Combining with the SEM results in Figure 3, it is confirmed that rough surface of explosive crystals can remarkably improve the mechanical strength of PBXs, providing favorable advantages for their practical applications.

Furthermore, DMA test was performed to determine the creep resistance performance of spherical LLM-105 based PBXs, the creep curves are presented in Figure 5d. Apparently, the creep resistance was largely improved for both the two spherical samples. The creep characteristic of raw LLM-105 based PBX was already discussed in Figure 2d, showing a steady creep under 2 MPa and the cylinder was fractured under 4 MPa. However, no fracture was found for spherical LLM-105 based composites under the same condition. For the two spherical samples, the creep behavior under 2 MPa was similar, the creep strain of LLM-105-SS and LLM-105-SR based PBXs was 2.63×10^{-4} and 2.67×10^{-4}, respectively, which is about 18% lower than that of raw LLM-105. As the stress was increased to 4 MPa, typical steady creep curves were still maintained, and LLM-105-SR with rough surface exhibited a lower strain (3.54×10^{-4}) than that of LLM-105-SS (4.47×10^{-4}) after 3600s. The creep resistance results were in good consistence with the other mechanical tests, indicating the spherical morphology and rough surface of LLM-105 crystals can be beneficial to the mechanical performance and dimensional stability under external environmental stimulations.

It is known that the particle size and distribution have significant influence on the mechanical performance of PBX materials [21]. In order to confirm that the improvement of mechanical performances for LLM-105 based PBXs was contributed to the roughed interface rather than particle size distribution, the PSD test was conducted, and the results are shown in Figure 6a. Clearly, the particle size distribution of LLM-105 samples before and after surface treatment displayed similar curves. Specifically, the recrystallized samples before (LLM-105-H) and after acid treatment (LLM-105-HA) had the average particle size of 74.7 μm and 71.4 μm, respectively. For the spherical samples with smooth and rough surface, LLM-105-SS and LLM-105-SR showed the average particle size of 55.8 μm and 58.7 μm, respectively. Therefore, it can be confirmed that such surface treatment had negligible influence on the PSD of samples, and the improvement of mechanical performance are resulted from the surface roughness.

Furthermore, as small amounts of acid impurities can lead to significant decrease in thermal sensitivity for energetic materials, excessive hot water was used to wash the acid treated samples carefully for several times, followed by heated vacuum drying at 100 °C for 48 h to confirm that the HNO_3 had been removed thoroughly. To study the possible effect of acid treatment on the thermal stability of LLM-105, the thermal decomposition curves of different LLM-105 samples in this work were obtained by DSC, as shown in Figure 6b. Obviously, the LLM-105 samples before and after acid treatment showed similar curves and decomposition peaks. Specifically, LLM-105-H and LLM-105-HA showed the decomposition temperature of 359.5 °C and 358.8 °C, respectively. The difference can be ignored, indicating that acid treatment followed by careful washing will not cause negative effects for the thermal stability of LLM-105. As for the spherical samples, LLM-105-SS and LLM-105-SR exhibited the decomposition temperature of 356.9 °C and 356.3 °C, respectively, indicating that the surface roughness has no effect on the thermal decomposition.

4 Conclusions

LLM-105 based PBXs with different crystal quality, morphology and surface roughness were prepared, the granulating, moldability, compressive, tensile and creep performances were studied and compared. Crystals with high quality after recrystallization had low density and poor tensile strength,

which could be visibly improved by acid treatment. Spherical morphology was profitable for both the mechanical strength and creep resistance, especially for the samples with rough surface, which was confirmed in groups of experiments. This work not only points out the mechanical problems caused by increased crystal quality of explosives in spite of improved safety, but also provides the strategy how to effectively improve the mechanical performance. Surface roughness had critical influence on the mechanical performance because the anchor points created can be helpful for the bonding between energetic crystals and polymer matrix, thus increasing the interfacial strength and dimensional stability. It is worth noting that LLM-105-SR, the spherical and rough sample, exhibited fairly high compressive and tensile strength of 78 MPa and 6.4 MPa, respectively, both of which was more than 60% high than that of normal LLM-105 with polyhedral structure. It was confirmed by PSD test that the improvement of mechanical performances for LLM-105 based PBXs was contributed to the roughed interface rather than particle size distribution, and acid treatment followed by careful washing will not cause negative effects for the thermal stability of LLM-105. The results and conclusions based on the present work can not only broaden the application value of LLM-105 in military use and national defense, but also provide strong generality to improve the mechanical performance for the other commonly used high explosives, such as HMX, RDX and TATB.

Acknowledgements

This work was supported by the National Natural Science Foundation of China (NSFC 51703211, 21875232, 11702267, 21875229).

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Manuscript received: March 30, 2019
Revised manuscript received: July 21, 2019
Version of record online: August 14, 2019

