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Study on the relation between microstructural change and compressive creep stress of a PBX substitute material

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Abstract: A polymer-bonded explosive, also called PBX or plastic-bonded explosive, is an explosive material in which explosive powder is bound together in a matrix using small quantities (typically 5%–10% by weight) of a synthetic polymer. A PBX substitute material was made from sugar granules and polymer binder. Its compressive creep properties were investigated at room temperature. The creep deformation was found to depend strongly on the applied stress amplitude. Under an applied stress near the strength, creep deformation developed and reached the final rupture very quickly. A power law relationship, \( \varepsilon = 4.14 \times 10^{-8} \sigma^{0.25} \), was established between steady creep rate and applied stress. Microscopic observations show that the damage mechanism processes include mainly the intergranular and transgranular fractures, binder fracture, and peeling. Both porosity and granule size decrease almost linearly with increasing applied stress.

Keywords: compressive creep; creep deformation; damage mechanism; microscopic observation; PBX substitute material.

1 Introduction

During the long-term storage and transport period for polymer-bonded explosives (PBX), creep deformation may occur, this deformation will degrade the mechanical properties of PBX. These mechanical properties could in turn affect the explosive performance to some degree. Therefore, the security and reliability of PBX requires good properties against creep deformation.

A PBX is composed of particles and binder. Due to the predominant content of particles, which could be more than 90%, initial defects such as pores and tiny cracks are often left inside the material, resulting in different failure mechanisms [1–3]. The particle size is generally several dozen micrometers on average, although nanoparticle explosives are of increasing interest and the corresponding study on creep properties could refer to nanocomposites [4–6]. Experimentally, diverse methods have been developed to capture the creep deformation and analyze the damage and failure mechanisms. High-resolution moiré interferometry was applied to study the microscopic failure mechanisms of PBX [7]. Real-time microscopic examinations were undertaken for deformation and failure of PBX [8, 9] by using an environmental scanning electron microscope (SEM) and a SEM equipped with a loading stage, respectively. Both tensile and compressive creep tests were carried out to investigate the deformation behavior under different stress amplitudes and temperatures [10, 11]. In addition, the strain rate is also a key factor to be taken into account when studying the creep properties of PBX [12]. The evolution of initial defects such as voids and the effects of type and molecular structure of binders on creep deformation were studied under different stress amplitudes and temperatures [13–16]. Theoretically, the Burgers-Kelvin viscoelastic constitutive model, refined recovery-creep model, and one-dimensional viscoelasticity damage model, among others, were proposed and used to predict the rheological behavior and damage of PBX [17–20].

Previously, Funk et al. [21] found that PBX 9501 sugar mock closely resembled the behavior of the PBX 9501 explosive. Furthermore, to take safety issues into account and to avoid possible explosions, substitute materials for PBX are often used to carry out laboratory studies [22, 23]. In this work, a study on the compressive creep behavior of a PBX substitute material is undertaken. The emphasis is placed on the relationship between creep properties and
microstructural evolution, which includes not only the damage phenomena such as interfacial debonding and cracking but also the variation of granule size and porosity. Therefore, it provides new aspects into creep deformation and damage mechanisms.

2 Materials and methods

2.1 Material preparation

The studied material, referred to as a PBX substitute material, is composed of sugar granules (Chinese Medicine Reagent Company, Shanghai, China) with sizes ranging from 20 μm to 300 μm and a specific SWJ-9301 glue binder. The two constituents were weighted in a proportion of 9:1, and then mixed completely by mechanical stirring. Afterward, the mixture was poured into a special stainless steel mold and slightly pressed on top and then kept at 70°C for 5 h. Finally, cylindrical samples with dimensions of 25 mm × 25 mm (height × diameter) were obtained for creep tests.

2.2 Compressive creep tests

The compressive creep tests were conducted at room temperature with a creep testing machine (RDJ30; Mechanical Science Research Institute Co., Ltd, Changchun, China). The stress amplitudes were kept constant at 4.00, 4.50, 5.00, 5.25, and 5.50 MPa, respectively. The SEM (S-4800, Hitachi, Tokyo, Japan) was used to observe the damage and failure mechanisms. The sizes of the sugar granules before and after the tests were measured with a laser particle size analyzer (model LS-POP(6); Changchun Institute of Machinery Co., Ltd., Changchun, China).

3 Results and discussion

3.1 Mechanical behavior under compressive creep tests

Figure 1B shows the monotonic compressive stress-strain curve of the studied material, in which the stress amplitudes for creep tests were chosen from 4.00 to 5.50 MPa. The choice for the stress amplitude is based on the following considerations: the lowest stress value of 4.00 MPa is still in the linear section of the stress-strain relation, whereas 4.50 MPa is just at the limit of linearity (elastic limit). The highest value of 5.50 MPa is approximately 95% of the compressive strength, whereas the two stress amplitudes of 5.00 and 5.25 MPa are situated between the elastic limit and compressive strength. In this way, we can completely study the compressive creep properties of materials ranging from elastic to plastic deformation. Figure 1A shows the appearance of samples under monotonic compression. It can be seen that several deep cracks appear along the vertical direction accompanied by some small cracks on the side face of the sample.

The creep strain versus time curves under different applied stresses are given in Figure 2. The creep tests were stopped at 25 h, and thus the tertiary stage of creep deformation was not obtained. Primarily, the strain increases quickly with an almost instantaneous linear trend, which

Figure 1: The process and result of monotonic compression test: (A) Appearance of the sample under monotonic compression and (B) the corresponding stress-strain curve of the studied material.
is termed an elastic response. Then, creep strain occurs and the strain rate gradually decreases until the steady state. As expected, the strain rate remains constant in the second steady stage, and the strain amplitude gradually increases with increasing stress amplitude. However, a jump in the strain amplitude was noted when stress increased from 4.50 to 5.00 MPa. According to the monotonic stress-strain curve shown in Figure 1B, a transition from elastic to plastic deformation happened at 4.50 MPa. At 5.00 MPa, plastic deformation had already been initiated, and thus resulted in a jump in strain. From 4.00 to 4.50 MPa, the deformation is always elastic, and it remains plastic when the stress increases from 5.00 to 5.25 MPa, and thus the strain increment is progressive. The average differences of strain amplitude are given in Table 1 to clarify quantitatively the dependence of creep deformation on the stress amplitude. To show the dependence of primary creep deformation on stress amplitude, the amplified curves of the primary stage are also shown in Figure 2 (inset). As it can be seen, the instantaneous elastic deformation began first, and came to an end faster and at a higher peak with increasing stress amplitude.

Figure 3 displays the creep curve under a stress of 5.50 MPa, which is almost 95% of the ultimate strength. Under such high stress, the sample is unstable and is very quickly damaged, and the entire three-stage creep deformation process is noticed. By adopting a routine analysis method for creep deformation, the primary stage was extended whereas the secondary stage was shortened and quickly followed by the tertiary stage until final failure. This three-stage creep deformation can be defined as the “apparent” creep deformation due to the significant differences compared with those in Figure 2. During the “apparent” creep deformation process, high creep stress causes very early creation and accelerated development of damage in the material. Therefore, the creep deformation increased rapidly and the final failure occurred within a short time.

Under applied stress, $\sigma$, the creep rate of the steady stage can be expressed in general by the equation $\dot{\epsilon} = A\sigma^n$. The coefficient $A$ represents the creep factor and $n$ refers to the creep stress exponent. The curve of the steady creep rate as a function of stress is shown in Figure 4. The linearity of the data fits very well with the linear correlation coefficient of 0.94, providing a creep stress exponent of 2.50 and a creep factor of $4.14 \times 10^{-8}$. The creep stress

| Table 1: Average strain difference in the steady creep stage. |
|-----------------|-----------------|-----------------|-----------------|
| Stress range (MPa) | 4.00–4.50 | 4.50–5.00 | 5.00–5.25 |
| Strain difference (%) | 0.17 | 0.83 | 0.11 |

Figure 2: The creep strain-time curves under different stresses.

Figure 3: The “apparent” creep curve at less than 5.50 MPa.

Figure 4: Relationship between steady creep rate and applied stress.
exponent of 2.50 is relatively small considering the long failure time and service life.

3.2 Porosity evolution

Figure 5 shows SEM micrographs of cryo-fractured cross-sections of samples before and after the creep tests. As it can be seen from Figure 5A, there are initial pores in the as-prepared samples; their existence comes from the sugar granules being incompletely coated by binder and from the residual air left inside after fabrication. They are distributed in the encountering corners and sides of granules with irregular shapes. During creep deformation, the pores are compressed more and more with time, which contributes greatly to the macroscopic strain. This process is similar to post-curing, which makes the sample more compact. After creep deformation, the pore size is evidently reduced, as shown in Figure 5B–D. The cryo-fractured surfaces are rougher than the original one. As the creep stress increased further, the morphology of the cryo-fractured surfaces (Figure 5E,F) became much more different from the original one (Figure 5A). They are uneven and some small granules coming from the breakage of larger ones can be clearly seen. In this case, the rupture of large granules happened due to mutual squeezing among them under high stress. Some cracks were even created as indicated by the arrow in Figure 5F.

Based on SEM micrographs and by using Photoshop software, the porosity was measured and plotted against the applied stress as shown in Figure 6. The porosity decreases almost linearly with the increase of applied stress. Changes at creep stresses of <4.00 MPa are unknown and are thus presented with a discontinuous line. The porosity is as high as 3.46% for the original sample, and this was reduced to 0.94% after the creep tests at an applied stress of 5.50 MPa. This phenomenon is different from that of tension, in which the porosity

Figure 5: The microscopic observation of porosity evolution: (A) stress of 0.00 MPa; (B) stress of 4.00 MPa; (C) stress of 4.50 MPa; (D) stress of 5.00 MPa; (E) stress of 5.25 MPa; (F) stress of 5.50 MPa.
increases with stress. The reduction of porosity comes from two sources: the evolution of the original pores and the formation of new cracks. Under compressive stress, the pores were increasingly pressed and their sizes were reduced progressively with creep time. At the same time, new cracks were created from the fracture of large granules and debonding at the granule and binder interface, bringing an increase of porosity. Because the pores are much larger and more abundant than cracks, the reduction in pore volume predominated, leading to the total reduction of porosity.

Figure 7 shows enlarged SEM micrographs. In Figure 7A, both large and small pores in the as-prepared sample are seen at the corners of some granules, as indicated by the arrows. These are approximately a few micrometers to hundreds of micrometers in size. It can be seen from Figure 7B that small pores may exist at the bottom of large ones, forming several arrangement levels, as indicated by the arrows. This reflects the difficulty of preparing no-pore samples due to the high content of granules. With the increasing stress of 4.5 MPa, i.e. the limit of elasticity, intergranular fractures occurred (arrow, Figure 7C), signifying the appearance of new damage.

Figure 6: Decrease of porosity with applied stress.

Figure 7: SEM micrographs showing creep damage mechanisms: (A) stress of 0.00 MPa; (B) stress of 4.00 MPa; (C) stress of 4.50 MPa; (D) stress of 5.00 MPa; (E) stress of 5.25 MPa; (F) stress of 5.50 MPa.
mechanisms. With continuous creep deformation, the relative sliding of granules resulted in debonding along the boundary of the granules. As the applied stress continued to increase to 5.0 MPa, the breakage of granules or transgranular fracture occurred (arrow, Figure 7D). This new mechanism of damage comes from the stress concentration at the boundary of the pore, as well as the high contact stress between two neighbor granules. When the applied stress further increased to 5.25 MPa, the binder fracture and peeling, as well as feasible pore deformation, were observed as indicated by the arrow in Figure 7E. Under a very high stress of 5.50 MPa, the cracks were largely open and some pores were filled with small pieces of broken granules, as indicated by the arrows in Figure 7F. This signifies that deformation and stress concentration were so great that damage and failure had occurred quickly. The previous observations outlined the damage mechanisms under creep tests, i.e. a sequential occurrence of intergranular failure, binder fracture and peeling, cracking, and transgranular failure with increasing stress. It should be indicated that some mechanisms may also occur simultaneously.

3.3 Reduction of granule size

It is shown in Figure 7 that some granules were broken during creep deformation, and thus their sizes were reduced. To obtain a quantitative variation of granule size, its differential distribution was measured after creep tests as shown in Figure 8. It is clear that the initial granule size had a broad distribution. After the creep tests, it became narrower and narrower with increasing creep stresses. This signifies that the larger granules tend to be broken into smaller ones whereas the smaller ones seem to be more resistant. The breakage of granules became one of the main damage mechanisms of creep deformation in this case. It comes from the squeezing of closely packed granules under compressive load, as well as the stress concentration at the corner of pores. The average granule size as a function of creep stress is given in Figure 9. It is noted that the granule size decreased almost linearly with creep stress, from an initial value of 46 μm to the minimum of 31 μm. This also indicates that the internal damage was gradually aggravated with the increasing creep stress.

4 Conclusions

In this work, a PBX substitute material was made of sugar granules and polymer binder, its creep behavior was studied experimentally under five stress amplitudes. It was found that a remarkable jump in the creep strain happened when the applied stress was increased from the elastic limit to the plastic flow stress. The steady creep rate gradually increased with the increasing stress, between them, a power law relationship \( \dot{\varepsilon} = 4.14 \times 10^{-6} \sigma^{0.25} \) exists. A low value of the creep stress exponent means that the material is not very sensitive to the creep stress level. The microscopic observation and measurements show that both porosity and granule size decreased almost linearly with the creep stress amplitude. Additionally, intergranular and transgranular fractures, as well as binder fracture and peeling, are the main damage mechanisms. Under very high amplitudes of creep stress, the creep behavior shows an "apparent" three-stage deformation that is much shorter than traditional creep deformation.
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References