Microstructural simulation of initial particle-matrix interfacial defect in HTPB solid propellants

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Abstract: Impregnation capability at particle-matrix interfacial deteriorates during the preparation of composite solid propellant due to manufacturing and environmental factors, which leads to the initial defects in the HTPB solid propellant. In the present work, the effects of initial particle-matrix interfacial defects on the debonding, nucleation, and crack propagation of the composite solid propellant is investigated experimentally and numerically. A random packing model is developed based on the molecular dynamics method by embedding AP particles in the HTPB propellant. The strength of particle-matrix interface obeys Weibull function. Moreover, a zero-thickness cohesive element is used to simulate the particle-matrix interfacial debonding and matrix rupture. According to the simulation and experiment results, the initial interface defects makes a marked effect on the initial modulus, limit strength and elongation at fracture of the composite solid propellant. The initial modulus, limit strength and elongation at fracture of the composite solid propellant decrease with the increase of the initial interface defects.

1. Introduction
Solid propellants are extensively used in rocketry systems due to their high energy density, safety and long shelf life. Usually, the solid propellants are composed of ammonium perchlorate (AP) particles, metal powder and matrix which lead the mechanical properties of the solid propellants depend on the mechanical properties of the particles, the matrix and the particle/matrix interface [1]. Van Ramshorst et al. [2] studied the relationship between the microscopic morphology and macroscopic deformation and failure of the solid propellants based on scanning electron microscope (SEM), and found that the particle/matrix interface debonding and cracks grow along the particle/matrix interface under loading. Therefore, it is necessary to analyze the microscopic damage of the solid propellants.

Matouš and Geubelle [3] simulate the particle/matrix interface debonding process with small deformation theory. They proved that the interface debonding lead a non-homogeneous stress. H. Arora [4] developed 2D circles, 2D polygons and 3D spheres to compare the effects of the particle geometries on the damage evolution of particulate reinforced materials. Li Gaochun [5] found that the strength of the interface is the key factors to enhance the mechanical properties of the solid propellants. To describe the viscoelastic mechanical properties of the solid propellants, Cui [6] proposed a novel time-dependent cohesive zone model (CZM) based on the Maxwell box to simulate relaxation responses. However, above studies have ignored the performance defects of the solid propellants in production which causing errors in the simulate result and experimental result. Balzer [7] observed the particle/matrix interface by using SEM, and found that there were so many micro holes in the solid propellant, especially in the particle/matrix interface. This is because the material properties of the particles and the matrix are different during the curing process of the solid propellant, which causing gaps between particle and...
matrix, resulting in the existence of randomly distributed initial interface defects in the solid propellants. The initial interface defect will change the failure path of the solid propellants which accelerated damage. Wang [8] found that the effect of the initial defects on the macro-mechanical properties of the particulate reinforced materials cannot be ignored.

The present research investigated the mechanical properties and fracture behavior of the HTPB propellant through microstructure-based numerical simulations. The strength density function of the particle/matrix interface obeys Weibull distribution to analyze the influence of initial defect on the mechanical properties of the HTPB propellants.

2. Microstructure-based numerical model

2.1. RVE model generation

A two-dimensional (2D) representative volume element (RVE) model is developed by using Fortran language, as shown in Fig. 1. It is considered as effective when the RVE model size is 3 times the maximum particle size [9]. Therefore, the 500 μm × 500 μm edge-sized RVE model is selected.

![Figure 1 Microscopic particle filled model](image)

2.2. Cohesive zone model

In order to capture the particle/matrix interfacial debonding and matrix rupture, the zero-thickness cohesive elements are embedded in the packing model by using Python scripting language (Fig. 2). Fig. 2(a) shows the initial mesh of the particle and the matrix and the mesh type is CPE4R. Fig. 2(b) shows the cohesive element of the interface, the matrix and the mesh type is COH2D4.

![Figure 2 Embedded cohesive element diagram](image)

The cohesive zone model was first proposed by Dugdale [10] and Barenblatt [11]. A fracture process area at the crack tip is proposed which is determined by the normal traction and tangential traction. The cohesive zone models are divided into bilinear, trapezoidal, and exponential, etc. Every cohesive zone model has its scope of application. According to the damage characteristics of the HTPB propellant, the exponential cohesive zone model is used to describe the damage evolution process of the particle-matrix interface, and the trapezoidal cohesive zone model is used to describe the rupture of the matrix.

The exponential cohesive zone model and trapezoidal cohesive zone model have been obtained by using Fortran language. The fracture energy \( \phi(t) \) of the exponential cohesive zone model is:
\[
\phi(t) = \phi_n + \phi_r \exp\left(-\frac{t_n}{\delta_n}\right)\left[1 - r + \frac{t_n}{\delta_n}\left(1 - q + \frac{r - q}{r - 1} \exp\left(-\frac{t_r}{\delta_r}\right)\right)\right],
\]
(1)

where \(\delta_n\) and \(\delta_r\) are the characteristic length of the normal and tangential. The cohesive element normal traction and tangential traction are obtained by the following equation.

\[
T_n = -\frac{\phi_n}{\delta_n} \exp\left(-\frac{t_n}{\delta_n}\right) \left[\frac{t_n}{\delta_n} \exp\left(-\frac{t_r}{\delta_r}\right) + \frac{1 - q}{r - 1} \left[1 - \exp\left(-\frac{t_r}{\delta_r}\right)\right]\right],
\]
(2)

\[
T_t = -2 \frac{\phi_n}{\delta_t} \frac{t_t}{\delta_t} \left[q + \frac{r - q}{r - 1} \frac{t_n}{\delta_n}\right] \exp\left(-\frac{t_n}{\delta_n} - \frac{t_t}{\delta_t}\right),
\]
(3)

The fracture energy \(\phi(t)\) of the trapezoidal cohesive zone model is:

\[
\phi(t) = \frac{1}{2} \sigma_{\text{max}} (\delta_j + \delta_i - \delta_f)
\]
(4)

The cohesive element normal traction and tangential traction are obtained by the following equation.

\[
T_{n,t} = \left\{ \begin{array}{ll}
\frac{\sigma_{\text{max}}}{\delta_i} & \delta < \delta_i \\
\frac{\sigma_{\text{max}}}{\delta_f - \delta_i} (\delta_j - \delta_i) & \delta_i \leq \delta \leq \delta_2 \\
\frac{\sigma_{\text{max}}}{\delta_f - \delta_2} (\delta_f - \delta_2) & \delta_2 \leq \delta \leq \delta_f
\end{array} \right.
\]
(5)

During the mixing and curing of the HTPB solid propellant, the bonding of the particle-matrix interface will decrease, and voids will be generated between the particle and the matrix interface, as shown in Fig. 3. The initial defects causing the instability mechanical propose of the HTPB propellant. Therefore, it is necessary to consider the initial defects between the particle and the matrix interface when predicting the mechanical properties of the propellant. In this paper, the density function of the bonding strength between the particle and the matrix is set as the Weibull function. The mesoscopic numerical model with initial defects is developed. The Weibull function is:

\[
f(x) = \frac{m}{\lambda} \left(\frac{x}{\lambda}\right)^{m-1} \exp\left[-\left(\frac{x}{\lambda}\right)^m\right],
\]
(6)

where \(x\) represents a random function. \(m\) is a shape parameter. \(\lambda\) is a proportional parameter. The Weibull function with different shape parameters \(m\) is shown in Fig. 4.

![Figure 3 Initial defect of solid propellant](image-url)
3. Results and discussion

3.1. Damage behavior

Under uniaxial tension, the deformation process of the packing model was divided into four stages-Ⅰ, Ⅱ, Ⅲ, and Ⅳ, as shown in Fig. 5. In stage Ⅰ, the particle and the matrix experienced elastic deformation and interface between the particle and matrix is bonded well. However, the rigidity of the particles and the matrix is different, causing the extremely uneven stress distribution within the model, and the particle stress is significantly higher than the matrix stress. In stage Ⅱ, due to the non-uniformity of the interfacial bonding strength distribution, the location where the particle and the matrix interface damage content is higher will be the first to debond. In stage Ⅲ, the deformation of the matrix increases, and the amount of separation displacement of the particles from the matrix increase, causing the particles to be exposed form the holes. In stage Ⅳ, as the strain continues to increase, the pores after debonding of the particles and the matrix converge, and the microcracks formed inside the matrix.

3.2. Influence of initial defects on HTPB solid propellant

The effect of initial defects on the mechanical property of the HTPB solid propellant is examined by varying the m values. With the increase of the m values, the mechanical property of the HTPB solid propellant changed significantly, as shown in Fig. 6. The initial modulus, limit strength and elongation increase with the increase of the m values. This is because the m value determines the distribution law of the bonding strength between the particle and the matrix. The smaller of the m value, the more dispersed the bonding strength, the more initial defects inside the RVE model. As a result, the interface between the particle and the matrix is more likely to form debonding damage. Moreover, the load transfer capacity of the interface decreases, resulting in a decrease in the overall strength of the model. Finally, the initial modulus, the limit strength and elongation decreases. On the contrary, the larger of the m value, the more concentrated the bonding strength distribution, and the higher the initial modulus, the limit strength and elongation obtained from the RVE model.

The initial defect models better explain the discreteness of the HTPB solid propellant experimental results. It also proved the randomness of the HTPB solid propellant fracture position. Therefore, the initial defect model is more coincident with the real situations.
4. Conclusion

A RVE model of HTPB solid propellant is developed to explore the influences of initial interfacial defect on the mechanical properties. The cohesive elements are presented to simulate the particle-matrix interfacial debonding and matrix rupture. It is found that the damage evolution of the HTPB solid propellant from the particle-matrix interfacial debonding to the matrix rupture. Moreover, the initial defect model also presents that the initial modulus, the limit strength and the elongation decreases with the increases of the initial interfacial defect.

Reference

[1] Francqueville, J., Diani, J., Gilormini, P., et al. (2021) Use of a micromechanical approach to understand the mechanical behavior of solid propellants[J]. Mech Mater, 153: 103656.
[2] Vanramshorst, M.C.J., Benedetto G.L., Duvalois, W., et al. (2016) Investigation of the Failure Mechanism of HTPB/AP/Al Propellant by In-situ Uniaxial Tensile Experimentation in SEM[J]. Propell, Explos, Pyrot, 41(4): 700-708.
[3] Matouš, K., Geubelle, P.H. (2006) Finite element formulation for modeling particle debonding in reinforced elastomers subjected to finite deformations[J]. Comput Method Appl M, 196(1-3): 620-633.
[4] Arora, H., Tarleton, E., Li-Mayer, J., et al. (2015) Modelling the damage and deformation process in a plastic bonded explosive microstructure under tension using the finite element method[J]. Comp Mater Sci, 110: 91-101.
[5] Li, G., Wang, Y., Jiang, A., et al. (2018) Micromechanical investigation of debonding processes in composite solid propellants[J]. Propell, Explos, Pyrot., 43(7): 642-649.
[6] Cui, H., Shen, Z., Li, H. (2018) A novel time dependent cohesive zone model for the debonding interface between solid propellant and insulation[J]. Meccanica, 53(14): 3527-3544.
[7] Balzer, J.E., Siviour, C.R., Walley, S.M., et al. (2004) Behaviour of ammonium perchlorate–based propellants and a polymer–bonded explosive under impact loading[J]. Proceedings of the Royal Society of London. Series A: Mathematical, Physical and Engineering Sciences, 460(2043): 781-806.
[8] Wang, J., Qiang, H., Wang, Z., et al. (2020) Numerical Study of Mechanical Properties of Composite Solid Propellant with Initial Defects[C]/Journal of Physics: Conference Series. IOP Publishing, 1634(1): 012146.

[9] Bernard, F., Kamali-Bernard, S., Prince, W. (2008) 3D multi-scale modelling of mechanical behaviour of sound and leached mortar[J]. Cement Concrete Res, 38(4): 449-458.

[10] Dugdale, D.S. (1960) Yielding of steel sheets containing slits[J]. J Mech Phys of Solids, 8(2): 100-104.

[11] Barenblatt, G. I. (1962) The mathematical theory of equilibrium cracks in brittle fracture[M]. Adv Appl Mech, 7: 55-129.