Numerical Study of Mechanical Properties of Composite Solid Propellant with Initial Defects

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Abstract. In order to study the effect of initial interface defects and initial void defects on the macro tensile mechanical properties of composite solid propellant, based on the Python script, the particle filling model of composite solid propellant with meso pores and interface defects was established in ABAQUS. Firstly, zero thickness cohesive elements were embedded at the adjacent elements of the matrix and the particle/matrix interface, and the traction separate law was used to simulate the fracture failure of composite solid propellant. Then, the influence of different shapes, porosity and interface defect ratio on the elastic modulus and tensile strength of composite solid propellant was studied. Accordingly, the relationship between the initial damage of the composite solid propellant and the porosity and interface defects was established. Moreover, the initial damage surface of the composite solid propellant was depicted. Results showed that the initial damage of polygon filling model is larger than that of circular particle filling model under the same initial defect condition; The elastic modulus of composite solid propellant decreases linearly with the increase of porosity and interface defect ratio; The tensile strength of composite solid propellant decreases linearly with the increase of interface defect ratio, and decreases logarithmically with the increase of porosity.

1. Introduction
Composite solid propellant is mainly composed of oxidant, metal fuel additive, organic binder, various functional additives and performance regulators. Its macro mechanical properties are closely related to the performance, structure, distribution, content and initial defects of each component. At present, the research on the mechanical properties of composite solid propellant is mainly carried out from three aspects: experimental study, theoretical analysis and numerical simulation. Due to the limitation of experimental conditions and costs, it is difficult to control the structure and content of internal defects of solid propellant quantitatively, and it is difficult to carry out the initial meso damage experimental research on the mechanical properties of solid propellant. At present, the theoretical models used to predict the macro mechanical properties of composite solid propellants have made many simplifications on the meso structure, so it is difficult to reveal the influence of internal defects on the mechanical properties of composite solid propellants. Therefore, the research on the relationship between meso mechanical properties and macro mechanical properties of composite solid propellants is mainly based on numerical simulation presently.

A lot of numerical studies have been carried out on the relationship between the meso-structure and the macro-mechanical properties of composite solid propellants. Knott \cite{1} established the meso particle filling model of composite solid propellant based on molecular dynamics method for the first time. Matous\cite{2} developed RVE (Representative Volume Element) model of solid propellant by using self-developed software, and he studied the performance of propellant under different loading conditions.
from the meso-damage mechanism. Based on the maximum tensile stress and Mohr Coulomb criterion, Kang [3] simulated the fracture process of polymer binder and explosive particles by the numerical manifold method taking the initial interfacial debonding, the initial holes in polymer binder and the initial microcracks in explosive particles into consideration. Feng [4] established a meso-model of composite solid propellant filled with circular particles by using molecular dynamics method, and he studied the effects of particle breakage, initial interfacial debonding and micropores on the mechanical properties of composite solid propellant through finite element simulation calculation, but the damage and fracture of binder matrix were not taken into account. The damage of composite solid propellant mainly includes particle/matrix interface debonding and matrix fracture. Considering both interfacial debonding and matrix damage, Zhao[5] simulated the complete evolution process of composite solid propellant failure by using global bilinear cohesion model. The process of crack initiation and propagation of composite solid propellant at mesoscopic scale was given, but the initial defects in the propellant were not taken into account in the calculation model.

In order to reveal the relationship between the initial defects and the macroscopic mechanical properties of composite solid propellant, two kinds of meso-particle filling models of composite solid propellant, circular and polygonal, were established by using stochastic continuous algorithm. The zero thickness cohesive elements was embedded at the adjacent elements of the matrix and the particle/matrix interface, and the mesoscopic numerical simulation of composite solid propellant was carried out. The tensile stress-strain curves of composite solid propellant from microcrack initiation to complete failure were obtained. Moreover, the influence of void and interface defects content on the tensile mechanical properties of composite solid propellant was studied, and the fitting equation between initial damage of composite solid propellant and porosity and interface defect was established.

2. Establishment of meso-model

2.1. Bilinear cohesive force model

The bilinear cohesion model was first proposed by Dugdale [6] and Barenblatt [7] to solve the elastic-plastic fracture problem of ductile metal materials. Its basic idea is to treat the material interface as an adhesive element with a certain thickness and bonding strength, and the mechanical response of the interface was defined through the traction separation rule. The typical two-line cohesive force model are shown in figure 1, n and t represent the normal and tangential directions of the interface respectively:

![Figure 1. Bilinear cohesive model. (a) Normal direction; (b) Tangential direction](image)

The failure process of interface damage can be divided into three stages: elastic stage, damage stage and failure stage. When the interface opening displacement reaches the critical value of damage $\delta^0$, the interface stress reaches the maximum value. With the increase of the displacement, the damage begins and the interface stress decreases. When the interface opening displacement is greater than the
maximum value $\delta^f$, the interface completely debonds. $\phi^f$ represents interfacial bonding energy. The tension displacement calculation equation of bilinear cohesion model is as follows:

$$
T = \begin{bmatrix}
\sigma \\
\tau
\end{bmatrix} = \begin{bmatrix}
(1 - D)k_n & 0 \\
0 & (1 - D)k_t
\end{bmatrix} \begin{bmatrix}
\delta_n \\
\delta_t
\end{bmatrix}
$$

(1)

Where $\sigma$ and $\tau$ represent the normal and tangential stresses on the interface respectively; $k_n$ and $k_t$ represent the normal and tangential stiffness of the interface respectively; $\delta_n$ and $\delta_t$ represent the normal and tangential opening displacements of the interface, $D$ is the interface damage variable, and the damage variable $D$ of the bilinear cohesion model was defined as:

$$
D = \begin{cases}
0 & (\delta \leq \delta^0) \\
\frac{\delta^f (\delta - \delta^0)}{\delta(\delta^f - \delta^0)} & (\delta > \delta^0)
\end{cases}
$$

(2)

Under the pure normal or pure tangential load, the damage variable can be used to judge whether the interface is damaged or not. However, under the condition of two-dimensional loading, the mixed mode damage initiation criterion as equation (3) should be used:

$$
\left(\frac{\sigma}{\sigma_{\text{max}}}\right)^2 + \left(\frac{\tau}{\tau_{\text{max}}}\right)^2 = 1
$$

(3)

The secondary energy release rate as equation (4) was often used as the failure criterion of mixed mode:

$$
\left(\frac{\phi_n}{\phi_{n_{\text{max}}}}\right)^2 + \left(\frac{\phi_t}{\phi_{t_{\text{max}}}}\right)^2 = 1
$$

(4)

where $\phi_n$ and $\phi_t$ represent the normal and tangential energy release rates respectively.

2.2. Meso finite element calculation model

At the mesoscale, the composite solid propellant could be regarded as a three-phase composite material composed of filling particles, binder matrix and the interface transition zone between them. Taking the initial hole defects and interface defects in the composite solid propellant into account, the Circular and polygonal particle fill models with random distribution of void defects was generated in ABAQUS by writing Python script. Polygonal particles are generated on the basis of circular particles. The position and size of polygonal particles are determined based on the radius of the circular particles and the coordinates of the center of the circle:

$$
\begin{align*}
\alpha_i &= \frac{2\pi i}{N} + \frac{\pi}{N} \cdot \text{random}(1) \\
x_i &= x_p + r \cdot \cos \alpha_i \\
y_i &= y_p + r \cdot \sin \alpha_i
\end{align*}
$$

(5)

Where $N$ is the number of sides of the polygon, and the sides number of the generated random polygon is $8 \sim 10$; $r$ is the radius of the circumscribed circle of the polygon particle, $(x_p, y_p)$ is the center coordinate of the circle, $\alpha_i$ is the angle between the polygon vertex / center line and the X axis of the global coordinate system; $(x_i, y_i)$ is the coordinate of the polygon particle vertex. Some studies have shown that the crack mainly propagates along the interface of large-size filled particles under the condition of loading at room temperature and low strain rate. In order to reduce the number of elements, and improve the calculation speed and ensure convergence, only the meso structure composed of larger particles and matrix was considered in the meso-model. According to the content of HTPB particles given in table 1, the random particle filling model as shown in figure 2 was generated. The calculation model size of composite solid propellant specimen is 1500μm×1500μm.
Table 1. The particle gradation and parameters of meso-geometric model

| Particle diameter/µm | Mass fraction/% | Volume fraction/% |
|----------------------|-----------------|------------------|
| AP particle          | 330             | 50               | 44.3             |
|                      | 130             | 14               | 12.2             |

In order to study the effects of void and interface defects on the macroscopic mechanical properties of composite solid propellant, the numerical simulation of 16 kinds of particle filling models with initial defects under four different porosity (0, 0.5%, 1%, 1.5%) and four different interfacial defects (0, 10%, 20%, 30%) were carried out in the numerical model. Three groups of parallel specimens were generated for each condition to eliminate the influence of random distribution of particles and defects. By writing Python script program, zero thickness cohesive elements were inserted between matrix / particle interface and matrix elements.

![Figure 2. Mesoscopic model of composite solid propellant.](image)

(a) Circular particle; (b) Polygonal particle

The interface defects in the meso calculation model were realized by defining the interface cohesive force elements with weakened mechanical properties. A certain proportion of cohesive force elements were randomly selected at the particle / matrix interface, and it is considered that these elements have incomplete adhesion defects with the filled particles. In the simulation modeling, the mechanical parameters of these interfacial defect elements are set to be 10% of that of well bonded interface elements.

The Poisson's ratio of propellant matrix is 0.49, which is approximately incompressible. The hybrid element CPE4H with plane strain 4-node quadrilateral linear integration was used to mesh the propellant matrix. CPE4 element was used to mesh AP particle matrix. A penalty function contact constraint was defined at the particle / matrix interface of AP to prevent matrix particles from invading each other. The global mesh size is 0.01mm and the maximum offset factor is 0.1. In this paper, the uniform displacement boundary condition was adopted. During the stretching process, the model's edges were kept straight, and the quasi-static smooth displacement loading was simulated. The ABAQUS/Standard implicit solver was used to calculate. In order to increase the convergence of the calculation, a certain viscosity was introduced into the bonding element. The initial step size is $10^{-5}$ and the minimum step size is $10^{-12}$, which meets the requirements of convergence.

The linear elastic model was used for the mechanical properties of propellant matrix and AP particles; the bilinear cohesive force model was used for the simulation calculation of interface elements, and the criteria of secondary nominal stress and secondary energy release rate were used as the damage initiation and damage evolution criteria of the interface bonding elements. The meso
components and relevant mechanical parameters of the interface were selected according to the reference [5], as shown in table 2 and table 3:

Table 2. Mechanical properties of meso-components

| component       | The elastic modulus/Mpa | Poisson ratio |
|-----------------|--------------------------|---------------|
| Matrix          | 0.46                     | 0.49          |
| AP particle     | 32450                    | 0.14          |

Table 3. Interfacial mechanical properties

| Interface                  | $\sigma_{max}$ (Mpa) | $\delta f$ (mm) | k/(MPa/mm) |
|----------------------------|-----------------------|-----------------|------------|
| Particle/matrix interface  | 0.4                   | 0.02            | 5000       |
| Matrix element interface   | 1.2                   | 0.013           | 460        |

3. Calculation results and analysis

3.1. Calculation results

Figure 3 and figure 4 show the stress nephogram when the strain of meso model of composite solid propellant reaches 5%, 10% and 15%.

Figure 3. Circular particle filling model Mises stress distribution (porosity 1%, interface defect ratio 20%). (a) strain 5%; (b) strain 10%; (c) strain 20%
Figure 4. Polygon particle filling model Mises stress distribution (porosity 1%, interface defect ratio 20%). (a) strain 5%; (b) strain 10%; (c) strain 20%

Because of the large number of figures, the Mises stress nephogram of the filling model with circular particles and polygonal particles are given only when the porosity is 1% and the interface defect is 20%. Figure 3 (a) and Figure 4 (a) show the Mises stress nephogram when the average strain is 5%. Due to the difference of elastic modulus between the filled particles and the matrix, the deformation of the matrix is greater than that of the particles. In the meso model, the stress-strain distribution is uneven, and microcracks initiate at the particle / matrix interface. The microcracks tend to appear around the particles with large size. Figure 3 (b) and Figure 4 (b) show the Mises stress nephogram when the strain is 10%. With the increase of strain, the incomplete coating area on the particle surface increases, and the cracks between the particles / matrix gradually expand and converge. Meanwhile, the reinforcement effect of the particles was weakened. Figure 3 (c) and Figure 4 (c) show the Mises stress nephogram when the strain is 20%. The crack displacement at the particle / matrix interface increases and converges with the matrix crack to form a fracture surface. The bearing capacity of the specimen disappears completely and fracture occurs.

3.2. Effect of initial damage on tensile properties

Furtherly, in order to study the change law of initial tensile elastic modulus and tensile strength of composite solid propellant under different void ratio and interface defect rate, based on the definition method of propellant tensile mechanical property parameters shown in figure 5, the initial elastic modulus and tensile strength of the propellant were obtained from the stress-strain curves obtained by simulation. The initial elastic modulus was defined as the slope of the stress-strain curve at the origin,
and the tensile strength was defined as the stress value corresponding to the highest point on the stress-strain curve\(^9\).

Figure 5. Stress-strain curves for propellant defining the mechanical parameters

According to the above definition method of mechanical property parameters, the stress-strain curves of different initial defect conditions were analyzed, and the curves of initial tensile modulus and tensile strength with initial defects were obtained, as shown in figure 6 and figure 7.

![Stress-strain curves for propellant](image)

**Figure 6. Effects of Initial defects on elastic modulus of propellant.**

(a) Effects of interface defect ratio on elastic modulus; (b) Effects of porosity on elastic modulus

It can be seen from figure 6 that the tensile elastic modulus of composite solid propellant decreases with the increase of interface defect rate under the condition of the same void ratio, and the influence degree of interface defect on elliptical and circular particle filled propellant is approximately the same. When the interface defect rate increases from 0 to 30%, the elastic modulus of composite solid
propellant decreases by about 40%. Under the condition of the same interface defect rate, the tensile elastic modulus of composite solid propellant decreases with the increase of void ratio. The effect of void ratio change on the elastic modulus of propellant filled with polygonal particles is more obvious than that with round particles.

Considering that the influence of interface defect rate and void ratio on tensile modulus of elasticity is approximately linear, the following equation was used for fitting:

$$E_t = E_{t0} [1 - (\alpha K + \beta P + \gamma)]$$  \hspace{1cm} (6)

Where:

- $E_t$ is the tensile elastic modulus of composite solid propellant,
- $E_{t0}$ is tensile elastic modulus of ideal composite solid propellant,
- $K$ is interface defect rate (%),
- $P$ is void ratio (%), and
- $\alpha$, $\beta$, $\gamma$ are fitting parameters.

The numerical results were fitted and the fitting parameters were shown in table 4. The correlation coefficients between the fitting curve of tensile elastic modulus of composite solid propellant filled with circular particles and polygonal particles are 0.9898 and 0.9824 respectively, which indicates that the formula (6) can well be used to predict the tensile modulus of composite solid propellant.

| Fitting parameters       | $E_t$/MPa | $\alpha$   | $\beta$   | $\gamma$   | Fitting correlation coefficient |
|--------------------------|-----------|------------|------------|------------|-------------------------------|
| Circular particle filling| 5.98      | 0.01223    | 0.10062    | 0.01609    | 0.9898                        |
| Polygon particle filling | 5.93      | 0.01118    | 0.17499    | 0.03489    | 0.9824                        |

It can be seen from figure 7 (a) that the tensile strength of composite solid propellant decreases with the increase of interface defect rate under the condition of the same void ratio, and the influence of interface defect rate on the tensile strength of composite solid propellant is approximately linear. At the same time, it is also found that the tensile strength of composite solid propellant filled with polygonal particles is smaller than that filled with circular particles with the same void ratio. It can be seen from figure 7 (b) that the tensile strength of composite solid propellant decreases nonlinearly with the increase of void ratio under the condition of the same interface defect rate.
Figure 7. Effects of Initial defects on tensile strength of propellant.
(a) Effects of interface defect ratio on tensile strength; (b) Effects of porosity on tensile strength

In order to study the quantitative relationship between the tensile strength of composite solid propellant and the micro initial defects, the numerical results were fitted and analyzed. Considering that the effect of interface defect rate on the tensile strength of composite solid propellant is approximately linear, and the effect of void ratio on the tensile strength of composite solid propellant is approximately logarithmic, the following equation was adopted to fit the curve:

$$f_t = f_{t_0}[1 - (\eta K + \mu) \ln(\varphi P + \lambda) + \nu]$$  

(7)

Where:
- $f_t$ is the tensile strength of composite solid propellant,
- $f_{t_0}$ is tensile strength of ideal composite solid propellant,
- $K$ is interface defect rate (%),
- $P$ is void ratio (%), and
- $\eta$, $\mu$, $\varphi$, $\lambda$, $\nu$ are fitting parameters. The numerical results were fitted and the fitting parameters were shown in table 5. The correlation coefficients between the fitting surface and the numerical results are 0.9974 and 0.9983 respectively, which indicates that equation (7) can well be used to predict the tensile strength of composite solid propellant.

Table 5. Fitting parameters of tensile strength equation

| Fitting parameters | $f_{t_0}$/N | $\eta$     | $\mu$     | $\varphi$ | $\lambda$ | $\nu$     | Fitting correlation coefficient |
|--------------------|-------------|------------|------------|------------|------------|-----------|--------------------------------|
| Circular particle filling | 0.365       | -0.00537   | 0.31191    | 0.08539    | 0.06914    | -0.8279   | 0.9974                           |
| Polygon particle filling | 0.356       | -0.00385   | 0.21782    | 0.06604    | 0.02692    | -0.7876   | 0.9983                           |

3.3. Initial damage analysis of composite solid propellant

Due to the difference of thermal conductivity between the filled particles and the matrix, the micro defects such as micropores and incomplete interfacial adhesion will be produced in the process of pouring, curing and cooling of composite solid propellant\[^{10}\]. The initial defects will degrade the macro mechanical properties such as tensile strength and initial elastic modulus of solid propellant. It is of great significance to establish the relationship between the macro mechanical properties and micro structural parameters of composite solid propellant for the structural integrity analysis of solid rocket motor grain.

According to the damage mechanics theory, the change of tensile strength of composite solid propellant caused by initial meso structure defect can be defined as initial damage. The initial damage variable of composite solid propellant can be calculated by the following equation\[^{11}\]:

$$f_t = f_{t_0}[1 - (\eta K + \mu) \ln(\varphi P + \lambda) + \nu]$$  

(7)
\[ D = \frac{(f_t - f_t^0)}{f_t^0} \] (8)

Where \( D \) is the initial tensile damage value of composite solid propellant; \( f_t \) is the tensile strength of composite solid propellant, \( f_t^0 \) is the ideal tensile strength of composite solid propellant without damage. The equation of initial tensile damage surface of solid propellant can be obtained by simultaneous equation (8) and tensile strength fitting equation (7) of composite solid propellant:

\[ D = (\eta K + \mu) \ln(\rho P + \lambda) - \nu \] (9)

Substituting the fitting parameters in Table 6 into equation (9), the relationship between the initial tensile damage surface of composite solid propellant and the initial meso defect parameters was shown in figure 8 and figure 9. The initial damage value of composite solid propellant increases with the increase of void ratio and interfacial defect rate, and the initial tensile damage surface shows obvious nonlinearity.

4. Conclusion

Based on Python script program, the particle filling model of composite solid propellant with micro holes and particle / matrix interface defects was established in ABAQUS. Cohesive force elements were embedded in the interface of particle / matrix and matrix element, and the tensile failure process of composite solid propellant was simulated. The complete failure process of solid propellant fracture...
was given, and the relationship between initial defect and macro tensile mechanical properties of propellant was studied. The results show that the elastic modulus of composite solid propellant decreases linearly with the increase of void ratio and interfacial defect rate. The tensile strength decreases linearly with the increase of interface defect rate, and logarithmically decreases with the increase of void ratio. On mesoscale, the initial tensile damage surface of composite solid propellant shows obvious nonlinearity, and the initial damage value increases with the increase of void ratio and interface defect rate. Under the same initial defect condition, the initial damage value of polygonal particle filling model is larger than that of circular filling particle model. The time and temperature dependence of mechanical properties of propellants and the damage of filled particles were not considered in the model. In the next step, a more perfect simulation model of material properties will be established to improve the accuracy of the model.

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