Full Paper

Propellants, Explosives, Pyrotechnics

DOI: 10.1002/prep.201900277

Study on the Fracture Properties of HTPB Propellant at Low Temperature

Bing Long,*[a] Hongliang Wang,[a] and Shuangsheng Gao[a]

Abstract: In order to study the fracture behaviour of hydroxyl-terminated polybutadiene (HTPB) propellant, the low temperature fracture toughness experiment of HTPB propellant was carried out by using centrally cracked sheet tensile specimens. The fracture toughness values of HTPB propellant with mode I crack at different temperature and loading rate were obtained, and the effect of specimen thickness on the fracture toughness was investigated. The experimental results show that the fracture toughness value continuously increases with the decreasing of temperature and the increasing of loading rate, and a linear relationship exists between the fracture toughness and the logarithm of loading rate. The effect on the fracture toughness by the change in loading rate is more notable at a lower temperature. Both at 25 °C and −40 °C, the fracture toughness of thick specimens is higher than that of thin specimens, and at low temperature the change is more obvious. A master curve of quadratic function for the fracture toughness was obtained. The influence of temperature on the fracture toughness is more obvious than that of the strain rate with double factor analysis of variance.

Keywords: Fracture toughness · HTPB propellant · Low temperature · centrally cracked sheet tensile specimen

1 Introduction

Hydroxyl-terminated polybutadiene (HTPB) propellant is widely used in solid rocket motors. In recent years, with the diversification of military missions and the deepening of space exploration activities, the range of service environment temperature of solid rocket engines has become wider and wider, and the risk of facing extremely low temperature is increasing. The mechanical properties of the matrix, particles and interface change, and the fracture properties of the propellant will change significantly at low temperature compared with the normal temperature. A crack in the propellant grain provides additional burning area, and affects the ballistic performance of the solid rocket engines. It also affects the mechanical properties which will destroy the structural integrity of the propellant grain, and may cause serious accidents such as ignition explosion in extreme case [1]. Thus, it is necessary to study the fracture properties of propellants at low temperature.

The effects of temperature and strain rate on the quasi-static crack propagation in a particulate composite material have been studied [2]. A power law relationship exists between the crack growth rate and the Mode I stress intensity factor, and under the test conditions considered, the effect of strain rate on crack growth is relatively small. The crack growth rate at low temperature is larger than that at normal temperature. Microstructure damage and fracture processes of H-24 propellants at three loading rates (3.2, 6.4, and 8.4 mm/min) and three temperatures (−54, 25, and 71 °C) were investigated by using centrally cracked sheet tensile samples [3]. A solid propellant has been tested to determine the critical crack size by applying both standard and fracture-mechanical test methods, and the master curve for the fracture toughness has been obtained using MT specimens. Master curves for the fracture energy G_F and the critical COD (crack-opening displacement) have been obtained by using the Wedge-Splitting Tests technique [4]. The effect of ageing on the crack growth mechanism in a composite propellant has been studied to obtain an understanding of the fracture process under service life conditions [5], and a distinct difference in the mechanism of crack growth was observed for the accelerated aged specimens. A fracture mechanics method based on the J integral was adopted to study the fracture behavior of a hydroxyl-terminated polybutadiene propellant with the assistance of a high-speed microscopic camera and multiple-sample test procedure [6].

In recent years there has been an increasing studies on temperature/strain rate effects in polymer-bonded explosives. Temperature-time response of the energetic composition EDC37 in compression has been studied. Failure stress is a monotonic function of applied strain rate or temperature, which is dominated by the relaxation properties of the polymeric binder. Similarities between the compressive strain rate/temperature data sets can be understood by temperature-time superposition [7]. The mechanical response of a polymer bonded explosive has been

[a] *B. Long, H. Wang, S. Gao*
Changzhou Institute of Technology, Changzhou, Jiangsu, 213032, P. R. China
**e-mail: longbingfh@126.com*

researched using a Split Hopkinson Pressure Bar. The failure mechanism is found to transition from shear-banding with crystal debonding fracture to brittle failure with some evidence of crystal fracture [8]. The strain rate and temperature sensitivity of Rowanex 1100 Type 1 A (a polymer-bonded explosive) is investigated. The stress at high rates of deformation ($1750 \pm 225\ s^{-1}$) is found to be about an order of magnitude greater than that supported at low rates ($0.015\ s^{-1}$), and temperature is also found to have a large effect [9].

In the presented work, the fracture behaviors of HTPB propellant at different temperatures and loading rate were studied, and fracture toughness values of the propellant at different conditions were obtained. A master curve for the fracture toughness was obtained, and the effect of specimen thickness on fracture toughness value was researched.

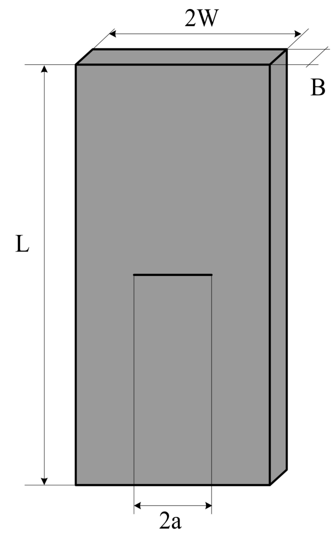

Figure 1. Geometrical shape of specimen.

2 Experimental Section

2.1 Materials and Sample Configuration

In this paper, the propellant material was HTPB propellant which was formulated and produced at the Fourth Academy of China Aerospace Science and Technology Corporation. The specimen material is manufactured by casting method comprising 86% solid particles (AP/AL), 10% HTPB binder and 4% others. All the particle sizes obey normal distribution, and the mean value of large and small AP particles is about 215,150 μm, the variance is 19.5, 10.55 μm, respectively. The mean value and the variance of metallic aluminum particles is about 120 and 8 μm, respectively. A centrally cracked sheet tensile sample was chosen in this test, and a $100 \times 50 \times 5$ mm rectangle specimen was die-cut out of propellant slabs, a crack was made by a razor-sharp blade of the required length at the center(Figure 1), the initiation crack length $2a = 16$ mm. The thickness of blade was 300 μm, comparable with the size of the larger AP particles, so that the notch produced through the blade could be assumed to be as sharp as a crack obtained by a preliminary subcritical cycling stage. Since the rectangular specimens cannot be directly loaded on the test machine, they are bonded to the Aluminum chuck using epoxy resin (Figure 2).

Figure 2. Specimen of propellant.

The size of $100 \times 50 \times 10$ mm specimen was fabricated to study the influence of the thickness on the fracture toughness of the propellant.

2.2 Experimental Design Conditions

In this study, five temperatures (25 °C, −10 °C, −30 °C, −40 °C and −50 °C) and four crosshead speeds (5 mm/min, 20 mm/min, 50 mm/min and 200 mm/min) were used. The specimens were kept in an incubator at the required temperature for at least 1 hour and then tested on a drawing mill with an incubator which was cooled by dry ice. Three specimens were measured under every test condition, and the fracture toughness value was taken as the average value. The fracture toughness value of thick specimens were measured under the conditions of 25 °C, 20 mm/min, −40 °C and 20 mm/min respectively.

2.3 Fracture Toughness Calculation

The crack tip stress intensity factor corresponding to the tensile instability point of the centrally cracked sheet tensile sample is the fracture toughness value. With the maximum force and the specimen geometry (Figure 1), the fracture toughness can be calculated as [10]:

$$K_{IC} = \frac{P}{B\sqrt{W}} f\left(\frac{a}{W}\right) \quad (1)$$

where P is the tensile force corresponding to the tensile instability point, B and W are the thickness and width of the specimen respectively, and f is a geometric factor computed with the following expression:

$$f\left(\frac{a}{W}\right) = \sqrt{\frac{\pi a}{4W} \sec \frac{\pi a}{2W}} \left[1 - 0.025\left(\frac{a}{W}\right)^2 + 0.06\left(\frac{a}{W}\right)^4\right] \quad (2)$$

3 Results and Discussion

3.1 Stress-Strain Curve

The stress-strain curves of the HTPB propellant fracture test at 25 °C and −40 °C are shown in Figure 3 and the characteristics are represented as follow:

Temperature and strain rate significantly influenced the mechanical behavior of the HTPB propellant. A decrease in the temperature and an increase in the strain rate generated an obvious increase in the stress. Additional, the propellant exhibited strong nonlinear mechanical behaviors under each experimental condition.

3.2 Fracture Toughness

With the load-displacement curve of the tensile fracture process, the fracture toughness value (K_{IC}) of the propellant specimen are summarized in Table 1.

The fracture toughness value with temperature changes is shown in Figure 4. As it's shown, the fracture toughness value continuously increases with the decreasing of temperature. This is mainly due to the fact that the propellant performs brittle property, the bond strength of the matrix and the matrix/particle interface increases at low temperatures. At the same temperature, the fracture toughness of the propellant gradually increases with the strain rate increasing.

The relationship between fracture toughness and loading rate at different temperatures is shown in Figure 5. As it's shown, a linear relationship exists between the fracture toughness and the logarithm of the loading rate, the fitting formulas are shown in Table 2. The slope of the line increases with the temperature decreasing, which indicates that changing the loading rate has a more significant effect on fracture toughness at lower temperature.

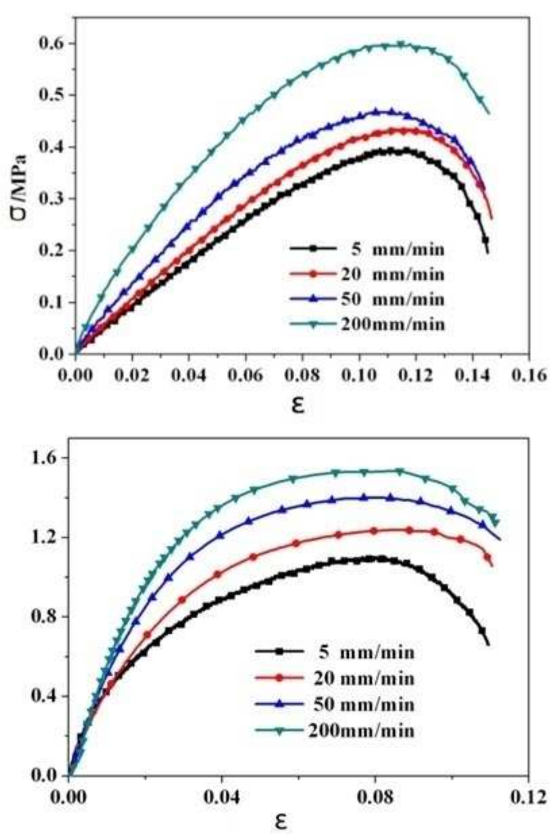

Figure 3. stress-strain curves.

Table 1. Experimental results of $K_{IC}/(MPa \cdot mm^{0.5})$.

Temperature/°C	Load rate ε'(mm/min)			
	5	20	50	200
25	1.76	2.31	2.74	3.43
−10	3.31	4.16	4.90	5.94
−30	4.56	5.62	6.58	7.72
−40	5.87	6.81	8.08	9.10
−50	7.38	8.46	9.72	11.47

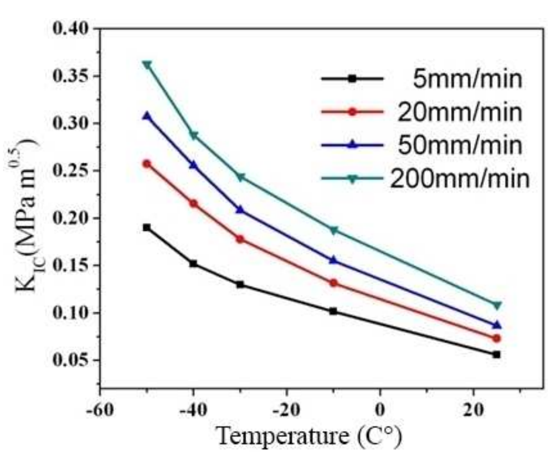

Figure 4. Plot of K_{IC} dependence on temperature.

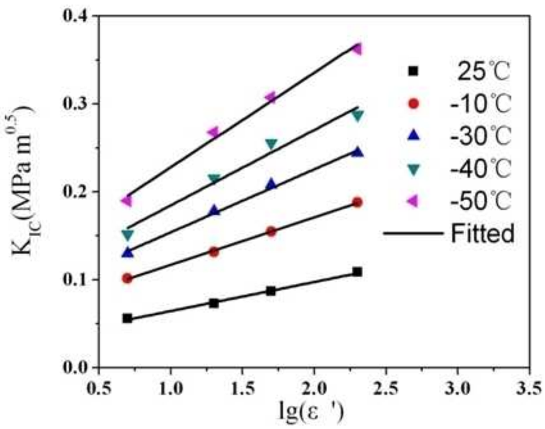

Figure 5. Plot of K_{IC} dependence on loading rate (loading rate in mm/min).

Table 2. The relationship between K_{IC} and strain rate.

Temperature/°C	Fitting relationship	R
25	$K_{IC} = 0.0330 \cdot \lg(\varepsilon') + 0.0314$	0.9950
−10	$K_{IC} = 0.0541 \cdot \lg(\varepsilon') + 0.0628$	0.9985
−30	$K_{IC} = 0.0717 \cdot \lg(\varepsilon') + 0.0823$	0.9920
−40	$K_{IC} = 0.0856 \cdot \lg(\varepsilon') + 0.0989$	0.9615
−50	$K_{IC} = 0.1075 \cdot \lg(\varepsilon') + 0.1206$	0.9877

3.3 The Thickness Dependence of the Fracture Toughness

Results of the toughness for different thickness specimens are shown in Table 3. Both at 25 °C and −40 °C, the fracture toughness of thick specimens is larger than that of thin specimens, which is agreement with the results of [4]. For some materials, a thicker specimen gives lower fracture toughness due to the size effect, but it seems to be contrary for the composite solid propellant [11]. It can also be seen from Table 3 that, compared with the normal temperature, the change of the fracture toughness for thick specimens at −40 °C is more distinct than that of thin specimens. This may be due to the increase in bond strength of the matrix and the matrix/particle interface at low temperatures, which inhibits voiding, resulting in a decrease of the fracture process zone and crack opening displacement at the crack tip. It is believed to allow the development of transverse constraint in the thick specimens and caused the material to behave almost as a single phase continuum and the thick test piece exhibits typical brittle fracture behavior [12,13].

Table 3. Experimental results of K_{IC} with different thickness specimens/($MPa \cdot mm^{0.5}$).

Temperature/°C	Thickness/mm		Change rate/%
	5	10	
25	0.0703	0.0748	6.4
−40	0.2154	0.2517	16.9

3.4 Master Curve for Fracture Toughness

Toughness data can be correlated as conventional failure properties using the temperature reduced strain rate [14]. Tests at different temperature and strain rate with the same $\alpha_T \cdot \varepsilon'$ value produce equal fracture toughness. A master curve for the toughness is thus obtained (Figure 6), and the equation of the curve can be obtained by the least-squares regression as:

$$\begin{aligned} K_{IC} &= 0.2074 \cdot \log^2(\alpha_T \cdot \varepsilon') \\ &+ 0.4025 \cdot \log(\alpha_T \cdot \varepsilon') + 1.4198 \end{aligned} \tag{3}$$

Where, correlation coefficient R = 0.9072.

The fracture toughness has a quadratic function relationship with $\log(\alpha_T \cdot \varepsilon')$, including the relationship between temperature, strain rate and fracture toughness. Based on the relevant fracture failure criterion, the propellant failure prediction under different conditions can be analysed by using the master curve.

3.5 Double Factor Analysis of Variance

In order to analyze the impact of temperature and strain rate on fracture toughness, a double factor analysis of variance is employed. Taking the fracture toughness as the index, the temperature and strain rate as the influencing factors, the significance level is 0.05, and the *F* value of the variance analysis result is shown in Table 4, where F_{crit} is the critical value of *F*.

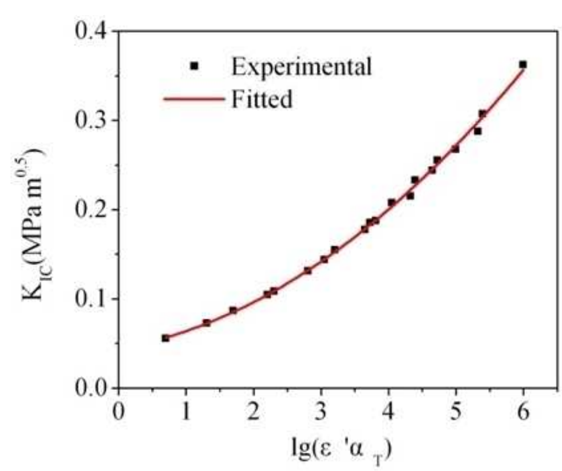

Figure 6. Master curve of K_{IC}.

Table 4. Analysis results of double variance for propellant fracture toughness.

Test statistic	Temperature	Strain rate	Interaction
F	1782.25	552.84	11.43
F_{crit}	2.87	3.10	2.12

As it's shown in Table 4, the *F* value of temperature and strain rate to the fracture toughness is significantly greater than the value of F_{crit} (2.87, 3.10), both temperature and strain rate have a significant effect on the fracture toughness of the propellant. And the *F* value of the temperature is larger than the *F* value of the strain rate, which indicates that the influence of temperature on the fracture toughness is greater than the influence of the strain rate. The dependence on strain rate is rather small, in agreement with the fact that the speed of stress relaxation for HTPB propellants is more sensitive to temperature than the strain rate between glass transition and the rubbery region. This is mainly because the speed of stress relaxation for HTPB propellants between the glassy and rubbery region is more sensitive to temperature than the strain rate, which is consistent with the results of [4].

4 Conclusion

The fracture properties of HTPB propellant at different temperatures and strain rates were studied, and the fracture toughness was calculated. The fracture toughness is obviously sensitive to temperature and loading rate, it increases with the decreasing of temperature and the increasing of loading rate, and a linear relationship exists between the fracture toughness and the logarithm of loading rate.

A temperature strain rate reduction has been obtained for the fracture toughness and the master curve can be expressed by the equation:

$$\begin{aligned} K_{IC} &= 0.2074 \cdot \log^2(\alpha_T \cdot \varepsilon') \\ &+0.4025 \cdot \log(\alpha_T \cdot \varepsilon') + 1.4198 \end{aligned} \tag{4}$$

Within the appropriate temperature and loading rate, the equation can be used for temperature and loading rate conversion.

Both at 25 °C and −40 °C, the fracture toughness of thick specimen is larger than that of thin specimen, and at low temperature the change is more distinct.

The influence of temperature on the fracture toughness is greater than the influence of the strain rate at the conditions of this paper.

Symbols and Abbreviations

HTPB Hydroxyl-terminated polybutadiene
K_{IC} critical stress intensity factor in mode I
F_{crit} critical value of F

References

[1] *Structural assessment of solid propellant grains. AGARD-AR-350*, France, Advisory Group for Aerospace Research & Development, **1997**.

[2] C. T. Liu, C. W. Smith, Temperature and Rate Effects on Stable Crack Growth in a Particulate Composite Material, *Exp. Mech.* **1996**, *36*, 290–295.

[3] C. D. Bencher, R. H. Dauskardt, R. O. Ritchie, Microstructural Damage and Fracture Processes in a Composite Solid Rocket Propellant, *J. Spacecr. Rockets* **1995**, *32*, 328–334.

[4] G. S. Tussiwand, V. E. Saouma, R. Terzenbach, L. T. D. Luca, Fracture Mechanics of Composite Solid Rocket Propellant Grains: Material Testing, *J. Propul. Power*, **2009** *25*: 60–73.

[5] K. M. Ide, S. Y. Ho, D. Williams, Fracture behavior of accelerated aged solid rocket propellants, *J. Mater. Sci.* **1999**, *34*, 4209–4218.

[6] G. P. Zhou, X. C. Yin, A. Q. Li, Study on the Fracture Toughness of Hydroxyl-Terminated Polybutadiene Solid Rocket Propellant, *J. Propul. Power* **2015**, *31(3)*, 912–918.

[7] D. M. Williamson, C. R. Siviour, W. G. Proud, S. J. P. Palmer, R. Govier, K. Ellis, P. Blackwell, C. Leppard, Temperature-time response of a polymer bonded explosive in compression (EDC37)" *J. Phys. D* **2008**,*41*, 085404.

[8] D. R. Drodge, D. M. Williamson, S. J. P. Palmer, W. G. Proud, R. K. Govier, The mechanical response of a PBX and binder: Combining results across the strain-rate and frequency domains, *J. Phys. D* **2010**, *43*, 335403.

[9] S. M. Walley, N. E. Taylor, D. M. Williamson, Temperature and strain rate effects on the mechanical properties of a polymer-bonded explosive, *Eur. Phys. J. Special Topics* **2018**, *227*, 127–141.

[10] T. L. Anderson, *Fracture Mechanics, Fundamentals and Applications.* 2nd ed, CRC Press, Boca Raton, FL, **1995**, 373–513.

[11] V. E. Saouma, D. Natekar, E. Hansen, Cohesive Stress and Size Effects in Elasto-Plastic and Quasi-Brittle Materials, *Int. J. Fract.*, **2003**, *119*, 287–298.

[12] W. G. Smith, C. T. Liu, *Global and near tip response of cracked solid propellant.* **1993**, ADA 418742.

[13] C. T. Liu, W. G. Smith, *Near-tip behavior in a particulate material under constant strain rate including temperate and thickness effects.* **2001**, ADA 410504

[14] A. J. Kinloch, R. J. Young, *Fracture Behaviour of Polymers*, Applied Science Publishers, London, **1983**.

Manuscript received: August 12, 2019
Revised manuscript received: September 10, 2019
Version of record online: October 22, 2019

