Storage Life Prediction of Composite Propellant Based on Reaction Kinetics

Zhengqiang Pan, *Member, IAENG*, and Ruibing Bian

Abstract—**The physical and chemical properties of composite propellants are greatly affected by environmental factors, among which temperature is one of the most important factors. The composite propellant will gradually deteriorate under the influence of temperature. In view of the insufficient utilization of current composite propellant test data and low accuracy of life estimation model, this paper takes a certain type of composite propellant as the object to carry out the storage life prediction research by its destructive accelerated storage test data. From the perspective of thermal analysis kinetics, the tensile strength is taken as the key performance parameter, and the performance degradation model based on the n^{th} order reaction kinetics equation is built. Next, the optimal order n is chose based on the AIC criterion. Finally, the estimation methods of storage reliability, storage life and their lower confidence limits are given. This work indicates that the proposed model gives some good technological approaches for solving the storage life estimation problem.**

Keywords—**composite propellant, reaction kinetics, storage reliability, storage life, lower confidence limit**

I. Introduction

The composite propellant is a type of propellant composed of a mixture of polymeric binder, solid powder oxidant, powdered metal fuel, and other additional ingredients. It is also called heterogeneous propellant because it is non-uniform in physical structure and has a distinct interface between the ingredients. The aging mechanism of the composite solid propellant is different depending on the binder, thus it is very complicated. Its typical aging mechanisms include post-cure, oxidative cross-linking, and chain scission of the polymer.

Extensive research on the storage life of composite propellants has been conducted both here and abroad. The performance parameters of solid propellants are generally mechanical properties such as relaxation modulus, Poisson's ratio, tensile strength and maximum elongation. At present, the methods reported in the literatures generally adopt an exponential function, a power function or a linear function to fit the variation of the performance characteristic parameters of the explosives, and obtain a pseudo-life value under each stress level according to the failure threshold. Some typical acceleration equations are used to model the storage life under normal stress, such as Arrhenius equation, the Berthelot equation, the Eyring model, etc. [1-7]. In view of the shortcomings of such models, some scholars have revised the traditional methods to improve the accuracy of propellant life prediction to some extent [8-11]. However, in order to further improve the accuracy of propellant life prediction, there is still a lot of work to do just by relying on the modified aging model. Therefore, it is necessary to explore some aging models with high precision and good stability.

Accelerated life test of composite propellant is a typical destructive test. In recent years, aim at the physical and chemical characteristics of propellant, some scholars have carried out some studies from the perspective of thermal analysis kinetics, and described the degradation mechanism of its performance parameters by the reaction kinetics equation. For instance, Wang [12] established the relationship model between aging degree and temperature of NEPE propellant by using reaction kinetic equation. Lee [13] et al. described the stabilizer consumption process of single-base propellant by using kinetic equation, and gave the optimal kinetic equation series according to its accelerated test data, so as to predict the storage life. In order to investigate the aging kinetics of composite solid propellant under the alternating temperature load and reveal its aging mechanism, Wang et al. [14] established an alternating temperature accelerated aging kinetic model, gave the calculation method of equivalent temperature and equivalent cycle time and the calculation formula for the activation energy in an alternating temperature environment, and put forward the aging mechanism of the composite solid propellant under alternating temperature load. Son et al. [15] introduced the use of reaction kinetics equations to solve the problem of storage reliability estimation of destructive measurement products, and provided a good solution for the estimation of propellant storage life.

In this paper, the accelerated storage test is introduced for a certain type of composite propellant, and the tensile strength is chose as the key performance parameter. Combined with the distribution of the test data at each measurement time, the performance degradation model based on the n^{th} order reaction kinetic equation is established. Furthermore, the storage reliability, storage life and their lower confidence limits are obtained, which provides a good means for solving the estimation of the explosives storage life.

II. Experimental

For a certain type of composite propellant, accelerated storage test was carried out at three temperature levels to

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track the change of tensile strength during the aging process of the propellant. The temperature levels are 55 ℃ (Case 1), 65 ℃ (Case 2) and 75 ℃ (Case 3), respectively. Since the test is destructive in the experiment, only a data can be obtained for each sample. At each stress level, 4 measurements are performed, and the number of samples per measurement is 5. Therefore, the total number of samples is 120.

The test data collected according to the test scheme is shown in fig. 1.

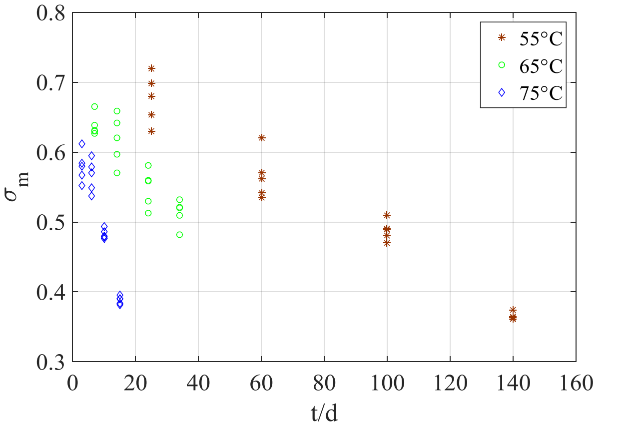

Fig. 1. accelerated storage test data of a certain type of composite propellant

According to the conclusion of the failure mechanism analysis, the composite propellant mainly exhibits the chain scission of the polymer during the aging process, so its tensile strength decreases by test time. It can be seen from fig. 1 that the temperature has a great influence on the composite propellant storage life. As the temperature level elevates, the decline rate tensile strength increases remarkably. Therefore, the measurement intervals at each temperature level are significantly different during the test. The higher the temperature is, the shorter the detection interval and the test time are. In this test, the cut-off times at the three temperature levels of 55 ℃, 65 ℃, and 75 ℃ were 140 days, 34 days, and 15 days, respectively. According to the relevant research conclusions, it can be considered that the composite propellant fails if the tensile strength drops to 0.5 MPa.

III. DEGRADATION MODELING OF COMPOSITE PROPELLANT BASED ON REACTION KINETICS

The failure mechanism of composite propellant is very complicated, and it is difficult to know its real chemical reaction process. Therefore, this paper takes advantage of the reaction kinetics to estimate the storage life of composite propellant. This method is usually used to estimate the storage reliability of one-shot products with accelerated degradation data when chemical reaction or degradation trend is unknown. Specifically, the n^{th} reaction kinetics of physical model is adopted to analyze the degradation phenomenon of one-shot products using destructive degradation data, and an accelerated degradation model is established to estimate the storage reliability or life at a normal temperature.

A. Model description

As shown in fig. 1, the tensile strength of the composite propellant decreased significantly with the test time during the test. Let $Y(t,T)$ denotes the tensile strength of the composite propellant under stresses T at time t with the initial content, $Y_0 = Y(t=0)$, and define the rate of the fraction degraded as

$$\alpha(t,T) = \frac{Y_0 - Y(t,T)}{Y_0}. \tag{1}$$

According to the principle of thermal analysis kinetic, the rate of the fraction degraded over time $\alpha(t,T)$ could be determined by both reaction rate coefficient $K(T)$and n-th order kinetics from kinetics of a reaction. Therefore, the rate could be expressed as

$$\frac{d}{dt}[\alpha(t,T)] = K(T)\big(1-\alpha(t,T)\big)^n. \tag{2}$$

From the integration of (2) with the initial condition $\alpha(0,T) = 0$,the fraction degraded over time could be rewritten using $K(T)$ and n as

$$\alpha\,(t,T) = \begin{cases} 1 - e^{-K(T)t}, & n = 1 \\ 1 - [(n-1)K(T)t + 1]^{\frac{1}{1-n}}, & n \neq 1 \end{cases}, \tag{3}$$

Substituting (3) into (1) and arrangement provide the response at time t as

$$Y(t,T) = \begin{cases} Y_0 e^{-K(T)t}, & n = 1 \\ Y_0[(n-1)K(T)t + 1]^{\frac{1}{1-n}}, & n \neq 1 \end{cases}. \tag{4}$$

Assuming that the tensile strength values detected at each moment follow a normal distribution, the mean can be expressed as

$$\mu(t,T) = E[Y(t,T)] = \begin{cases} Y_0 e^{-K(T)t}, & n = 1 \\ Y_0[(n-1)K(T)t + 1]^{\frac{1}{1-n}}, & n \neq 1 \end{cases}. \tag{5}$$

For temperature stress T in Kevin, the reaction rate coefficient has a form for Arrhenius model as

$$K(T) = A\,exp\left(-\frac{E_a}{kT}\right), \tag{6}$$

where A stands for pre-exponential factor, E_a for activation energy, and k for Boltzmann's constant. For the convenience of notation, let $\gamma_0 = Y_0$, $\gamma_1 = A$, $\gamma_2 = \frac{E_a}{k}$.

It is assumed that the standard deviation of the tensile strength at each moment is independent of the stress level and the detection time, that is, $\sigma(t) = \sigma$, which is a constant.

Assuming that the test sets Q stress levels, $q = 1, 2, \cdots, Q$, the number of detections is r_q under the stress level T_q, the number of i-th detection samples is n_i, and the detection time is t_i, $i = 1,2,\cdots,r_q$, the performance parameter value of the j-th sample is denoted as $Y_j(t_i,T_q)$, $j = 1,2,\cdots,n_i$, and the likelihood function is expressed as

$$L\big(\boldsymbol{\theta}\big|Y(t,T)\big) = \prod_{q=1}^{Q}\prod_{i=1}^{r_q}\prod_{j=1}^{n_i}\left\{\frac{1}{\sqrt{2\pi}\sigma}\exp\left(-\frac{1}{2}\left(\frac{Y_j(t_i,T_q)-\mu(t_i,T_q)}{\sigma}\right)^2\right)\right\}, \tag{7}$$

where $\boldsymbol{\theta} = (\gamma_0, \gamma_1, \gamma_2, \sigma)$, the corresponding logarithmic likelihood function is

$$\ln L\big(\boldsymbol{\theta}\big|Y(t,T)\big) = \sum_{q=1}^{Q}\sum_{i=1}^{r_q}\sum_{j=1}^{n_i}\left\{-ln(\sqrt{2\pi}) - ln(\sigma) - \frac{1}{2}\left(\frac{Y_j(t_i,T_q)-\mu(t_i,T_q)}{\sigma}\right)^2\right\}. \tag{8}$$

The Maximum likelihood estimators to the likelihood function are obtained from the optimization problem as follows.

$$\hat{\boldsymbol{\theta}} = max\{\ln L\big(\boldsymbol{\theta}\big|Y(t,T)\big)\}.$$

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B. Case Study

For the n^{th} order reaction kinetic equation, the typical n value is selected as

$$n = \left[0, \frac{1}{4}, \frac{1}{3}, \frac{1}{2}, \frac{2}{3}, 1, \frac{3}{2}, 2, \frac{5}{2}\right].$$

Then, we can obtain the other parameters of the model for different n values. Meanwhile, AIC (Akaike Information Criterion) is used to choose the best model for propellant storage life estimation. AIC is defined as

$$AIC = -2 \times max\ lnL(\theta|Y(t,T)) + 2m$$

where m is the number of unknown parameters in the model. AIC has been used as a model selection criterion in many places. If several potential models are available, their AIC values are calculated, respectively. Finally, the model with the minimum AIC value is considered to be the best choice.

The model parameter and AIC values obtained according to (8) are shown in table I.

TABLE I
MODEL PARAMETER ESTIMATION AND AIC VALUE

n	γ_0	γ_1	γ_2	σ	AIC value
0	0.6943	3.2427E+14	1.2865E+4	0.0336	-339.18
1/4	0.6987	3.8588E+14	1.2902E+4	0.0340	-337.61
1/3	0.7002	4.0959E+14	1.2915E+4	0.0342	-337.06
1/2	0.7031	4.6255E+14	1.2941E+4	0.0345	-335.94
2/3	0.7062	5.2393E+14	1.2968E+4	0.0349	-334.79
1	0.7126	6.7806E+14	1.3024E+4	0.0356	-332.41
3/2	0.7230	1.0199E+15	1.3113E+4	0.0367	-328.75
2	0.7348	1.5751E+15	1.3206E+4	0.0378	-325.08
5/2	0.7486	2.5024E+15	1.3303E+4	0.0389	-321.48

According to table I, the AIC value of 0^{th} reaction kinetic model is the minimum. Therefore, the degradation model of this type of composite propellant can be described by 0^{th} reaction kinetic equation.

IV. PREDICTION OF STORAGE LIFE OF COMPOSITE PROPELLANTS

A. Storage reliability estimation

It can be seen from fig. 1 that the tensile strength of the composite propellant is gradually decreasing. For the pre-defined failure threshold D_f, storage reliability for a storage temperature T at time t could be estimated as

$$R(t,T) = P(Y(t,T) > D_f) = \Phi\left(\frac{\mu(t,T) - D_f}{\sigma}\right). \quad (9)$$

The lower confidence limit of storage reliability can be expressed as follows [16].

$$R_L(t,T) = R(t,T)/exp\left\{\frac{Z_{C.L}\sqrt{V[R(t,T)]}}{R(t,T)}\right\}, \quad (10)$$

where $Z_{C.L}$ is 1.2816 and 1.6449 for confidence levels, 90% and 95%, respectively. The variance in (10) was evaluated using Fisher's information matrix for $\boldsymbol{\theta}$.

$$V[R(t,T)] = \boldsymbol{h}_R I(\boldsymbol{\theta})^{-1} \boldsymbol{h}'_R, \quad (11)$$

where

$$\boldsymbol{h}_R = \left(\frac{\partial R(t,T)}{\partial \gamma_0}, \frac{\partial R(t,T)}{\partial \gamma_1}, \frac{\partial R(t,T)}{\partial \gamma_2}, \frac{\partial R(t,T)}{\partial \sigma}\right)\Bigg|_{\boldsymbol{\theta}=\hat{\boldsymbol{\theta}}}, \quad (12)$$

$$\boldsymbol{I}(\boldsymbol{\theta}) =$$

$$\begin{pmatrix} E\left(-\frac{\partial^2 \ln L(\boldsymbol{\theta})}{\partial \gamma_0{}^2}\right) & E\left(-\frac{\partial^2 \ln L(\boldsymbol{\theta})}{\partial \gamma_0 \partial \gamma_1}\right) & E\left(-\frac{\partial^2 \ln L(\boldsymbol{\theta})}{\partial \gamma_0 \partial \gamma_2}\right) & E\left(-\frac{\partial^2 \ln L(\boldsymbol{\theta})}{\partial \gamma_0 \partial \sigma}\right) \\ E\left(-\frac{\partial^2 \ln L(\boldsymbol{\theta})}{\partial \gamma_1 \partial \gamma_0}\right) & E\left(-\frac{\partial^2 \ln L(\boldsymbol{\theta})}{\partial \gamma_1{}^2}\right) & E\left(-\frac{\partial^2 \ln L(\boldsymbol{\theta})}{\partial \gamma_1 \partial \gamma_2}\right) & E\left(-\frac{\partial^2 \ln L(\boldsymbol{\theta})}{\partial \gamma_1 \partial \sigma}\right) \\ E\left(-\frac{\partial^2 \ln L(\boldsymbol{\theta})}{\partial \gamma_2 \partial \gamma_0}\right) & E\left(-\frac{\partial^2 \ln L(\boldsymbol{\theta})}{\partial \gamma_2 \partial \gamma_1}\right) & E\left(-\frac{\partial^2 \ln L(\boldsymbol{\theta})}{\partial \gamma_2{}^2}\right) & E\left(-\frac{\partial^2 \ln L(\boldsymbol{\theta})}{\partial \gamma_2 \partial \sigma}\right) \\ E\left(-\frac{\partial^2 \ln L(\boldsymbol{\theta})}{\partial \sigma \partial \gamma_0}\right) & E\left(-\frac{\partial^2 \ln L(\boldsymbol{\theta})}{\partial \sigma \partial \gamma_1}\right) & E\left(-\frac{\partial^2 \ln L(\boldsymbol{\theta})}{\partial \sigma \partial \gamma_2}\right) & E\left(-\frac{\partial^2 \ln L(\boldsymbol{\theta})}{\partial \sigma^2}\right) \end{pmatrix}, \quad (13)$$

The elements of Fisher information matrix in (13) are calculated as

$$E\left(-\frac{\partial^2 \ln L(\boldsymbol{\theta})}{\partial \gamma_g{}^2}\right) = \sum_{q=1}^{Q} \sum_{i=1}^{r_q} \sum_{j=1}^{n_i} \left[\frac{\left(\frac{\partial \mu(t_i, T_q)}{\partial \gamma_g}\right)^2}{\sigma^2}\right], \ g = 0,1,2,$$

$$E\left(-\frac{\partial^2 \ln L(\boldsymbol{\theta})}{\partial \gamma_g \partial \gamma_h}\right) =$$

$$\sum_{q=1}^{Q} \sum_{i=1}^{r_q} \sum_{j=1}^{n_i} \left[\frac{\left(\frac{\partial \mu(t_i, T_q)}{\partial \gamma_g}\right)\left(\frac{\partial \mu(t_i, T_q)}{\partial \gamma_h}\right)}{\sigma^2}\right], \ g \neq h, g, h = 0,1,2,$$

$$E\left(-\frac{\partial^2 \ln L(\boldsymbol{\theta})}{\partial \gamma_g \partial \sigma}\right) = 0, \ g = 0,1,2,$$

$E\left(-\frac{\partial^2 \ln L(\boldsymbol{\theta})}{\partial \sigma^2}\right) = \frac{2N}{\sigma^2}$, N is the total number of samples.

When $n = 0$,

$$\mu(t,T) = \gamma_0[-K(T)t + 1],$$

then

$$\frac{\partial \mu(t_i, T_q)}{\partial \gamma_0} = -\gamma_1 e^{-\frac{\gamma_2}{T_q}} t_i + 1,$$

$$\frac{\partial \mu(t_i, T_q)}{\partial \gamma_1} = -\gamma_0 e^{-\frac{\gamma_2}{T_q}} t_i,$$

$$\frac{\partial \mu(t_i, T_q)}{\partial \gamma_2} = \frac{\gamma_0 \gamma_1}{T_q} e^{-\frac{\gamma_2}{T_q}} t_i.$$

In (12), each element of $\boldsymbol{h}_R$ is expressed as

$$\frac{\partial R(t,T)}{\partial \gamma_g} = \phi\left(\frac{\mu(t,T) - D_f}{\sigma}\right) \cdot \frac{1}{\sigma} \cdot \frac{\partial \mu(t,T)}{\partial \gamma_g}, g = 0,1,2,$$

$$\frac{\partial R(t,T)}{\partial \sigma} = -\phi\left(\frac{\mu(t,T) - D_f}{\sigma}\right) \cdot \frac{\mu(t,T) - D_f}{\sigma^2}.$$

B. Storage life estimation

According to (9), the life equation of composite propellant can be derived as

$$\Phi^{-1}(R)\hat{\sigma} - \hat{\mu}(t, \boldsymbol{T}) + D_f = 0. \quad (14)$$

Given the reliability R, the reliability life t_R of the propellant at the stress level T can be obtained according to (14).

$$t_R = \begin{cases} \dfrac{\ln\left(\frac{\Phi^{-1}(R)\sigma + D_f}{\gamma_0}\right)}{-K(T)}, & n = 1 \\ \dfrac{\left(\frac{\Phi^{-1}(R)\sigma + D_f}{\gamma_0}\right)^{1-n} - 1}{(n-1)K(T)}, & n \neq 1 \end{cases}.$$

According to the analysis in the previous section, the 0-th reaction kinetic equation was chose to describe the degradation process of the composite propellant, which was further determined

$$t_R = \frac{1 - \left(\frac{\Phi^{-1}(R)\sigma + D_f}{\gamma_0}\right)}{K(T)}.$$

Using the δ method, the asymptotic variance of t_R could be written as

$$\mathrm{Avar}(t_R) = \boldsymbol{h_t I(\theta)}^{-1}\boldsymbol{h}_t',$$

Where, $\boldsymbol{I(\theta)}$is shown in (13), and

$$\boldsymbol{h_t} = \left(\frac{\partial t_R}{\partial \gamma_0}, \frac{\partial t_R}{\partial \gamma_1}, \frac{\partial t_R}{\partial \gamma_2}, \frac{\partial t_R}{\partial \sigma}\right)\Big|_{\boldsymbol{\theta}=\hat{\boldsymbol{\theta}}}. \quad (15)$$

The expression of each element of $\boldsymbol{h_t}$ is

$$\frac{\partial t_R}{\partial \gamma_0} = \frac{\Phi^{-1}(R)\sigma + D_f}{\gamma_0^2 K(T)},$$

$$\frac{\partial t_R}{\partial \gamma_1} = -\left(1 - \frac{\Phi^{-1}(R)\sigma + D_f}{\gamma_0}\right)\frac{1}{\gamma_1 K(T)},$$

$$\frac{\partial t_R}{\partial \gamma_2} = \left(1 - \frac{\Phi^{-1}(R)\sigma + D_f}{\gamma_0}\right)\frac{1}{TK(T)},$$

$$\frac{\partial t_R}{\partial \sigma} = -\frac{\Phi^{-1}(R)}{\gamma_0 K(T)}.$$

The lower confidence limit of reliable storage life can be expressed as follows [16].

$$\hat{t}_{RL} = \hat{t}_R / exp\left\{\frac{z_{C.L}\sqrt{\mathrm{Avar}(\hat{t}_R)}}{\hat{t}_R}\right\}. \quad (16)$$

C. Case study

According to the analysis of relevant engineering practice, when the tensile strength of this type of composite propellant decreases to 0.5MPa, it fails to meet the requirements and is deemed as failure, that is, $D_f = 0.5MPa$. Based on the analysis in this section, the storage reliability and its lower confidence limit, the storage reliability life and its lower confidence limit can be estimated.

According to the subsection A of Section IV, the reliable life of this composite propellant at 25 ℃ and its 90% and 95% lower confidence limit can be obtained as shown in fig. 2.

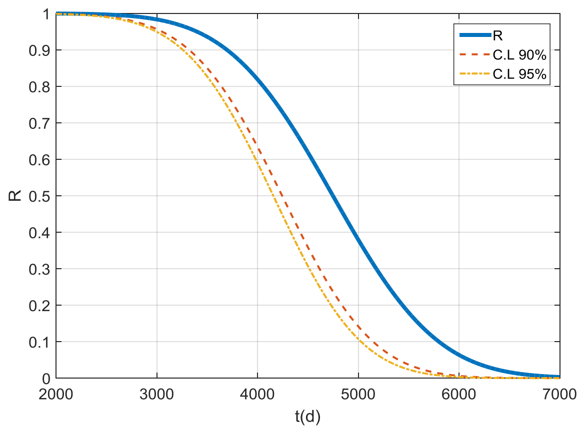

Fig. 2. Reliability of this composite propellant at 25 ℃ and its lower confidence limit

According to the above calculation process, the storage reliability and the lower confidence limit of the type of composite propellant under typical storage time can be estimated, as shown in table II.

TABLE II
STORAGE RELIABILITY OF A CERTAIN TYPE OF COMPOSITE PROPELLANT AND ITS LOWER CONFIDENCE LIMIT ESTIMATION

Storage time (year)	Reliability	90%Lower confidence limit	95%Lower confidence limit
5	0.9998	0.9994	0.9993
6	0.9991	0.9972	0.9967
7	0.9962	0.9893	0.9874
8	0.9870	0.9656	0.9597
9	0.9626	0.9081	0.8932
10	0.9095	0.7965	0.7671

Meanwhile, according to the analysis of subsection B of Section IV, the reliable storage life of the composite propellant and its lower confidence limit under different storage reliability can be calculated, as shown in table III.

TABLE III
STORAGE RELIABILITY LIFE OF A CERTAIN TYPE OF COMPOSITE PROPELLANT AND ITS LOWER CONFIDENCE LIMIT ESTIMATION

Reliability	Reliable life (year)	90% Lower confidence limit (year)	95%Lower confidence limit (year)
0.99	7.78	6.46	6.13
0.95	9.31	7.85	7.48
0.9	10.13	8.58	8.18
0.8	11.12	9.44	9.02
0.5	13.01	11.07	10.58

V. Conclusion

The paper studies the storage reliability and life estimation of a certain type of composite propellant. In view of the insufficient utilization of current composite propellant test data and low accuracy of life estimation model, we take the tensile strength as the key performance parameters, and establish a performance degradation model based on n^{th} order reaction kinetics equation from the perspective of thermal analysis kinetics. And then, the AIC criterion is utilized to choose the reaction kinetics equation order n. Furthermore, we gave the estimation method of the composite propellant storage reliability, reliable storage life and their lower confidence limits.

In fact, it is very difficult to accurately determine the form of the reaction kinetics equation of the composite propellant. The results of this paper are based on the reaction mechanism of function $h(\alpha) = (1-\alpha)^n$. In order to be as much as possible close to the true description of the reaction kinetic behavior of the composite propellant, we can take advantage of the $SB(m,n)$ kinetic model, i.e., $h(\alpha) = \alpha^m(1-\alpha)^n$ to depict its reaction kinetics behavior, and then derives its storage reliability and storage life index. At present, we are carrying out relevant research work, which is expected to be introduced in detail in the subsequent papers.

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