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Investigation on reliability of nanolayer-grained Ti_3SiC_2 via Weibull statistics

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Abstract Weibull modulus of bending strength of nanolayer-grained ceramic Ti_3SiC_2 was estimated with over 50 specimens, using the least square method, the moment method and the maximum likelihood technique, respectively. The result demonstrated that the *m*-value of this layered ceramic ranged from 25 to 29, which is much higher than that of traditional brittle ceramics. The reason of high Weibull modulus was due to high damage tolerance of this material. Under stress, delamination and kinking of grains and shear slipping at interfaces give this material high capacity of local energy dissipation and easy local stress relaxation, leading to the excellent damage tolerance of Ti_3SiC_2. The effect of amounts of specimens on the reliability of the estimated *m*-values was also investigated. It was confirmed that the stability of the estimated *m*-value increased with increasing numbers of specimens. The parameter obtained using the maximum likelihood technique showed the highest reliability than other methods. The ranges of failure probability were determined using the Weibull estimates calculated from the maximum likelihood technique.

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Introduction

For brittle materials, Weibull modulus is an important parameter for characterizing the reliability of strength and damage tolerance, so that this parameter plays an important role for the safe applications and risk evaluation. Generally, ceramics are brittle and sensitive to defects, and the random of the defects distribution leads to great scatter of strength data and low Weibull modulus. Weibull modulus of ceramics normally ranges from 5 to 20 [1, 2]. This low value of ceramic reflects the low reliability, which is a main obstacle for engineering applications. If a material is insensitive to flaws, Weibull modulus should be high. Generally, the value of Weibull modulus mainly depends on two key factors: (i) damage tolerance and ii) uniformity of a material. Therefore, great efforts have been made for enhancing the damage tolerance and uniformity of ceramics. However, there is not yet remarkable breakthrough for brittle ceramics.

During past decades, layered ternary ceramic Ti_3SiC_2 attracted great attentions of material scientists because this material displayed high damage tolerance and low ratio of hardness to elastic modulus. Therefore, a high Weibull modulus is expected for this ceramic. But this prediction has not been confirmed because a credible Weibull statistics of strength data needs large amounts of specimens from a uniform bulk material. Recently, a solution to fabricate large bulk Ti_3SiC_2 (135 mm in diameter and 20 mm in thickness) has been achieved in our laboratory. This gives us the opportunity to investigate the Weibull modulus of Ti_3SiC_2.

In this work, the Weibull modulus of Ti_3SiC_2 was estimated by means of the least square, the moment method and the maximum likelihood technique, respectively. The effect of amounts of samples on estimated m-value was also investigated. The mechanism of the high Weibull modulus of this layer-grained ceramic was analyzed. Based on the obtained Weibull modulus, the failure probability of this material was calculated.

Estimations of Weibull modulus

Weibull statistics based on a weakest-link hypothesis are widely used in the characterization of strength distribution of ceramics [3, 4]. Based on the fracture mechanics, the failure in a solid material is controlled by the most serious flaw that is subjected to the highest stress intensity factor. The effect of flaws depends on the size, shape, orientation and location of the flaws. Therefore, only the flaws in tensile zone are effective for the failure of a sample in bending. The common two-parameter Weibull function is given by

$$P = 1 - \exp\left[-\left(\frac{\sigma}{\sigma_0}\right)^m\right] \quad \text{for } \sigma > 0$$
$$P = 0 \quad \text{for } \sigma \leq 0 \tag{1}$$

where P is the failure probability, m is the Weibull modulus and σ_0 is the Weibull characteristic strength corresponding to a failure probability of 63% [4, 6].

The Weibull modulus, m, normally ranges from 5 to 20 for most brittle ceramics [1]. The value of m reflects the stability of the strength data. A low m-value implies a great dispersion in strength and a low reliability. Estimation of Weibull parameters is often affected by many factors. Among them, the numbers of test specimens, N, play an important role [2, 5, 6]. It has been noticed that the estimated Weibull modulus remains almost constant when the amounts of testing samples are over 30. Therefore, testing 30 specimens or above is acceptable for a reliability prediction [2, 6]. The Weibull modulus can be calculated by different methods. In this work, three methods including the linear least-squares method, the moment method and the maximum likelihood technique, were used for comparison. In the first method, Eq. 1 should be rewritten into a linear function [6]

$$\ln\ln\left(\frac{1}{1-P}\right) = m\ln\sigma - m\ln\sigma_0 \tag{2}$$

Eq. 2 is appropriate to apply a least square analysis to obtain the Weibull parameter m and σ_0 , using a set of ranked strength data (σ_i, P_i). The P_i-value is estimated with $P_i = (i–0.5)/N$, where P_i is the fracture probability of the ith ranked specimen.

In the moment method, the Weibull modulus can be directly obtained from the mean value and standard deviation of the strength data [4].

$$m = 1.278(\bar{\sigma}/S) - 0.621 \tag{3a}$$

$$\sigma_0 = \bar{\sigma}/\Gamma(1 + 1/m) \tag{3b}$$

where S is the standard deviation and $\bar{\sigma}$ is the mean value of the strength data. It means that the estimated m-value is in reverse proportion to the relative deviation $S/\bar{\sigma}$. In the light of Eq. 3, if the standard deviation of strength data is close to zero, the estimated m-value will be infinite.

According to the latest international standard [6], the parameter obtained using the maximum likelihood technique is unique (for a two-parameter Weibull distribution). If the testing samples are enough, this method is more efficiently than other techniques. The system of equations obtained by differentiating the log likelihood function for a sample with a single flaw population is given by [6]

$$\frac{\sum_{i=1}^{N}(\sigma_i)^{\hat{m}}\ln(\sigma_i)}{\sum_{i=1}^{N}(\sigma_i)^{\hat{m}}} - \frac{1}{N}\sum_{i=1}^{N}\ln(\sigma_i) - \frac{1}{\hat{m}} = 0 \tag{4a}$$

and

$$\hat{\sigma}_0 = \left[\left(\sum_{i-1}^{N}(\sigma_i)^{\hat{m}}\right)\frac{1}{N}\right]^{1/m} \tag{4b}$$

where σ_i is the maximum stress in the ith test specimen at failure and N is the number of test specimens in the sample being analyzed. The parameters (the Weibull modulus, $\hat{m}$, and the characteristic strength, $\hat{\sigma}_0$) are determined by taking the partial derivatives of the logarithm of the likelihood function with respect to $\hat{m}$ and $\hat{\sigma}_0$, and equating the resulting expressions to zero. Equation 4 should be solved numerically because an analytical solution is impossible. It is very effective to use the estimated m-value from the method of moment as the initial value for the numerical calculation in the maximum likelihood method. A merit of the maximum likelihood method is that the upper bound and the lower bounds of the Weibull parameters could be estimated based on the sample size [6], by using the equations

$$\hat{m}_{\mathrm{low}} = \hat{m}/q_{0.95} \text{ and } \hat{m}_{\mathrm{up}} = \hat{m}/q_{0.05} \quad (5a)$$

$$\begin{aligned}\hat{\sigma}_{0\mathrm{low}} &= (\hat{\sigma}_0)\exp(-t_{0.95}/\hat{m});\\ \hat{\sigma}_{0\mathrm{up}} &= (\hat{\sigma}_0)\exp(-t_{0.05}/\hat{m})\end{aligned} \quad (5b)$$

where $q_{0.05}$, $q_{0.95}$, $t_{0.05}$ and $t_{0.95}$ are bound factors depending on the sample size, for 90% confidence interval, and the values of them are displayed in the international standard [6].

Generally, the standard deviation of strength data is related to the damage tolerance of the test material, i.e., the higher the damage tolerance is, the smaller the standard deviation will be, thus the higher the Weibull modulus should be. Therefore, the damage tolerance is an important factor influencing the Weibull modulus of ceramics. The damage tolerance for brittle materials is controlled by both the crack tolerance factor $K_{\mathrm{IC}}/\sigma_{\mathrm{b}}$ and the energy-dissipation capacity factor E/H [7], so the product of them is defined as a damage tolerance parameter for a quantitative evaluation. Thus, the damage tolerance parameter D_{t} of ceramics was quantitatively evaluated from four basic mechanical parameters [8].

$$D_{\mathrm{t}} = \frac{K_{\mathrm{IC}}}{\sigma_{\mathrm{b}}} \cdot \frac{E}{H} \quad (6)$$

where σ_{b} is the bending strength, K_{IC} the fracture toughness, E the elastic modulus and H the hardness. The D_{t} value represents a resistance to brittle failure and it is convenient for comparing the damage tolerance of different materials. It has been confirmed that the damage tolerance of the quasi-plastic ceramic Ti_3SiC_2 is much higher than that of brittle ceramics, so a higher Weibull modulus is expected.

Experimental and discussion

Polycrystalline Ti_3SiC_2 was prepared by hot-pressing mixed powders of Ti (99%, 300 mesh), Si (99%, 400 mesh), graphite (98%, 200 mesh) in a ϕ135 mm graphite mode at 30 MPa under flowing Ar atmosphere at 1,560 °C for 60 min, and subsequently annealed at 1,400 °C for 30 min. The thickness of the as-fired sample was 20 mm and the density was 4.48 g/cm^3. Over 50 specimens with dimension of $3 \times 4 \times 36$ mm^3 were electrical-discharge machined from the bulk Ti_3SiC_2. The tension surfaces of samples during bending tests were polished using 1 μm diamond paste. Three-point bending tests with 30 mm span were carried out using a crosshead rate of 0.5 mm/min. The measured strength ranges from 406 MPa to 471 MPa, and the ranked strength distribution is shown in Fig. 1. Using the least square analysis, a linear plot based on Eq. 2 is depicted in Fig. 2, from which the Weibull modulus m is 28.1 and the characteristic strength σ_0 is 449 MPa for this layered ceramic.

As a comparison, the Weibull estimates from the three methods are displayed in Table 1. From Table 1, the Weibull moduli calculated from the least square

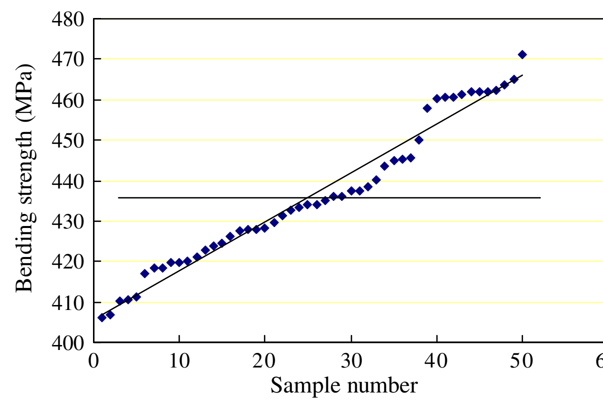

Fig. 1 Distribution of bending strength for 50 Ti_3SiC_2 samples, measured by three-point bending at room temperature

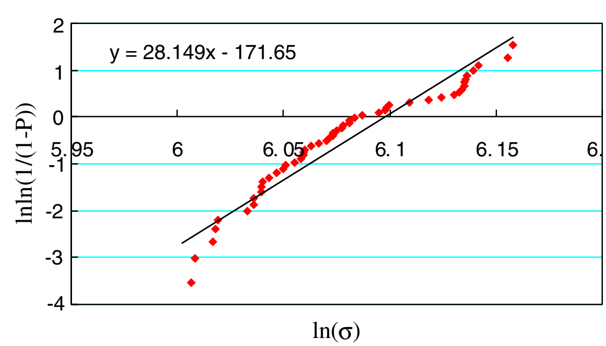

Fig. 2 Weibull modulus evaluated by the linear least squares analysis with 52 specimens cut from a bulk disk. The m-value was estimated as 28.1, using failure probability estimator $P_i = (i–0.5)/N$

Table 1 Weibull parameters estimated by three different approaches for Ti_3SiC_2 ceramics, using three-point bending data of 52 specimens

Estimation approach	Estimated m-value	Characteristic strength (MPa)
The maximum likelihood	24.6	446
The least square method	28.1	449
The method of moment	29.3	445

and the moment method are similar, but the value obtained from the maximum likelihood technique is relatively low. The Weibull parameters estimated by the method of moment depends on only the average value and the standard deviation of strength data, which is very convenient for estimating the Weibull parameters. But the estimates are often a little bit higher than that from other techniques. The Weibull modulus calculated from the least square involves the influence of the estimator of failure probability that related to the ranked strength data. For example, using $P(\sigma_2) = 1.5/N$, where i is the ith datum and N is the number of specimens, the supposed failure probability is $P(\sigma_i) = 0.5/N$ for the first specimen and $P(\sigma_2) = 1.5/N$ for the second specimen even if they have the same strength value. In this work, there are several specimens that showed similar strength value (461.8 MPa), so there is obvious fluctuation in Fig. 2 at corresponding data. The maximum likelihood technique provides stable but the lowest Weibull estimates. Nevertheless, the obtained m-value for Ti_3SiC_2 is higher than that of traditional ceramics whose Weibull moduli range from 5 to 20.

For investigating the reliability of the estimated m-value and the effect of numbers of samples on this value, we randomly selected six groups of specimens with different amounts (sample size N = 12, 20, 30, 40, 46, 52, respectively) for six times, i.e., Weibull parameters were estimated with six different data groups for each sample size. The corresponding m-values and deviations were list in Table 2. It is shown that the stability of the estimated Weibull modulus increases with increasing the numbers of samples, but the mean m-value decreased with increasing sample size. The difference between the upper and lower bounds in the maximum likelihood method and the variation of estimated Weibull modulus by the method of moment are decreasing functions of N, as depicted in Fig. 3. In the method of moment, the error bar of the estimated Weibull modulus becomes very small when the number of the specimens is over 40. It is worth noting that the reliability and stability of strength data is usually in proportion to the value of Weibull modulus. Whereas the reliability and stability of estimated Weibull modulus should increase with increasing number of specimens.

The failure probability varying with the applied stress is analyzed using various estimators and compared to the Weibull statistical prediction with the parameters from the least square and the maximum likelihood approaches. The results are displayed in Fig. 4 which indicates that the failure probability obtained from estimators $P = (i–0.5)/N$ and $P = (i–0.3)/(N + 0.4)$ are almost the same, and the failure probability predicted by Weibull estimates from the maximum likelihood is a little higher than that estimated by the least square method in major stress region.

Since the Weibull parameters estimated by the maximum likelihood method possess a lower and upper bounds [6], the predicted failure probability should also have the lower and upper bounds. To determine the lower and upper bounds, failure probability curves are plotted by using Weibull modulus and the characteristic strength estimated via the maximum likelihood method. The lower and upper bounds of those parameters are also included. For a statistical sample with 52 Ti_3SiC_2 specimens, the Weibull modulus estimated by the maximum likelihood method is 24.6 with a lower bound m_{low} = 20 and upper bound m_{up} = 28.8; and the estimated characteristic strength is 446 MPa, with a lower bound σ_{0low} = 441 MPa and upper bound σ_{0up} = 450 MPa, according to the international standard [6]. Failure probability curves determined by using various combinations of those parameters make up of a zone of failure probability with clear lower and upper bounds. For example, when $\sigma = \sigma_0 = 446$ MPa, the failure probability is in the range from 0.538 to 0.749, instead of 0.632. The failure probability zone for the 52 Ti_3SiC_2 specimens is shown in Fig. 5. It shows that the lower bound of the failure probability is determined by the parameter pair related to the upper limit of the

Table 2 Weibull parameters estimated by two methods and corresponding standard deviation of the Weibull estimates (S_m) varying with the sample sizes, using 6 group data for each sample size

Sample size, N	The maximum likelihood			The method of moment		
	Lower to upper limit	$\hat{m}$	σ_0 (MPa)	$\bar{m}$	Deviation S_m	$S_m/\ \bar{m}$ (%)
12	16.4–36.7	27.6	436	31.33	4.96	15.83
20	18.3–33.5	26.5	449	30.41	2.81	9.24
30	19.1–31.1	25.5	450	30.02	2.23	7.43
40	19.7–29.9	25.1	452	29.73	1.49	5.01
46	19.6–28.9	24.5	446	29.24	0.71	2.41
52	20.0–28.8	24.6	446	29.35	0.15	0.52

The lower and upper limit were calculated by [6] $\hat{m}_{low} = \hat{m}/q_{0.95}$, $\hat{m}_{up} = \hat{m}/q_{0.05}$

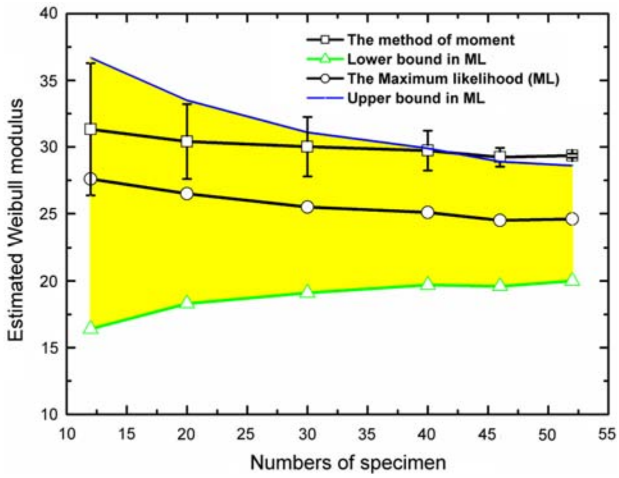

Fig. 3 Estimated Weibull modulus and corresponding standard deviation versus the numbers of specimens of Ti_3SiC_2, showing the increase of reliability with increasing sample size for the m-estimation. The shadow denotes the Weibull modulus distribution zone determined by the maximum likelihood estimation. The data with error bar denotes the estimates obtained from the method of moment, randomly taking data from a sample containing 55 specimens

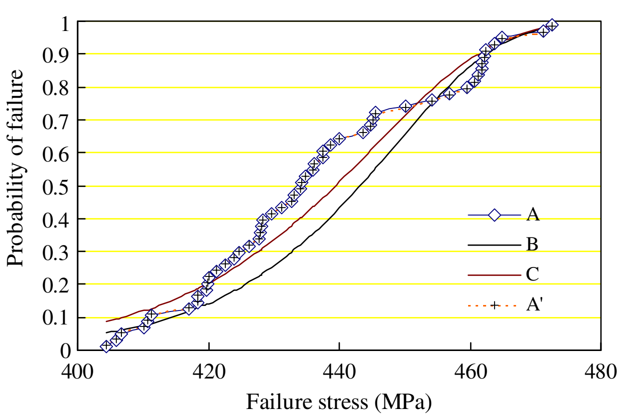

Fig. 4 Comparison in the failure probability predicted by different approaches for 52 Ti_3SiC_2 specimens. A: $P = (i–0.5)/N$; A′: $P = (i–0.3)/(N + 0.4)$; B: Theoretic calculation using Weibull estimates obtained from the least square; C: Theoretic calculation using Weibull estimates obtained from the maximum likelihood

characteristic strength, i.e., $(m_{\mathrm{up}}, \sigma_{0\mathrm{up}})$ for $\sigma \leq \sigma_{0\mathrm{low}}$ and $(m_{\mathrm{low}}, \sigma_{0\mathrm{up}})$ for $\sigma > \sigma_{0\mathrm{low}}$, while the upper bound of failure probability is determined by the lower bound of σ_0.

The stress resulting in 1% failure probability is defined to be the minimum strength, so the minimum strength would decrease with decreasing m-value. For example, the minimum strength of the layered ceramic with high Weibull modulus is close to the characteristic strength, but for brittle ceramic with low Weibull modulus, it is much lower than the characteristic

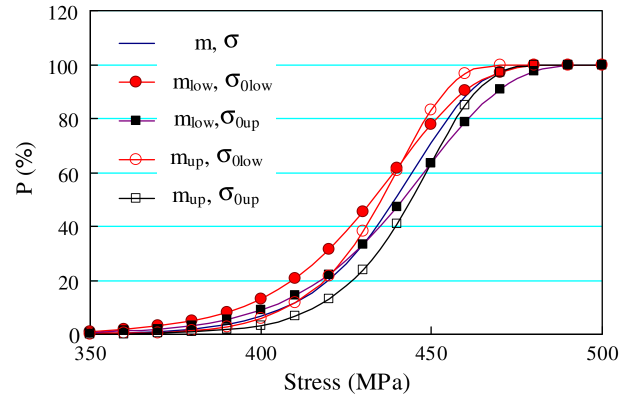

Fig. 5 Failure probability zone consisting of failure probability curves depending on the lower and upper bounds of Weibull parameters estimated via the maximum likelihood technique, for a strength statistical sample of 52 Ti_3SiC_2 specimens

strength. Obviously, in engineering design, minimum strength is a safer parameter than characteristic strength or mean strength, especially for ceramics with low Weibull modulus. The difference between the minimum strength and the characteristic strength increases with the decrease of the Weibull modulus. In addition, it can be seen from Fig. 5 that the minimum strength corresponding to the upper bound curve of the failure probability is lower than that corresponding to the lower bound curve, so the minimum strength determined by the upper bound curve of the failure probability is safer for strength design. Therefore, the minimum strength deduced from the upper limit of the failure probability in Weibull statistics, instead of the mean strength or the characteristic strength, is supposed as a threshold stress for safe strength design of brittle materials. Based on Fig. 5, the minimum strength, $\sigma_{\min}$, can be determined by the lower bounds of Weibull estimates at 1% failure probability via Eq. 2

$$\mathrm{In}\sigma_{\min} = \mathrm{In}\sigma_{0\mathrm{low}} - 4.6/m_{\mathrm{low}} \quad (7)$$

where the constant –4.6 = lnln(1/(1–0.01)). Substituting the obtained Weibull parameter pair $(m_{\mathrm{low}}, \sigma_{0\mathrm{low}}) = (20, 441)$ into Eq. 7, the minimum strength is calculated to be 351 MPa. The determined minimum strength is much lower than the mean strength although the Weibull modulus is high for this ceramic. Obviously, using the minimum strength is safer for strength design. The high Weibull modulus of Ti_3SiC_2 is mainly attributed to high damage tolerance, and the high damage tolerance is due to the nanolayered microstructure. Differing from brittle ceramics, the layered ceramics exhibit quasi-plastic fracture mode with slow crack growth rate and low shear resistance [9, 10]. Beside

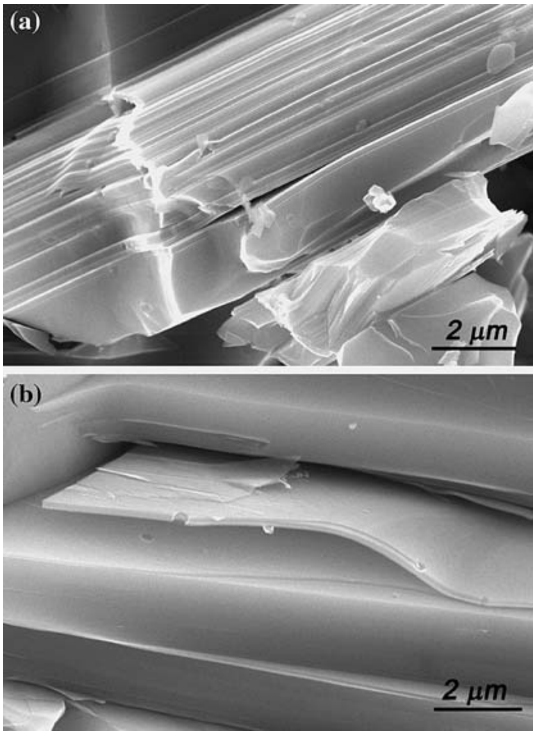

Fig. 6 SEM micrograph of delamination in nanolayer-grained Ti_3SiC_2 ceramic. (**a**) Initiation of the delamination near grain kink; (**b**) a single layer delaminated from a layered grain

conventional energy dissipation mechanisms such as crack deflection and branch, delaminating in grains and the interfacial slipping contribute to the damage tolerance of this quasi-plastic ceramics [11–13]. When a layered grain is subjected to buckling, torque or bending, delaminating occurs easily in the grain. The kinking and delaminating in Ti_3SiC_2 grains under compressive failure have been examined by SEM and are shown in Fig. 6, and this local damage can consume strain energy and results in local stress relaxation.

Indentation tests have exhibited the insensitivity of this ceramic to surface defects [14–16]. The local energy dissipation due to the delamination and slipping leads to local stress relaxation and then improves the damage tolerance. The local softening due to the delamination results in a low hardness and high energy-dissipation in the layered ceramic, so that the damage tolerance and Weibull modulus are greatly improved.

Conclusions

An ultrahigh Weibull modulus, $m = 25–29$, is estimated for Ti_3SiC_2 ceramic by using over 50 specimens in bending tests based on the least square method, the method of moment and the maximum likelihood technique, respectively. It is confirmed that the parameter estimates obtained using the maximum likelihood technique undergo less influence from strength data than other methods, and that the reliability of the estimated m-value increases with increasing numbers of specimens. The lower and upper bounds of failure probability were evaluated via the lower and upper limits of Weibull parameters estimated by the maximum likelihood technique. The minimum strength (here defined as the stress corresponding to 1% failure probability) determined by the upper bound curve of failure probability, instead of the mean strength, is proposed to be the reference strength for safe design of ceramic materials.

High Weibull modulus of the nanolayer-grained ceramic is mainly attributed to high damage tolerance, and the damage tolerance is due to the capacity of local energy dissipation and local stress relaxation caused by delamination in grains and shear slipping at interfaces.

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