Crushing Strength of Fuel Kernels for High-Temperature Gas-Cooled Reactors

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The crushing strength of thorium oxide and thorium-uranium mixed oxide fuel kernels for high-temperature gas-cooled reactors (HTGR) was estimated by application of the Hertz theory of contact to the failure load obtained in a simple crushing test. The strength data were interpreted assuming the Weibull distribution. The crushing strength of the ThO₂ kernels ranged from 1.7 to 1.9 GPa. The strength was found to be closely related to the microstructure of the kernel and to be increased by the kernel properties: small grain size and smooth surface. Fracture would occur in a transgranular manner for the fine-grained kernels and in an intergranular manner for the coarse-grained. Flaws, which are located at or near surface of the boundary of the contact area, act as fracture origins.

KEYWORDS: crushing strength, fuel kernels, HTGR type reactors, thorium dioxide, Hertz theory, failure load, Weibull distribution, microstructure, grain size, fracture origin

I. INTRODUCTION

The mechanical strength of high-temperature gas-cooled reactor (HTGR) fuel kernels is one of important properties, since it is related to their intactness to damage during fabrication processes, the evaluation of the characteristics of the kernel and the strength of HTGR coated particles. The qualities of the kernel such as density, grain size, porosity and flaw distribution may be correlated with the kernel strength. It is expected that the coated fuel particle with a strong kernel may have a higher strength than with a weak one. However, the kernel strength has not been regarded as of importance compared with the strength of the coated particles(1)〜(3) and the coatings such as silicon carbide(4)〜(8) and pyrolytic carbon(5)〜(6).

In the present study, kernel strength was measured using a simple crushing test, which had been applied to the measurement of the coated particle strength.

II. EXPERIMENTAL DETAILS

Crushing tests were conducted on spherical oxide fuel kernels: ThO₂ and (Th, U)O₂. The diameter, density and average grain size of the kernels are summarized in Table 1. Thorium oxide kernels (Nos. 1 and 2) were prepared by the sol-gel process(7). Thorium oxide (No. 3) and (Th, U)O₂ kernels were commercially available. The uranium concentration of the (Th, U)O₂ kernel was 6.3 mol%. The ThO₂—3 kernels were annealed at 1,100〜1,500°C for 5 h in air, so as to examine annealing effects on mechanical properties of the kernels.

Crushing strength was measured at room temperature in air using a soft-type compression test apparatus(3)〜(6). The test kernel was lubricated with a drop of silicon oil at

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its contact faces and then placed between two steel rods with plates of chemically vapor-deposited (CVD) silicon carbide on each contact face. Compressive load was applied at a rate of about 0.75 kg·min⁻¹ (~0.12 N·s⁻¹) until the kernel was fractured. The failure load and the displacement of distance between two steel rods can be read from an X-Y recorder. After test, the pieces of the broken kernel were removed and some were examined under optical and scanning electron microscopes.

Vickers hardness and surface area were measured in order to examine whether or not bulk strength and surface roughness of the kernel affect the kernel strength. The Vickers hardness measurement was conducted at a load of 100 g on the kernel mechanically and carefully polished to the equatorial plane. The BET surface area of the kernel was measured using a Kr adsorption technique.

### III. ESTIMATION OF CRUSHING STRENGTH OF FUEL KERNELS

When a fuel kernel is pressed onto a solid plane surface, a complex stress field is set up in the kernel. It should be noted that the failure load measured by a conventional manner does not necessarily correspond to the strength of the fuel kernel, since fracture should initiate from the regions of maximum tensile stress, which strongly depends on dimensions of kernels. We can estimate the stress within the kernel on the basis of the assumption that the stress is proportional to a mean contact pressure $P/pa^2$, where $P$ is the applied load and $a$ the radius of a circle of contact between the kernel and the SiC plate (see Fig. 1).

The subject of contact between a spherical indentor and a plane sample surface was discussed by Hertz. The Hertzian indentation has recently been applied to the measurements of fracture properties and fracture surface energy of brittle materials, such as TiC, ZrC, VC and WC(9), and nuclear fuel ceramics, such as UO₂(10), ThO₂(11), (U, Pu)CN and (U, Pu)N(12).

For a compression load $P$, the geometry of the contact area between the kernel and the SiC surface is given by:\(^{(9)}\)

$$a = \left\{\frac{3}{4}\pi(k_1 + k_2)PR\right\}^{1/3}$$

with

<table>
<thead>
<tr>
<th>Batch name</th>
<th>Number of kernels</th>
<th>Diameter ($\mu$m)</th>
<th>Density (g·cm⁻³)</th>
<th>Grain size ($\mu$m)</th>
<th>Annealing conditions</th>
</tr>
</thead>
<tbody>
<tr>
<td>ThO₂</td>
<td>1</td>
<td>50</td>
<td>9.76</td>
<td>4.8±0.3</td>
<td>—</td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>50</td>
<td>9.76</td>
<td>3.2±0.2</td>
<td>—</td>
</tr>
<tr>
<td></td>
<td>3</td>
<td>50</td>
<td>9.91</td>
<td>&lt;1</td>
<td>—</td>
</tr>
<tr>
<td>(Th, U)O₂</td>
<td>25</td>
<td>—</td>
<td>0.9±0.2</td>
<td>1,100°C, 5 h</td>
<td>—</td>
</tr>
<tr>
<td></td>
<td>25</td>
<td>—</td>
<td>3.2±0.2</td>
<td>1,350°C, 5 h</td>
<td>—</td>
</tr>
<tr>
<td></td>
<td>25</td>
<td>—</td>
<td>5.4±1.3</td>
<td>1,400°C, 5 h</td>
<td>—</td>
</tr>
<tr>
<td></td>
<td>25</td>
<td>—</td>
<td>7.9±1.1</td>
<td>1,450°C, 5 h</td>
<td>—</td>
</tr>
<tr>
<td></td>
<td>25</td>
<td>—</td>
<td>12.9±2.1</td>
<td>1,500°C, 5 h</td>
<td>—</td>
</tr>
</tbody>
</table>

Table 1 Characteristics of fuel kernels

\(^{(8)}\)
where \( \nu_1, E_1 \) and \( \nu_2, E_2 \) are the Poisson ratios and the Young's moduli of the kernel and the SiC plate, respectively.

Inside the contact area, the stress is compressive and the maximum stress \( Q_{\text{max}} \) is given by

\[
Q_{\text{max}} = (3/2)(P/\pi a^4). \tag{3}
\]

Outside the contact area, the radially tensile stress \( \sigma_\phi \) is created in the kernel such that

\[
\sigma_\phi(y, z = R - \delta) = \frac{1 - 2\nu_1}{3} Q_{\text{max}}(a/y)^2, \tag{4}
\]

where \( \delta \) is the displacement of the kernel (see Fig. 1). Thus the radially tensile stress is maximum at \( y = a \) (the boundary of the contact area) such that

\[
\sigma_{\phi_{\text{max}}} = \frac{1 - 2\nu_1}{2} (P/\pi a^4). \tag{5}
\]

Equation (5) combined with Eq. (1) becomes

\[
\sigma_{\phi_{\text{max}}} = \frac{1 - 2\nu_1}{3} \left( \frac{6P}{\pi^4(k_1 + k_2)^2 R^2} \right)^{1/3}. \tag{6}
\]

The equation indicates that the stress \( \sigma_{\phi_{\text{max}}} \) is proportional to \((P/R^2)^{1/3}\) and therefore increases with decreasing diameter. The \( \sigma_{\phi_{\text{max}}} \) was calculated as a function of \( P \) for the three kernels and shown in Fig. 2, where the \( E_1 \) and \( \nu_1 \) of ThO\(_2\) were assumed to be \(2.5 \times 10^2\) GPa and 0.28, respectively, approximately corresponding to the values of ThO\(_2\) pellets of less than a few percents in porosity\(^{13,15}\). Room temperature Young’s moduli of from \( \sim 3 \times 10^2 \) to \( \sim 4.4 \times 10^2\) GPa have been measured for CVD SiC\(^{16}\). The \( E_2 \) and \( \nu_2 \) of \(3.5 \times 10^2\) GPa and 0.25 were used for the present calculation. Possible errors in the values of \( \sigma_{\phi_{\text{max}}} \) and \( a \) result from uncertainties in the elasticity data used. Assuming that the elastic constants already contain errors of \( \pm 5\% \) and \( \pm 30\% \) for ThO\(_2\) and SiC, respectively, the errors in the \( \sigma_{\phi_{\text{max}}} \) and \( a \) are estimated to be about \( \pm 5\% \).

In the following chapters, the crushing strength is defined as the stress \( \sigma_{\phi_{\text{max}}} \) at the applied load \( P \).

**IV. Statistical Treatment of Crushing Strength**

Crushing strength data for 50 or 25 kernels per batch can be statistically treated. In general, fracture strength in brittle materials does not fit the normal and lognormal dis-
tributions, since there are many irregularly shaped flaws, such as inclusions, pores and large grains, which are intrinsic to the materials. Thus the fracture strength should be evaluated from a statistical flaw-size analysis\(^{17,18}\).

We treated the crushing strength data of the kernel as a member of Weibull distributed population. The relative survival probability \((1-S)\) at a given strength level \(F\) can be defined as

\[
1-S = \exp\left(-\left(\frac{F}{F_0}\right)^m\right),
\]

where \(F_0\) and \(m\) are empirically determined constants. Equation (7) allows us to obtain \(F_0\) and \(m\) by plotting \((1-S)\) vs. \(F\) and fitting using a least-squares method. It can be seen that the plots of \(\ln(-\ln(1-S))\) vs. \(\ln F\) should be a straight line, when the Weibull distribution is reasonable for the present data.

The Weibull modulus \(m\), which characterizes the nature, severity and dispersion of flaws contained in the material, may be related to the amount of scatters observed in the test data. Thus a low value of \(m\) indicates a material in which the severity of flaws is highly variable and characteristic of brittle materials. On the other hand, a high value of \(m\) indicates a uniform distribution of highly homogeneous flaws which exhibits a small scatter in the fracture stresses.

V. RESULTS AND DISCUSSION

1. Comparison of Crushing Strengths

In Table 2, a meadian failure load \(F_m\) (the load at 50% failure probability), which was obtained by fitting the failure loads to Eq. (7), can be written in the form

\[
F_m = \left(\frac{1}{2}\right)^m F_0.
\]

Corrected strength ratios were obtained by dividing the \(F_m\) by \(R^{2/3}\). We think that the Vickers hardness may be proportional to the bulk strength, or the crushing strength. The ratios appear to be in good agreement with the ratios of the Vickers hardness. This suggests that application of the Hertz theory to estimation of the crushing strength of the kernel should be valid.

Table 2 Comparison of meadian failure loads, corrected strength ratios and Vickers hardnesses of kernels

<table>
<thead>
<tr>
<th>Batch name</th>
<th>Meadian failure load (kg)</th>
<th>Strength ratio</th>
<th>Vickers hardness value (ratio)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Load</td>
<td>Corrected</td>
</tr>
<tr>
<td>ThO(_2)</td>
<td>4.81</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>1</td>
<td>3.86</td>
<td>0.80</td>
<td>1.00</td>
</tr>
<tr>
<td>2</td>
<td>1.90</td>
<td>0.40</td>
<td>0.89</td>
</tr>
<tr>
<td>3</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(Th, U)O(_2)</td>
<td>1.68</td>
<td>0.36</td>
<td>0.80</td>
</tr>
</tbody>
</table>

Figure 3 shows the Weibull plots of the crushing strength, or the maximum tensile stress \(\sigma_{\text{max}}\). For the present data, reasonable straight lines were obtained. This indicates that the crushing strength data fits the Weibull distributed population.

Table 3 shows the meadian strength, Weibull modulus and ratio of true surface area \(S_t\) to geometric one \(S_g\) of the kernel. The ratio \(S_t/S_g\) is approximately proportional to surface roughness of the kernel. The literature values of compressive strength of ThO\(_2\) pellets measured at room temperature are from \(\sim0.2\) to \(\sim1.5\) GPa\(^{14}\), depending on porosity.
Table 3 Median strengths calculated by Eq. (6), Weibull moduli and $S_i/S_g$ of kernels

<table>
<thead>
<tr>
<th>Batch name</th>
<th>Median strength (GPa)</th>
<th>Weibull modulus</th>
<th>$S_i/S_g$</th>
</tr>
</thead>
<tbody>
<tr>
<td>ThO$_2$</td>
<td>1.94</td>
<td>16.8</td>
<td>5.35</td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>12.3</td>
<td>5.81</td>
</tr>
<tr>
<td></td>
<td>3</td>
<td>20.7</td>
<td>2.54</td>
</tr>
<tr>
<td>(Th, U)O$_2$</td>
<td>1.55</td>
<td>38.4</td>
<td>1.65</td>
</tr>
</tbody>
</table>

The value of $\sim 1.5$ GPa for the ThO$_2$ pellet with low porosity ($\sim 6.7\%$) is in broad agreement with the kernel strength of $1.7\sim 1.9$ GPa. It can be considered that the kernels have higher strength than the ThO$_2$ pellets in the literatures, since the porosity of the former is very low, probably less than a few percents.

The crushing strength of the ThO$_2$ kernel appears to depend on uranium oxide content, and an addition of UO$_2$ to the ThO$_2$ kernel would lead to decrease in strength.

The Weibull moduli are $10\sim 20$ and $\sim 38$ for ThO$_2$ and (Th, U)O$_2$ kernels, respectively, and much higher than those of the strength of the pyrolytic coatings of SiC(4)(5) and C(6), suggesting that the kernels are highly homogeneous. On the other hand, the modulus decreases as the $S_i/S_g$ increases, i.e. as the surface roughness increases. This would indicate that the surface roughness is closely related to the crushing strength and the strength distribution, and surface flaws may act as fracture origins.

2. Annealing Effects on Crushing Strength

Figure 4 shows the Weibull plots of the crushing strength of the ThO$_2$-3 kernels which are annealed at 1,100 $\sim 1,500^\circ$C for 5 h each. Reasonable straight lines are obtained for the kernels annealed up to 1,400$^\circ$C, but the deviation from the least-squares fitted lines becomes large for those annealed above $1,450^\circ$C and indicates that the data do not necessarily fit the Weibull distribution. Some statistical parameters, the Vickers hardness and the $S_i/S_g$ are summarized in Table 4. The median strength and the Vickers hardness appear to decrease.
with decreasing $S_t/S_g$ (surface roughness), brought about by the annealing treatment. In general, annealing of surface and internal flaws, which act as fracture origins, cause an increase of strength. Thus it would be suitable that reduction of the kernel strength may be mainly caused by the increase in grain size with increasing annealing temperature, since the presence of grain boundaries should lead to an increase in strength, probably according to the Hall-Petch relation (19).

Table 4 shows the effects of annealing on some parameters of ThO$_2$-3 kernels.

<table>
<thead>
<tr>
<th>Annealing conditions</th>
<th>Weibull modulus</th>
<th>Median strength (GPa)</th>
<th>Vickers hardness</th>
<th>$S_t/S_g$</th>
</tr>
</thead>
<tbody>
<tr>
<td>As-received</td>
<td>20.7</td>
<td>1.73</td>
<td>951</td>
<td>2.54</td>
</tr>
<tr>
<td>1,100°C</td>
<td>20.1</td>
<td>1.74</td>
<td>849</td>
<td>2.00</td>
</tr>
<tr>
<td>1,350°C</td>
<td>23.4</td>
<td>1.73</td>
<td>809</td>
<td>1.53</td>
</tr>
<tr>
<td>1,400°C</td>
<td>34.1</td>
<td>1.68</td>
<td>752</td>
<td>1.57</td>
</tr>
<tr>
<td>1,450°C</td>
<td>19.9</td>
<td>1.62</td>
<td>778</td>
<td>1.46</td>
</tr>
<tr>
<td>1,500°C</td>
<td>21.6</td>
<td>1.63</td>
<td>771</td>
<td>1.50</td>
</tr>
</tbody>
</table>

Figure 5 shows the dependences of the Weibull modulus and the $S_t/S_g$ on the annealing temperature. The $S_t/S_g$ decreases gradually with increasing annealing temperature and eventually remains constant (~1.5). This indicates that the surface roughness may be reduced by annealing. On the other hand, the Weibull modulus increases slowly at first and then rapidly, and reaches a maximum at 1,400°C. After the maximum, apparently the modulus decreases rapidly. Assuming that the kernel fracture originates from the surface flaws, the sharpening of the strength distribution up to 1,400°C is probably due to the relaxation of the surface flaws. The decrease of the modulus and the increase of the deviation from the fitted distributions above 1,450°C may be closely related to the rapid increase in grain size, as shown in Table 1. The Weibull plots appear to be composed of two distributions with different $m$ values: ~20 and ~35, being approximately equal to the $m$ values of the as-received and 1,400°C-annealed kernels, respectively. This indicates that the fracture mechanism may change with the grain growth. In regard with the relation between the fracture mechanism and the grain growth, we shall discuss in Sec. V-4.

3. Estimation of Geometry of Contact

The geometry of the contact between the kernel and the SiC plate can be derived from Eq. (1) and the crushing strength data. The displacement of the kernel during loading can be approximately given by $\delta = R - (R^2 - a^2)^{1/2}$. Table 5 shows the observed and calculated displacements. The observed values are a little larger than the calculated ones, since the observed displacement is the sum of the kernel and SiC plate ones. As a result, the geometry of the contact can be approximately estimated using the Hertz theory for the contact between a spherical indenter and a plane sample, indicating the essential validity of the present analysis.

![Fig. 5](image-url)
Table 5  Comparison of observed and calculated values of kernel displacements and contact radii of ThO$_2$-3 kernels

<table>
<thead>
<tr>
<th>Annealing conditions</th>
<th>Displacement ($\mu$m)</th>
<th>Contact radius ($\mu$m)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Observed</td>
<td>Calculated</td>
</tr>
<tr>
<td>As-received</td>
<td>5.52±0.98</td>
<td>3.33</td>
</tr>
<tr>
<td>1,100°C</td>
<td>5.58±1.18</td>
<td>3.38</td>
</tr>
<tr>
<td>1,350°C</td>
<td>6.00±1.02</td>
<td>3.32</td>
</tr>
<tr>
<td>1,400°C</td>
<td>5.60±1.50</td>
<td>3.12</td>
</tr>
<tr>
<td>1,450°C</td>
<td>5.90±0.72</td>
<td>2.91</td>
</tr>
<tr>
<td>1,500°C</td>
<td>5.24±1.02</td>
<td>2.97</td>
</tr>
</tbody>
</table>

4. Observation of Fracture Face

Photograph 1 shows optical micrographs of typical fracture faces of the kernels with fine (ThO$_2$-1 and as-received ThO$_2$-3) and coarse (ThO$_2$-3 annealed at 1,500°C and (Th, U)O$_2$) grains, respectively. Stripes, which would correspond to a propagation of a crack, can be observed in the fracture faces of the fine-grained kernels. However, such is not observed in the coarse-grained, for which grains in the fracture face can be clearly observed. Consequently, it is considered that a transgranular fracture (cracks propagate through grains) may be predominant for the fine-grained kernels (less than $\sim$5 $\mu$m) and an intergranular fracture (cracks propagate along grain boundaries) for the coarse-grained.

![Photo 1(A) ~ (D)](attachment:image)

Photograph 1(A) ~ (D) shows optical micrographs of typical fracture faces of the kernels with fine (A), (B)) and coarse ((C), (D)) grains

The features of the fracture faces of the annealed ThO$_2$-3 kernels (Photo. 2) appear to show a similar dependence on grain size, since the fracture would occur in a transgranular manner up to 1,400°C and in an intergranular manner above 1,450°C. As a result,
the decrease of the Weibull modulus and the increase of the deviation from the fitted distribution above 1,450°C are probably attributed to the appearance of the intergranular fracture, which should be caused by the large increase in grain size.

An optical micrograph (Photo. 3(A)) of the ThO$_2$-1 kernel, to which loading was stopped before fracture, shows two kinds of cracks: radiating and circular cracks. The radiating cracks emanate from the center of the contact between the kernel and the SiC plate, whereas the circular cracks are around the contact area. Photograph 3(B) shows a series of clear arc-shaped lines on the fracture face, being analogous to the stripes as shown in Photo. 1(A). The curvature of each line provides information about the direction of crack propagation and the stress distribution. The crack propagates to an inner region of the kernel, since the direction is from the concave to the convex side of the lines. The fracture origin appears to be located at or near the kernel surface, as shown at higher
magnification in Photo. 3(C). These patterns would indicate that the fracture originates at or near surface of the boundary of the contact area, where the tensile stress is the largest, and finishes as a result of the propagation of the circular crack to the inner region.

VI. CONCLUSIONS

The crushing strength of HTGR fuel ThO₂ and (Th, U)O₂ kernels was estimated using a simple crushing test. The main conclusions drawn from the present study are the following:

(1) The crushing strength is regarded as the maximum tensile stress, which is connected with the failure load using the Hertz theory of contact.

(2) The crushing strength of the ThO₂ kernels ranged from 1.7 to 1.9 GPa, which agreed approximately with the compressive strength of low porosity ThO₂ pellets.

(3) The crushing strength was found to be closely related to the microstructure of the kernel and to be increased by the kernel properties: small grain size and smooth surface.

(4) Fracture would occur in a transgranular manner for the fine-grained kernels and in an intergranular manner for the coarse-grain. Surface flaws are associated with surface roughness and act as fracture origins. The fracture origins are located at or near surface of the boundary of the contact area, where the radially tensile stress is maximum.
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REFERENCES