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<th>Title</th>
<th>Fabrication and Mechanical Properties of Al₂O₃-SiC/TiC/Ni Functionally Graded Materials by SHS/HIP(Materials, Metallurgy &amp; Weldability)</th>
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<td>Author(s)</td>
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Fabrication and Mechanical Properties of $\text{Al}_2\text{O}_3$-SiC/TiC/Ni Functionally Graded Materials by SHS/HIP†

Youping REN*, Junshan LIN**, Yoshinari MIYAMOTO***, Guanjun QIAO**** and Zhihao JIN*****

Abstract

Dense Functionally Graded Materials (FGMs) in the systems of $\text{Al}_2\text{O}_3$/TiC/Ni and $\text{Al}_2\text{O}_3$/SiC/TiC/Ni were fabricated by SHS/HIP. Due to the rapid high temperature heating and the use of effective additives, the grain growth could be well controlled. The residual stress produced in the outer $\text{Al}_2\text{O}_3$ and $\text{Al}_2\text{O}_3$/SiC layers, which was induced by the thermal expansion mismatch between the inner TiC/Ni and the outer ceramic layers, was in the range of $-220$ MPa to $-500$ MPa. Two methods were used to measure the R-curve behavior of FGMs. For the indentation-strength method, the compressive stress of the outer ceramic layers showed steep R-curve behavior. For the single edge notch bend (SENB) method, the toughness of the FGM increased in the compression area and reached its maximum of $33\text{MPa m}^{1/2}$ at the interface of the compressive stress and the tensile stress areas. In the tension area the value of toughness decreased.

KEY WORDS: (FGM; Functionally graded materials) $(\text{Al}_2\text{O}_3$/TiC/Ni) $(\text{Al}_2\text{O}_3$/SiC/TiC/Ni) (SHS/HIP) (R-curve behavior) (Toughness) (Residual stress)

1. Introduction

The residual stress in a material can be induced by anisotropy or mismatch of thermal expansion or phase transformation. The magnitude and the state of residual stress in ceramics have a vital effect on mechanical properties such as fracture stress, fracture toughness and wear resistance①-③. Generally it is tailored to obtain high surface compression with a moderate bulk tension. The introduction of this concept can be traced back to the work of H. P. Kirchner④ in 1979. Some methods have been employed to produce surface compression, including heat treatments such as quenching, application of low expansion coatings, chemical modification of surfaces, and pressure induced phase transformation⑤.

Various symmetric graded materials such as TiC/Ni/TiC⑥ and $\text{Al}_2\text{O}_3$/TiC/Ni⑦ have been fabricated. The results show that it is easy to tailor deep compressive layers by using layered or graded structures which are preferable for restraining the initiation or growth of microcracks. This concept is applied to cutting tools⑧.

In the present study, symmetric FGMs in the system of $\text{Al}_2\text{O}_3$/SiC/TiC/Ni were fabricated by SHS/HIP and their mechanical properties such as strength, apparent fracture toughness and R-curve behavior were investigated. The SHS/HIP is a powerful and energy saving process to sinter FGMs⑨.

2. Experimental Procedure

The starting materials used were $\text{Al}_2\text{O}_3$, TiC, Ni, SiC powders with an average particle size of 0.4μm, 1.4μm, 1.0μm, and 1.0μm, respectively. These powders were wet-mixed in pre-determined compositions for over 48 hours by ball milling, and then dried in a vacuum oven. Each sample was designed to have a symmetric five-layer structure. The compositions of each layer are shown in Fig. 1. The green body was press-formed at 200MPa by CIP and put into a glass capsule and vacuum sealed at 820°C. The sample was covered with BN powder bed in the capsule. Then, the glass encapsulated samples were put into the chemical oven including silicon powders of 40g which is charged in a graphite container. Two pieces of small ignition pellets consisting of Al and Fe$_2$O$_3$ powder mixtures were placed at just below and over the capsule in a chemical oven. The samples were sintered in

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37
Fabrication and Mechanical Properties of Al$_2$O$_3$-SiC/TiC/Ni FGM by SHS/HIP

![Diagram of Al$_2$O$_3$, Al$_2$O$_3$+10, 15, 20 vol.%SiC, Al$_2$O$_3$+46.8vol.%TiC+2.5vol.%Ni, TiC+19.8vol.%Ni+6.5vol.%Mo$_2$C](image)

Fig. 1 Structure and composition of sintered FGM bodies.

The HIP equipment. All samples were fabricated at the HIP temperature of 1150°C for 30 mins under a nitrogen gas pressure of 100MPa. In the course of heating up, the combustion reaction of silicon and nitrogen was induced by the exothermic reaction of thermit pellets at about 1050°C, so that the temperature in the chemical oven could reach as high as 2500°C for a few minutes due to the reaction heat, resulting in rapid densification. The glass capsule was melt, but not flew down and the sample was sintered without reaction with the glass capsule due to the protection of BN powder bed.

The sintered samples were disks of 30mm diameter and 6 ~ 7 mm thickness. They were polished to a diamond surface finish of 3µm. The compressive stress at the surface of FGMs was determined by the $sin^2\psi$-2θ method using an X-ray diffraction peak (416) of Al$_2$O$_3$. Hardness and indentation toughness were measured by using a Vickers hardness-testing machine with an indentation load of 98N for 10 seconds. The indentation-induced crack length, 2C, was measured using an optical microscope and the indentation toughness, $K_{IC}$, was calculated using the following equation:

$$
(K_{IC}/H_v a^{1/2})/(H_v/E\phi)^{2/5} = 0.129(c/a)^{-3/2}
$$

where $\phi$ is a material-independent constant, $H_v$, $E$, and $a$ are Vickers hardness, Young's modulus and half-diagonal length of the indentation, respectively.

The sintered samples were cut into rectangular bar specimens with the size of 2x6x25 mm$^3$. The surfaces of a specimen were ground and the edges were slightly chamfered. The tensile face was polished to a 1µm finish. Flexural strength was measured by means of three-point bending test with a span length of 18 mm and cross head speed of 0.5 mm·min$^{-1}$ using an Instron 1185 machine. The indentation-strength method was used to evaluate the R-curve behavior of FGMs$^{11,12}$.

The indentations with loads from 9.8 to 196N were applied at the center of the tensile face of a sample beam, and subsequently fractured by three-point bending. The indentation strength, crack length and load in bending experiments were used to calculated the R-curve: $K_R = k(\Delta c)^m$, where $K_R$ is the fracture resistance, $\Delta c$ is crack extension, and $k$, $m$ are material constants. In addition, the toughness of FGMs according to the SEBN method was investigated. A diamond blade of 100µm thickness was used to introduce a notch into the sample. The toughness was calculated referring to ASTM Standard E-399$^{13}$. The fracture surface was observed by scanning electron microscopy (SEM).

3. Results and Discussion

Figure 2 gives the fracture surface image of different FGMs. The photos (a), (b), and (c) show the surface layers of the Al$_2$O$_3$, the Al$_2$O$_3$+10vol.%SiC, and Al$_2$O$_3$+15vol.%SiC. Photo (d), (e), (f), and (g) show the interfacial regions of the first/second layer, second/layer, interfacial region of the second/central layers, and central layer of the Al$_2$O$_3$/TiC/Ni system, respectively. Because the temperature as high as 2500°C is maintained only for several minutes, the grain growth is not severe. Also because the SiC particles can refine the microstructure, the grain size of the Al$_2$O$_3$+15vol.%SiC layer was controlled to be smaller than that of the simple Al$_2$O$_3$ layer.

The mechanical properties of different FGMs and monolithic Al$_2$O$_3$ are summarized in Table 1. The compressive residual stress at the surface layers of FGMs rises with the increasing of the SiC content. The reason is that the addition of SiC with a lower thermal expansion coefficient than Al$_2$O$_3$ increases the thermal expansion mismatch between the outer and inner layers. The toughness and strength of the outer layer could be effectively enhanced due to the residual compressive stress. In the case of the Al$_2$O$_3$+20vol.%SiC FGM, however, the sample was cracked when cutting due to a large tensile stress produced in the central layer.

Figure 3 shows indents introduced in the surfaces of two different FGMs and monolithic Al$_2$O$_3$. The effect of compressive residual stresses can be seen in the crack length for an indentation load of 10kg (98N); each crack length in the Al$_2$O$_3$+10vol.%SiC FGM, Al$_2$O$_3$+15vol.%SiC FGM, Al$_2$O$_3$+20vol.%SiC FGM and the monol-
Fig. 2 SEM images of the fracture surfaces; (a) Outer Al$_2$O$_3$ layer, (b) Outer Al$_2$O$_3$-10v.%SiC layer, (c) Outer Al$_2$O$_3$-15v.%SiC layer, (d) Interfacial region of Al$_2$O$_3$/Al$_2$O$_3$-TiC layer, (e) Al$_2$O$_3$-TiC layer, (f) Interfacial region of Al$_2$O$_3$-TiC/TiC-Ni layer, (g) Center TiC-Ni layer.
Table 1  Mechanical properties of FGMs and a monolithic Al₂O₃.

<table>
<thead>
<tr>
<th>Materials</th>
<th>Hᵥ(GPa)(10Kg)</th>
<th>Kᵥc(MPa·m¹/²)(10Kg)</th>
<th>Compressive stress (MPa) (by X-ray)</th>
<th>Strength(MPa)</th>
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<tbody>
<tr>
<td>Al₂O₃</td>
<td>18</td>
<td>4.0</td>
<td>0</td>
<td>400</td>
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<tr>
<td>Al₂O₃ FGM</td>
<td>20</td>
<td>6.0</td>
<td>-220</td>
<td>900</td>
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<tr>
<td>Al₂O₃-10vol%SiC FGM</td>
<td>21</td>
<td>6.9</td>
<td>-340</td>
<td>940</td>
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<tr>
<td>Al₂O₃-15vol%SiC FGM</td>
<td>21.5</td>
<td>7.5</td>
<td>-500</td>
<td>900</td>
</tr>
<tr>
<td>Al₂O₃-20vol%SiC FGM</td>
<td>22</td>
<td>7.71</td>
<td>-500</td>
<td>-</td>
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</table>

Fig. 3  Optical micrographs of indented samples: (a) monolithic Al₂O₃, (b) Al₂O₃ FGM, (c) Al₂O₃-10v.%SiC FGM, (d) Al₂O₃-15v.%SiC FGM, (e) Al₂O₃-20v.%SiC FGM.
thick Al₂O₃ extends to 127μm, 115μm, 105μm, 100μm, and 160μm, respectively.

The residual stress can be determined according to the indentation fracture mechanics for a half penny flaw. The indentation toughness, $K_c$, and the crack length can be related by the following equation:\(^{20}\):

$$K_c = K_c^0 - 2\sigma_t (C/\pi)^{1/2}$$ (2)

where $K_c^0$ is the toughness of a ceramic without stress, $\sigma_t$ is the residual stress in a ceramic and $C$ is the indentation crack length. $K_c^0$ and $\sigma_t$ can be obtained by linear fitting associated with $K_c$ and $C^{1/2}$. Figure 4 shows the plots of $K_c$ versus $C^{1/2}$ of the three FGMs. The indentation loads are 49N, 98N, and 196N. For the Al₂O₃/TiC/Ni, the Al₂O₃-10v.%SiC/TiC/Ni FGMs, and the Al₂O₃-15v.%SiC/TiC/Ni FGM, the apparent fracture toughness rose with the increase of crack length. The calculated values for $\sigma_t$ are listed in Table 2. Both values showed good coincidences.
Table 2 Comparison of calculated and measured residual stress ($\sigma_r$)

<table>
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<tr>
<th>Materials</th>
<th>$\sigma_r$(MPa) Calculated</th>
<th>$\sigma_r$(MPa) Measured</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\text{Al}_2\text{O}_3$ FGM</td>
<td>-190</td>
<td>-220</td>
</tr>
<tr>
<td>$\text{Al}_2\text{O}_3$-10v.%SiC FGM</td>
<td>-260</td>
<td>-340</td>
</tr>
<tr>
<td>$\text{Al}_2\text{O}_3$-15v.%SiC FGM</td>
<td>-370</td>
<td>-500</td>
</tr>
</tbody>
</table>

R-curve behavior arises because additional energy is consumed in the process zone of a propagating crack besides the fracture energy dissipated at the crack tip. The shape of an R-curve reflects the ability of ceramic to tolerate the crack extension and thus the strength reliability. It is very important, therefore, to characterize and understand the R-curve behavior of ceramics as well as the development of materials having an appropriate R-curve behavior. It has been manifested that the surface compressive stress can introduce a steep R-curve in a ceramic by a three-dimensional finite element analysis\(^\text{15}\).

In this study, the compressive stress was induced by the thermal expansion mismatch between the outer and inner layers. Due to the addition of about 20v.% nickel whose thermal expansion coefficient is as high as $16\times10^{-6}/^\circ\text{C}$, an apparent difference in thermal expansion coefficient of $1 \sim 2\times10^{-6}/^\circ\text{C}$ exists between the central and outer layers of the $\text{Al}_2\text{O}_3$/TiC/Ni FGMs, which causes a strong compressive stress of $-220 \sim -300\text{MPa}$ in the outer layers. The central layer of TiC/Ni has a tensile stress of $300 \sim 400\text{ MPa}$, which can be withstood because of the high strength of about 10000MPa.

The fracture resistance $K_f$, and the crack extension length, $\Delta c$, satisfies the following relationship\(^\text{17}\): \[ K_f = k(\Delta c)^m \] (3)

where $k$ is a constant and $m$ is a characteristic exponent that describes the sensitivity of R-curve behavior. When $m$ is zero, $K_f$ is invariant with the crack extension. The exponent $m$ can be obtained from the slope $\beta$ of a log-log plot of post-indentation strength, $S$, versus indentation load, $P$.

\[ m = (1-3\beta)/(2+2\beta) \] (4)

\[ k = Y\alpha(\beta\gamma)^{\gamma}(1-\beta)^{(1-\gamma)} \] (5)

($\alpha$ is obtained from the intercept of the log-log plot; $\gamma = P/a_0^{3\beta+\gamma}$ where $a_0$ is the initial crack length)

The ratio of the crack length $a_i$ and the initial crack length, $a_i/a_0 = [4/(1-2m)]^{(3-2m)/(3-2m)}$ (6)

Table 3 gives the values of, $k$, and, $m$, of the two FGMs and the monolithic $\text{Al}_2\text{O}_3$. Figure 5 shows their R-curves. The FGMs exhibited steep R-curves with the residual compressive stress in the outer layers, that can lead to higher crack growth resistance and damage tolerance.

Figure 6 shows an optical microscope photo of a notched sample. Referring to the standard method for measuring plane-strain fracture toughness of metallic materials, ASTM E-399, the apparent fracture toughness of the grade materials was calculated from following formula:

\[ K_{IC} = P_f S Y(\alpha)/B W^{1/2} \] (7)

where $P_f$ is fracture load, $S$ is span, $B$ is the specimen thickness, $\alpha = a/W$ (a is the notch length and $Y(\alpha)$ is a nondimensional stress-intensity coefficient given by the following equation:

\[ Y(\alpha) = 3\alpha^2/[1.99-\alpha(1-\alpha)(2.15-3.93\alpha+2.7\alpha^2)]/2(1+2\alpha)(1-\alpha)^{-1/2} \] (8)

Figure 7 gives the apparent fracture toughness of the $\text{Al}_2\text{O}_3$-10v.%SiC FGM as a function of the notch depth. The toughness increases with the growth of notch depth in the compressive stress zone and is higher than that of the monolithic materials with compositions of the outer layer (4-5MPam\(^{1/2}\)) and the intermediate layer (7.2MPam\(^{1/2}\)). In the tensile area, the value decreases gradually. At the interface of tension and compression, the apparent fracture toughness reaches its maximum 33MPam\(^{1/2}\). J. Lin and Y Miyamoto\(^\text{9}\) calculated the stress distribution at the tip of a notch in the compression area of the $\text{Al}_2\text{O}_3$/TiC/Ni FGM by FEM. The result shows that the compression stress is over 1GPa around the tip of a notch due to the stress concentration. It is considered, therefore, that such high concentration of compressive stress at the crack tip effectively suppresses the crack initiation and extension from the notch root.
Table 3 The values of k and m.

<table>
<thead>
<tr>
<th>Materials</th>
<th>k</th>
<th>m</th>
</tr>
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<tr>
<td>Monolithic Al₂O₃</td>
<td>11.9</td>
<td>0.112</td>
</tr>
<tr>
<td>Al₂O₃ FGM</td>
<td>40.4</td>
<td>0.195</td>
</tr>
<tr>
<td>Al₂O₃-10v.%SiC FGM</td>
<td>80.5</td>
<td>0.252</td>
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<tr>
<td>Al₂O₃-15v.%SiC FGM</td>
<td>125.6</td>
<td>0.288</td>
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</table>

Fig. 5 R-curves of Al₂O₃-SiC FGMs and monolithic Al₂O₃.

Fig. 6 An optical image of a notched specimen (Al₂O₃-10v.%SiC FGM).
Fabrication and Mechanical Properties of Al₂O₃-SiC/TiC/Ni FGM by SHS/HIP

4. Conclusions

A dense Al₂O₃-SiC/TiC/Ni FGM was fabricated by SHS/HIP. Because of the compressive residual stress created in the outer ceramic layers and induced by the thermal expansion mismatch of the inner and outer layers, the outer ceramic layers were strongly toughened. The compressive residual stress in the surface leads to steep R-curve behavior of FGMs and significantly enhanced crack growth resistance and damage tolerance.

When the notch depth increases in the compressive stress area due to the stress concentration at the root of notch, the apparent fracture toughness evaluated by the SENB method increases sharply and reaches its maximum 33MPam¹/² at the interface of compression and tension fields. Once the notch enters the tension area, the toughness value begins to decrease.

References