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A novel fabrication approach for impact resistance laminated ceramics

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ABSTRACT

A novel approach via tape casting and reactive hot pressing was presented to fabricate impact resistance laminated ceramics, and the designability of soft and hard interbedded microstructure, the controllability of *in-situ* interfaces and the dynamic performance potentials of the laminated ceramics were investigated. Laminated $ZrO–Zr_2CN/Si_3N_4$ ceramics with designable microstructure and controllable *in-situ* interfaces were fabricated by synergistically designing the thickness of the matrix and controlling the heating rate. The dynamic compressive responses of the laminated ceramic were tested with a range of high strain rates from $1.1 \times 10^3\ s^{-1}$ to $3.3 \times 10^3\ s^{-1}$, and the results showed that the laminated ceramics obtain excellent dynamic compressive performance with high dynamic strength of 1.1–2.6 GPa, large pseudoplastic strain of 7.7–12.2% and good energy absorptivity of 23.34–34.59%. Multi-reflection and attenuation of stress wave at interfaces were the main reason of excellent comprehensive performance.

1. Introduction

Ballistic impact test results of laminated composites fabricated by intrusion and infiltration casting, with millimeter-scale ceramic plates, metal layers and weak bonding interfaces, show some degree multi-hit capability and structural integrity [1]. Laminated ceramics with weak interfaces [2,3] have been fabricated as structural ceramics [4,5,6], ultra-high temperature ceramics [7,8], erosion wear resistance ceramics [9] and wear resistant ceramics [10,11] due to well designability, high damage tolerance and high energy absorbing capability of laminar structure. However, laminated ceramics with designable soft and hard interbedded microstructure, which meet all the requirements of the impact resistance concept based on the energy absorption capacity and structural integrity [12], have not been used as impact resistance ceramics.

Up to now, the preparation methods of laminated ceramics are a process combining tape casting [5,8], slip casting [13], centrifugal casting [14] or dough rolling [3] with pressureless sintering [3,13,14], hot press sintering [8] or chemical vapor infiltration (CVI) [5]. Meanwhile, the existing methods retaining weak interfaces require a complex pre-treating process, such as coating [3], slip casting [13] or tape casting [15].

Amount of numerical and experimental studies on impact resistance materials have been done, and the results demonstrate that the capability of laminated materials resistance to impact is greatly affected together by the characteristic of matrix [16–19] and the characteristic of interfaces [1,20–23]. And a thin elastic interface with relatively high modulus [20,22,24] and strong interface bond [25] enhances the multi-hit capability and the efficiency of energy dissipation due to attenuation of stress wave at interfaces. The current methods fabricating laminated ceramics cannot achieve all the requirements, and reactive hot pressing, an *in-situ* processing technique with advantages of low energy requirement, high density and strong bonding interfaces, combined with tape casting is an available way. Research results indicated that the $ZrO_2–Si_3N_4$ system could easily react to form an interface of ZrO or ZrN at the surface of Si_3N_4 particles [26]. Our group found that $Zr–Si_3N_4$ system by introducing some carbon could fabricate laminated $ZrO–Zr_2CN/Si_3N_4$ ceramics, and the laminated ceramics obtained well dynamic compressive properties [27].

In this paper, a simple way combining tape casting with reactive hot pressing (RHP) was used to fabricate dense laminated ceramics with designing the *in-situ* characteristic of matrix and the characteristic of *in-situ* interfaces composed of Zr_2CN or a mixture of ZrO and Zr_2CN by designing the thickness of Si_3N_4 layer and controlling the heating rate. The laminated ceramics possess excellent dynamic performance of large strain, high dynamic strength and good energy absorptivity at various strain rates, and could achieve all the requirements for impact resistance application. Furthermore, much more work remains to be done in the design and optimization of impact resistance laminated ceramics.

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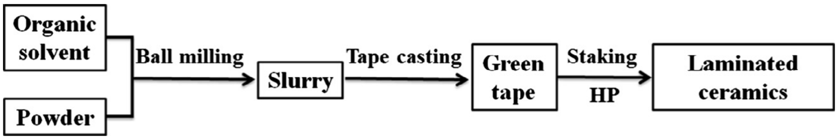

Fig. 1. Fabrication Flow for the laminated ceramics.

2. Experiments

2.1. Fabrication of laminated ceramics

The preparation process of laminated ZrO–Zr_2CN/Si_3N_4 ceramics was illustrated in Fig. 1. Si_3N_4 slurry was made by mixing Si_3N_4 powder (0.7 μm, 40 wt.%) with solvent (mixed isopropyl alcohol and methylbenzene), dispersing agent (triethyl phosphate) and sintering assistant (Al_2O_3 and Y_2O_3) and then milling for 48 h. Subsequently, binder (PVA) and plasticizer (DOP and Glycerol) were added and the slurry was milled for another 24 h. Zr Slurry was made in the similar way at a concentration of 64 wt.% of Zr powder (400 mesh). Tape casting was performed on mini experimental tape casting equipment (Beijing Orient Sun-Tec Co., Ltd.) with a blade height of 0.6 mm.

Laminated ceramics were fabricated by staking green Zr tapes and green Si_3N_4 tapes alternately. Subsequently, organic additives were removed at 600 °C for 1 h in argon flow. Finally, laminated ceramics were fabricated by reactive hot pressing at 30 MPa under argon atmosphere at 1860 °C for 30 min. The heating rate was 30 °C from room temperature to 1200 °C, and the heating rates from 1200 °C to the sintering temperature were 10 °C/min or 17 °C/min. The stacked structure of prepared laminated ceramics and heating rate are listed in Table 1.

2.2. Characterization

The cross surfaces of laminated ceramics were polished to 0.5 μm, then the micro-indentations were obtained at 98 N for 30 s by a Vickers hardness tester (Struers Duramin-A300, Denmark).

The strength of laminated ceramics was evaluated using three-point bending test at a constant crosshead speed of 0.5 mm/min. The dimensions of the samples were 3 mm × 4 mm × 40 mm (height × width × length, respectively.) and the span was 30 mm. The surfaces of samples were polished to 0.5 μm before conducting the three-point bending test.

Table 1
Structural design and heating rate of the laminated ceramics.

Samples	Stacked structure	Heating rate (°C/min)
A	Zr tape/single Si_3N_4 tape	10
B	Zr tape/4 Si_3N_4 tapes	10
C	Zr tape/8 Si_3N_4 tapes	10
D	Zr tape/4 Si_3N_4 tapes	17
E	Zr tape/8 Si_3N_4 tapes	17

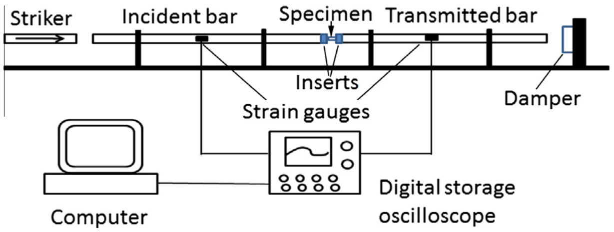

Fig. 2. Schematic diagram of the split Hopkinson bar apparatus for impact test.

The dynamic mechanical responses of laminated ceramics were measured in compression using a split Hopkinson press bar at room temperature. Fig. 2 shows the schematic diagram of the testing apparatus. To avoid the end effect, the inserts of hard SiC with a hardness of 25 GPa were used. The inserts were contacted to the incident and transmitted bars with lubricating oil, respectively, and then the specimen was contacted to the inserts with lubricating oil as lubricant agent. The wave speed and Young's modulus of the incident and transmitted bars are 4840 m/s and 210 GPa, respectively. The dimensions of the specimens were 3 mm × 4 mm × 4 mm (height × width × length, respectively). The loading direction is perpendicular to the layers.

Scanning electron microscopy images were taken from the cross surfaces after mechanical polishing to 0.5 μm of as-sintered laminated ceramics (SEM, S-4700, Japan), and with an energy dispersive spectroscopy (EDS) system.

3. Results and discussions

3.1. Designable microstructure

Comparing with other multilayer materials, such as aramid matrix laminates [28,29], 2D C/SiC composites [30] and laminated composites fabricated by intrusion and infiltration casting [1], the laminated ceramics fabricated by tape casting and RHP are flexible in the design of layer thickness [31].

Laminated ZrO–Zr_2CN/Si_3N_4 ceramics were fabricated by designing the thickness of Si_3N_4 layers (samples A–C with 1 tape, 4 tapes and 8 tapes of Si_3N_4, respectively.), and the microstructure is shown in Fig. 3. With increasing the thickness of Si_3N_4 layers, the structure changes from saw-tooth layer to tidy layer with uniform thickness, and the flexural strength increases from 218 MPa to 350 MPa as well as Young's modulus steadily increases from 136 GPa to 152 GPa and 175 GPa, as listed in Table 2.

The morphology and component distribution of matrix layers have to be designed effectively to absorb energy from incident and reflected waves. So far, multiphase composite ceramics are fabricated by adding the second phase, such as micrometer/nanometer-sized particles, platelets or whiskers [32], and the morphology and mechanical properties are controlled by complex particle pretreatment (surface treatment or dispersion process). However, reactive hot pressing is a simple way to control the morphology of ceramics by controlling the sintering condition.

To fabricated laminated ceramics with different phase distribution in ZrO–Zr_2CN layers and due to lower forming temperature of Zr_2CN [33] than that of Zr–O mixture [34], samples B and D were fabricated with different heating rates of 10 °C and 17 °C, respectively, as shown in Fig. 4. The ZrO–Zr_2CN layer of sample B is composed of continuous Zr–O mixture (gray area) surrounding Zr_2CN phase (light gray area), and little micron-sized SiC particles (black particle) (Fig. 4a). However, the ZrO–Zr_2CN layer of sample D is composed of Zr–O mixture surrounded by continuous Zr_2CN and submicron-sized SiC particle (Fig. 4b).

3.2. Controllable in-situ interfaces

During stress wave propagation through an impact resistance laminated material, the compressive wave will be reflected as a