Fabrication and Evaluation of Mechanical Properties of Reinforced Ceramic Composites by Means of Spark Plasma Sintering Method(放電プラズマ焼結法によるセラミックス複合材料の作製とその評価に関する研究)

著者	Rendon Juan Carlos Angeles
号	1903
発行年	1996
URL	http://hdl.handle.net/10097/7176

氏　　　　　名	レンドン　ファン　カルロス　アンヘレス Rendon Juan Carlos Angeles
授　与　学　位	博士　（工学）
学位授与年月日	平成９年３月２５日
学位授与の根拠法規	学位規則第４条第１項
研究科、専攻の名称	東北大学大学院工学研究科（博士課程）機械工学第二専攻
学位論文題目	Fabrication and Evaluation of Mechanical Properties of Reinforced Ceramic Composites by Means of Spark Plasma Sintering Method （放電ﾌﾟﾗｽﾞﾏ焼結法によるｾﾗﾐｯｸｽ複合材料の作製とその評価に関する研究）
指　導　教　官	東北大学教授　庄子　哲雄
論文審査委員	主査　東北大学教授　庄子　哲雄　東北大学教授　渡邊　忠雄 東北大学教授　嵐　治夫　東北大学助教授　橋田　俊之

論　文　内　容　要　旨

1. Introduction

In the present work, we have aimed at establishing a link between design, processing and evaluation for fiber reinforced ceramic composites. Therefore, initial experiments were performed in order to determine a suitable technique to sinter ceramics and ceramic composites. Our attention was focused on a new sintering process known as spark plasma sintering (SPS). In addition, we analyzed two groups of fiber ceramic composites; discontinuous random fiber and whisker composites, and continuous aligned fiber composites. Mechanical properties (fracture energy G_c and fracture toughness K_c) of fiber/whisker reinforced ceramic composites prepared by SPS process were determined by three point bending tests. Furthermore, this characterization included the determination of the principal mechanism that affects the mechanical response of the composites. Finally, based on the experimental results regarding the composite properties and the toughening mechanism, micromechanical models were proposed as a design tool for predicting the composite mechanical response in terms of fracture energy and strength.

2. Spark plasma sintering of alumina powders and its mechanical evaluation

Sintering experiments were performed to examine the densification behavior of pure Al_2O_3 powders at 1723 K for 5 min and a pressure of 49.0 MPa. It was observed that powder densification occurred earlier and faster in SPS process compared with in hot pressing (HP). This enabled us to sinter Al_2O_3 and ZrO_2 (with and without dopant addition) to full density at a temperature condition of 1873 K. Moreover, SiC powders were sintered to a relative density of 98.5 % at 2673 K. All the specimens were produced in an interval of time as short as 5 min. Microstructural analysis revealed the formation of homogeneous fine-grained structure (< 1.5 μm) in doped ceramics produced by SPS. The strength of the ceramics was determined by modified small punch tests. It was shown for the SPS specimens that the fine grained structure produced high strength at low temperature compared with the mechanical response exhibited by fire vacuum sintered (FVS) specimens. Moreover, significant decrease of strength observed on the specimens with dopant addition at elevated temperatures (beyond 1373 K) may be due to the presence of an intergranular phase which promotes grain boundary sliding. So far, full densification of Al_2O_3 powders without sintering aids has not been achieved by conventional process (HP or FVS). The Al_2O_3 without dopant produced by SPS, particularly showed a constant strength over a wide range of temperature up to 1773 K. The above mentioned results demonstrate that the SPS method is more suitable to produce monolithic ceramics than the conventional processes (HP and FVS).

3. Preparation of discontinuous random SiC fiber/Al_2O_3 composites by spark plasma sintering process, and their mechanical evaluation

SiC fibers with low oxygen content (0.5 wt.%) were used to form fiber reinforced Al_2O_3 composites. In order to achieve dense composites with the desired mechanical performance, experiments were performed to control the relative density of specimens during the sintering process. First, an analysis of the effect of processing parameters (sintering temperature and pressure) was done. The results showed that the composite properties such as density and interfacial shear strength were dependent strongly on the sintering temperature rather than the pressure. The decrease of the fracture energy observed beyond 1723 K was related to the high interfacial shear strength (23.5 MPa) developed at the fiber/matrix interface. Regarding the effect of micromechanical parameters such as fiber length, fiber volume fraction and fiber type on the composite fracture energy G_c, the following trends were observed: specimens produced with fibers of 2, 4 and 6 mm length showed a constant value of fracture energy irrespective of the fiber length. The fracture energy increased with increasing volume fraction of fibers up to 30 vol%. Beyond 30 vol%, a drop of fracture energy was observed due to the agglomeration of fibers and low bulk density. The effect of the oxygen content in the fibers was investigated using SiC fibers with an oxygen content of 12 wt.%. The results showed that the fracture energy of this composite was about half of that of the specimens produced with high purity fibers. The lower fracture energy observed for the high oxygen content is considered to be due to the higher interfacial shear strength brought about by a formation of SiO_2 thin film. All the specimens showed severe thermal microcracking which was induced during the cooling stage. It was also shown that the thermal microcracking severely reduced the load carrying capacity of composites. Microstructural observations revealed that the main mechanisms that controls the fracture energy in these composites were characterized by the fiber/matrix interface debonding, fiber pull-out and the considerable amount of fiber rupture at the matrix crack plane. It was observed that these mechanisms were influenced by the interfacial shear strength in combination with the fiber embedment length and its orientation with respect to the matrix crack plane.

4. Preparation of discontinuous random SiC whiskers/Al_2O_3 composites by spark plasma sintering process, and their mechanical evaluation

In an attempt to increase the load carrying capacity of Al_2O_3 composites SiC whiskers were used as a reinforcement. The thermal microcracking was prevented in all the SiC whisker/Al_2O_3 composites produced by SPS, resulting in higher fracture energy in comparison with the SiC fiber composites. Results on the evaluation of the processing parameters (sintering temperature and pressure) showed a trend similar as that observed for the SiC fiber composites. Moreover, the effect of whisker content and the dopant addition (Y_2O_3) on the composites produced by SPS were investigated. A considerable increase of the fracture toughness (6.5 MPa • $m^{1/2}$) was obtained when the whisker content was increased up to 40 vol%. The incorporation of dopants into the matrix represents one of the methods to control the feature of the interfacial property. The results obtained for 20 vol% whisker composites showed that both fracture energy and toughness were slightly increased when a content of 1 wt.% of Y_2O_3 was incorporated into the matrix (Al_2O_3). These mechanical properties were insensitive to the dopant addition over a content of Y_2O_3 between 1 to 5 wt.%. Microstructural analysis revealed that the main fracture mechanisms were the interfacial debonding and the whisker pull-out. In addition, it was shown that a great amount of whisker oriented at high and low angles contributed to the bridging action at the crack plane behind the crack tip. Furthermore, a distinct feature observed on these specimens was the whisker rupture at the matrix crack plane which was induced by the bending moment.

5. Development of the fiber pull-out rupture model for discontinuous random fiber composites

Recently, several models for discontinuous random fiber composites have been developed. The current models neglect the contribution of the fibers oriented at low angles. These models only consider a normal alignment of the fiber with the matrix crack plane. To account for the contribution of the fiber pull-out and fiber rupture to the composite toughness a model based on micromechanical parameters such as fiber geometry, fiber properties, volume fraction has been developed. The present model employs a bridging stress-crack opening displacement relationship derived for discontinuous random fiber composites. The model assumes fiber/matrix interface controlled by a constant frictional bond stress. It accounts for the effect of fiber orientation which leads to a local frictional effect called snubbing, and more importantly takes into account the effect of fiber rupture observed in both the SiC fiber and whisker/Al_2O_3 composites. The model predicted fracture energy values close to the experimental data, in comparison with the results predicted assuming no fiber rupture. The developed model was used to examine the effect of the composite microstructural parameters on the strength and fracture energy. It was particularly pointed out that there is an optimal length which improves composite properties.

6. Preparation of continuous carbon fiber/composites and a fiber distribution model

The use of continuous fibers in brittle matrices produces highest strength and toughness compared with the discontinuously distributed fibers and whiskers. These composites represent a promising candidate for structural applications despite their processing difficulty. Continuous aligned carbon fiber/SiC preform was sintered to 95% of the theorical density by SPS process at 2223 K for 1 min. Similar preforms were sintered by hot pressing at the same temperature for 60 min. The relative density achieved on this specimen was 90 % of the theorical density. The evaluation of the interfacial shear strength revealed that for composites produced by SPS a high interfacial shear strength (30.5 MPa) was developed at the fiber/matrix interface. This value was two times greater than that observed on the HP specimen. It was shown that the high interfacial shear strength improved the composite strength but lead to a reduction of the capacity of energy absorption. The presence of extensive fiber pull-out and the difference on the fiber pull-out length were clear evidence of fiber strength distribution. Based on the above experimental results for the composites sintered by SPS and HP, the fracture behavior was analyzed by means of a statistical model developed by Thouless and Evans. This model takes into account the effect of the strength distribution along the fiber axis. Composite fracture energy predictions by this model were in good agreement with the experimental values, validating its usefulness.

7. Conclusions

The spark plasma sintering process is a suitable process that can be used to synthesize ceramics and ceramic composites. The micromechanical models proposed here may provide the link between the constituent microstructure and the composite properties. Parametric studies showed that the use of high strength fibers is recommended to achieve sufficient composite mechanical performance in terms of composite fracture energy and strength.

審 査 結 果 の 要 旨

繊維強化セラミックス基複合材料を効率よく開発するためには、破壊靭性値を推定しうる力学モデルを開発し、設計モデルとして活用することが効果的である。加えて、効率の良い合成法を選択し開発することが重要である。本論文は、次世代型の焼結法として登場してきた放電プラズマ法を用い、代表的な2タイプの繊維強化複合材料すなわちランダム配合不連続繊維および一方向連続繊維セラミックス基複合材料を試作し、それぞれの微視破壊を詳細に観察・追跡し、破壊靭性値の推定モデルの開発に関する検討を行った結果をまとめたもので全編7章よりなる。

第1章は緒言である。

第2章では、放電プラズマ法を用いてアルミナ、ジルコニアおよび炭化ケイ素セラミックスの焼結実験を行い、焼結温度・時間ならびに加圧力に関する適切な焼結条件に関する検討を実施している。さらに、常圧焼結ならびにホットプレス法により作製された焼結材と比較することにより、放電プラズマ法が従来の方法と比較してより短時間で機械的特性に優れたセラミックスを作製できる方法であることを示している。

第3章では、ランダム配合不連続繊維複合材料の破壊機構に関する検討を目的として、放電プラズマ法によりアルミナをマトリックスとし、炭化ケイ素短繊維を強化素材とする複合材料の焼結実験を行っている。微視的観察に基づき、破壊機構は繊維のはく離、引抜けあるいは破断により特徴づけられることを示している。さらに、破壊靭性値に及ぼす繊維量、繊維寸法および界面特性の影響を明らかにしている。

第4章では、炭化ケイ素ウィスカーを強化素材として用いることにより熱応力割れを回避できることを示し、さらに、この場合においても破壊機構は短繊維複合材料で観察されたものと同一であることを確認している。

第5章では、ランダム配合不連続繊維複合材料の破壊靭性値を予測するための破壊力学的モデルを開発している。このモデルは観察された破壊機構を十分に踏まえたものであり、繊維のはく離、引抜けならびに破断過程を考慮している。さらに、実験結果と比較することにより、本モデルが繊維、マトリックスならびに界面特性に関するパラメータから破壊靭性値をほぼ推定できることを示している。これは材料設計に関する重要な知見である。

第6章では、一方向連続繊維複合材料を対象として、放電プラズマ法により炭素連続繊維で強化した炭化ケイ素複合材料を合成し、破面観察により、その微視破壊機構はマトリックス内での繊維の破断に起因した引抜けによっていることを明らかにしている。さらに、この微視破壊機構を踏まえて一方向連続繊維複合材料の破壊靭性値を推定するための有用な知見を得ている。

第7章は結言である。

以上要するに本論文は、放電プラズマ法を用いて繊維強化セラミックス基複合材料を高靭化するための合成法ならびに破壊靭性値の推定モデルに関する検討を行い、いくつかの重要な成果を得ており、機械工学ならびに機械材料学の発展に寄与するところが少なくない。

よって、本論文は博士（工学）の学位論文として合格と認める。