Damage mechanisms in silicon nitride materials under contact loading

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論 文 内 容 要 旨

Ceramic materials and more particularly technical ceramics provide very interesting mechanical, thermal and electrical properties suitable in various industrial applications. Among these materials, silicon nitride is considered as an ideal candidate for the development of ball bearings for the automotive and aeronautic industries. This thesis describes preparation and mechanical evaluation of yttria-doped silicon nitride ceramics. The potential applications of the silicon nitride ceramics include tribological elements and components such as ball bearings. Thus, this study focuses on characterization of their mechanical responses and damage mechanisms under contact loading. Notable features of this work are the use of spark plasma sintering in sample preparation, which enables the microstructure control, and sample characterization using focused ion beam (FIB) and transmission electron microscope (TEM), which provides the information for profound discussion on contact damage. Emphasis is also placed on the method to deduce the mechanical properties of the sample by a reverse analysis of the indentation results with the help of numerical simulation. The thesis is composed of four chapters. The outline of each chapter is presented below.

A bibliographic review of research and development for technical ceramics and their potential application areas is given in Chapter 1, focusing silicon nitride ceramics as the candidate material. Then, fabrication methods are examined including spark plasma sintering technique as well as some conventional routes. In addition, previous studies regarding the effect of microstructures on mechanical properties and properties evaluation methods are reviewed and discussed. Based on the

literature review, this chapter sets up two primary objectives for the thesis, (1) Development of methods for deducing and characterizing the nonlinear deformation response due to contact loading, and (2) Elucidation of the mechanisms of contact damage at micro- and nano-scale.

The experimental and numerical methods used in this work are described in Chapter 2. In order to prepare silicon nitride ceramics, a spark plasma sintering technique has been employed throughout this study. Yttria was used as a sintering agent. Silicon nitride ceramics with different grain sizes and microstructures were prepared by controlling the sintering temperature and pressure, and the amount of yttria. A gradual evolution from materials with fine equiaxed grains towards microstructures containing large elongated rod-like grains has been observed with increasing the amount of sintering aid form 1% to 5%.

A novel method for determining the elastic and plastic properties of the prepared silicon nitride ceramics was utilized, which couples instrumented nano-indentation testing and numerical computer simulation to predict the uniaxial stress and strain relationship of the ceramics material. (The tip radius of the indenter was 42 m.) A bilinear law was assumed for the uniaxial stress and strain relationship of the materials tested, and their Young’s modulus E, yield stress $_{y}$ and hardening coefficient K were determined. Hereafter, the numerical method coupled with indentation testing is referred to as inverse identification method. Numerical computations were also executed to study the plastic and damage behavior under contact rolling.

Contact damage was examined by using a bonded-interface (BI) sectioning technique and a focused ion beam (FIB) sectioning technique. Optical microscopy and scanning electron microscopy were used to observe the surface and subsurface contact damage for the specimens prepared with the BI sectioning technique. For the FIB sectioning technique, investigation using transmission electron microscopy was carried out.

Several experimental and numerical results obtained for the dense silicon nitride ceramics prepared by spark plasma sintering at higher pressures are presented in Chapter 3. Dense silicon nitride ceramics have been successfully prepared controlling the amount of yttria and the temperature in the SPS technique. The experimental results reported in this chapter include the effects of yittria contents on the microstructure (in particular, grain size) and phase composition, and mechanical properties such as Young’s modulus, Vickers hardness, flexural strength, and fracture toughness.

In addition to the above-mentioned mechanical properties, the inverse identification method was successfully applied to the dense silicon nitride ceramics in order to determine their yield stress and bi-linear coefficient that represents crucial parameters in the modeling of contact deformation and damage at microscopic scale. The inverse identification method based on the indentation testing of tip radius 42 m has yielded the values of Young’s modulus that compare well with those obtained from the ultrasonic technique, and provided fairly comparable indentation results obtained from a different indenter tip (0.2 mm of tip radius) such as the load vs. displacement records and the surface profiles. The results indicate the validity of the proposed inverse identification method, which provides an effective tool for evaluating the nonlinear mechanical properties of brittle ceramics materials to be subjected to contact loading. It has also been demonstrated that the grain size effect on the yield stress revealed by the inverse identification method is analogues to the Hall-Petch relationship.

TEM observations performed using FIB sectioning technique have revealed that the contact damage in the dense silicon nitride ceramics consists of distributed microcracks and dislocation motion as revealed by the lattice structure. The direct observation of dislocation motion in the ceramics may be considered to be the first in the world. The findings offer a new and important foundation for the characterization of the contact damage in silicon nitride ceramics.

Some experimental and numerical results obtained for porous silicon nitride ceramics are given in Chapter 4. The silicon nitride ceramics with different porosities were successfully produced by adjusting the pressure in the spark plasma sintering process. First, the values of Young's modulus and Vickers hardness obtained for the porous ceramics were correlated with the porosity based on the conventional empirical equations. Then, the effects of the indentation tip radius on the contact damage were examined using the BI sectioning technique, and the observed size effect was interpreted in light of the weakest-link theory and the dimensional ratio of intense strain region to porosity.

The inverse identification method was employed to deduce the uniaxial stress and strain relationship for the porous silicon nitride ceramics. It has been shown that the method allowed us to determine the nonlinear deformation response for the porous ceramics as well as the dense ceramics and also reproduces fairly well the surface profiles in the indented specimens using the different tip radius. The errors in the predicted indent depth were shown to be larger in the case of the porous ceramics compared to those for the dense ceramics. A plausible explanation for this slight discrepancy is that no damage mechanisms is taken into account in the current numerical simulation code used for analyzing the indentation surface profiles.

Experimental observations using the FIB sectioning technique have demonstrated the formation of distributed microcracks in the damage region and their further propagation parallel to the surface due to cyclic indentation loading. As described above, the Hertzian contact damages were examined at macroscopic (large scale) and mesoscopic scales (small scale) by using diamond spheres of different radii on dense and porous materials. At the large scale, experimental observations revealed a remarkable size effect on the damage mode for coarse microstructure: fracture at large scale and quasi-plastic deformation at smaller scale. In addition, the transition radius for a coarse Si3N4 microstructure was experimentally obtained. The difference with values reported in the literature is attributed to the strong assumptions made in previous studies. For the porous materials, it was found that increasing the porosity induced a transition from fracture at the surface to quasi-plastic damage mode at the macroscopic scale. For the reduced radius, the damage consists only of subsurface microcracks initiated from grains boundaries and residual pores. It is worth noting that this damage mode is not easily detectable but may deteriorate the lifetime of components. Therefore, it is essential to avoid the presence of pores or cluster of pores, and this by ensuring a proper control of the processing conditions.

Based on the experimental and numerical results, the outcome of this thesis is summarized in the section of General Conclusions and Prospects. The main results may be highlighted as follows:

1. Novel processing route was developed for preparing silicon nitride ceramics with different microstructures and densities
2. Coupled method of indentation testing and numerical simulation was proposed for characterizing the plastic deformation of the silicon nitride ceramics, and its validity was provided by comparison with experimental results regarding surface profiles of indents
3. TEM observations revealed that the fracture mechanisms in the silicon nitride ceramics under contact loading is the formation of dislocation and microcracks.

The results of this thesis is expected to provide fundamental and useful knowledge in the research community, particularly in the field of tribological applications of silicon nitride ceramics.

（別紙）

論文審査結果の要旨及びその担当者

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論文審査結果の要旨

摺動材料の高性能化は，各種構造物・要素における信頼性・効率等の向上に極めて効果的であることが示されている．窒化ケイ素系材料は，摺動材料としての高いポテンシャルを有するセラミックス材料であり，広範な適用が期待されている．しかしながら，その摺動応用における機械的特性に関する検討ならびに損傷発生に関する現象解明については今度の課題となっていた．本論文は，微細組織を制御した窒化ケイ素系材料の作製法に関する開発を行い，接触負荷下における塑性変形特性の評価ならびに損傷機構に関する検討を行い，それらの結果をまとめたもので全編５章よりなる．

第１章は緒論であり，本研究の背景を述べている．

第２章では，本研究で用いた窒化ケイ素系材料の作製方法と微細組織の制御方法，ならびに試作材の特性評価方法について説明している．試作においては，放電プラズマ焼結法を活用し，焼結助剤として用いたイットリアの添加量，焼結温度・圧力を調節することにより相対密度および結晶粒径の異なる窒化ケイ素系材料を合成できることを示している．また，接触負荷下における試作材の弾塑性変形特性を評価するために，ナノインデンテーション試験と数値解析を組み合わせた逆解析法を考案し，ナノインデンテーション試験を用いて弾性特性，降伏応力，硬化係数を簡易評価する方法を提案している．これらの作製・評価方法に関する提案は，いずれも学術上のみならず実用上も有意義なものである．

第３章では，緻密焼結体における評価結果について述べている．まず，イットリア添加量の微細組織，弾性特性，硬度，破壊特性に及ぼす影響について系統的に調査し，機械的信頼性ならびに損傷解析のための基礎を与えている．さらに，第２章で提案したナノインデンテーション試験に基づく逆解析法を適用し，緻密焼結体の弾塑性変形特性の評価に成功している．逆解析法で評価した弾性係数は超音波法で測定した結果と近い値を与えること，ならびに逆解析法で決定した降伏応力および硬化係数を用いて行った数値解析により，球状圧子インデンテーションによる表面形状を良く再現できることを示し，提案した手法の妥当性を与えている．加えて，接触負荷による損傷機構の解明を目的として，集束イオンビーム加工を用いて，圧痕直下の領域の TEM 観察を行うことより，接触負荷による塑性変形は転位すべりならびに微視き裂の形成によるものであることを初めて見出している．これは，極めて独創的かつ先駆的な成果であると評価される．

第４章では，多孔焼結体における評価結果について述べている．まず，弾性特性および硬度と空隙率との関係を明らかにし，その定式化を行っている．また，種々の寸法を有する球状圧子によるインデンテーション試験結果との比較検討により，弾塑性変形特性の逆解析法を多孔焼結体へも応用することができることを示している．加えて，圧痕形成における球状圧子寸法の影響を最弱リンク説により説明できること，また，その寸法効果は圧痕直下の強変形領域寸法と空隙率から構成したパラメータにより特徴づけることができることを示し，多孔焼結体の摺動材料応用のための基礎的知見を与えている．また，圧痕直下の領域の TEM 観察により，多孔焼結体の接触負荷による塑性変形は微視き裂の発生ならびに進展によることを明らかにしている．これらの成果は，摺動材料開発に貢献しうる貴重な知見を提供するものである．

第５章は結論である．

以上要するに本論文は，微細組織を制御した窒化ケイ素系材料の作製と評価に関する検討を行い，接触負荷下における塑性挙動の特性評価手法の提案ならびにその損傷機構解明に成功したものであり，環境科学ならびに材料工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(学術)の学位論文として合格と認める．