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TOPICAL REPORT

LOW-TEMPERATURE FABRICATION OF TRANSPARENT SILICON NITRIDE

May 31, 1994

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for

OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee 37831
managed by
MARTIN MARIETTA ENERGY SYSTEMS, INC.
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**Research sponsored by the U.S. Department of Energy,
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Summary

There are two approaches to fabricate Si_3N_4 ceramics from amorphous nano-size powder. The conventional method generally utilizes oxides as sintering aids in green ceramics to obtain full density Si_3N_4 . The sintering temperature might be reduced due to the presence of a glass phase, but grain size and pore size will be in micro or submicro-scale and cause a lack of transparency under visible light. The second method which is being explored in this project, differs from the conventional method in the use of gaseous lubricants during compaction but without the use of any extrinsic sintering aids. The primary pore distribution and dense packing are expected not only to provide transparency to the green body but also to improve the final properties of the ceramic and significantly decrease sintering temperature.

The notion that dense compacts of nano-size powders must be transparent prior to sintering has not been emphasized in the ceramic processing literature. Nevertheless, it could be of fundamental importance for fabrication of nano-phase dense ceramics via the nano-size powders route. Indeed, if nano-size particles are densely packed, the spaces between the particles should be of the same size or smaller than the particles themselves. The scale of porosity of such a compact varies on the order of 1/20th or less of the wavelength of visible light. Such compacts should be transparent to visible light, just as porous glass and xerogels are transparent despite having a density of about 50% of theoretical. Translucent or opaque compacts of particles should then have large light-scattering voids surrounded by a large number of nano-size particles. Such large pores cannot be removed via rapid surface and interface diffusion processes characteristic of nano-size particles. During sintering of conventional silicon nitride powders, for example, particle rearrangement and densification is assisted by low-viscosity silicate melts usually present at the grain boundaries and interfaces. In pure silicon nitride powders, however, there are no low-viscosity melts at grain boundaries at sintering temperatures.

Because full density might not be obtained without the addition of extrinsic sintering aids, we plan to determine the maximum density achievable with the nano-powders without sintering aids with the expectation that the small pore size and associated defects may not drastically reduce mechanical properties. This statement is based on our current experience and recent work by Danforth and his colleagues [1,2], Fatrez [3] and Tanaka [4]. Their work has shown that native Si-O layer on silicon nitride powders is sufficient to act as an intrinsic sintering aid. Cryogenic or other lubricant compaction techniques will be used to produce dense compacts (65 % or higher density at 2.5 GPa) from amorphous nano-size silicon nitride powder. Following this, the dense compacts will be hot-pressed at 1.0 GPa and 1000°C to increase contact area between particles. Finally, the compact will be pressureless sintered at 1400°C. This process is expected to promote a strong bonding at the contact area due to solid-state surface diffusion. The density of the new microstructure of silicon nitride ceramic

with nano-scale porosity and grain size is expected to be in the range of 65 - 80 % of theoretical with enhanced optical, electrical, and mechanical properties.

We plan to study selected nano-scale oxide powders to understand the basic phenomena involved during lubricant-aided compaction of powders to high green density and further densification by hot pressing followed by sintering. These studies aided by pore structure analysis using small angle neutron scattering (SANS) and transmission electron microscopy (TEM) will be used to develop an improved understanding of limitations to achieving full densification of nano-scale powders. These data will not only help us develop a new class of materials but also will have far reaching implications in the development of nano-powders processing technology.

Introduction

Often new fields of research in materials science have been stimulated by the synthesis of new types of materials. The production of nano-size particles has recently enabled the synthesis of a new class of ultrafine-grained materials by using non-conventional compaction and sintering of these particles. The resulting nano-phase materials, which may contain crystalline, quasicrystalline, or amorphous phases, can be monolithic metals, ceramics, or composites with properties different or improved from the normal coarse-grained polycrystalline materials. Much of the research completed to-date on the synthesis, characterization, and properties of these new materials has been recently reviewed, [5,6]. The advantages of nano-phase materials have been described elsewhere [6]. Some characteristics and advantages claimed for nano-phase ceramics are as follows: (1) small particle size during synthesis allows increased sinterability at lower temperatures and small residual pore sizes, resulting from a combination of high driving forces and short diffusion distances, avoiding the need for sintering aids; (2) exceptional physical and chemical control during gas-condensation (a common method of powder synthesis) lets the particles maintain clean surfaces during synthesis, allowing subsequent high grain-boundary purity and thus negligible interfacial phase formation; (3) a large fraction of atoms residing at interfaces, almost one-half in the case of a 5 nm grain size, may allow for new atomic arrangements at the interfaces, and thus provide novel and possibly improved ceramic properties. Properties which may be improved through higher grain-boundary purity and the absence of brittle phases therein or small grain sizes include, for example, more efficient deformation mechanisms and more effective crack dissipation than normally available in coarse-grained ceramics. We would like to emphasize that a number of these characteristics and advantages have yet to be proven.

Silicon nitride is an important ceramic with a wide range of structural, electronic and optical applications. In the area of structural applications, the high strength of pure silicon nitride up to 1500°C makes it a very attractive material for use in the ceramic gas turbine engine. It is considered to be one of the leading ceramics for use in the automotive industry which often requires wear resistance and high-temperature stability. The optical transparency, both in visible and infrared ranges, combined with high strength, and excellent corrosion and wear resistance at temperatures from room to 1500°C, make silicon nitride very attractive for use in optical windows and radomes.

All of the above properties are characteristics of pure silicon nitride, Si_3N_4 . In its pure state, silicon nitride exists in three forms: (1) amorphous, (2) α -crystalline, and (3) β -crystalline. The latter two forms have hexagonal symmetry of atomic arrangement and the former has no long-range symmetry. Amorphous silicon nitride crystallizes into α -phase at 1300-1400°C, and α -phase readily transforms into β -phase at 1500-1600°C. All three forms of pure silicon nitride have been claimed to possess excellent mechanical, electronic, and optical properties. These properties have been measured on small single crystals, thin films, or small polycrystalline samples produced using ultra-high pressures and temperatures.

The idea that nano-size particles with their high specific surface area can be sintered to high density at much lower temperatures than their micro-size counterparts is not new. Enhanced sintering of nano-size particles of many oxide ceramic materials has been shown in numerous publications. However, with nonoxide materials, such as silicon nitride, the sintering of nano-size particles has not been successful. The main problem is the fabrication of dense, transparent green compacts from nano-size particles of silicon nitride, which due to their small size, have a tendency to form hard agglomerates.

The feasibility of producing nano-phase Si_3N_4 with improved properties, and ultrafine-grained nano-phase transparent Si_3N_4 by working with amorphous nano-size powders without the use of sintering aids has been investigated in the present work. The approach uses cryogenic compaction of nano-size particles under liquid nitrogen followed by pressureless sintering [7]. This new procedure potentially offers improved reproducibility in manufacturing of ceramic parts because flaws can be easily seen owing to the transparent nature of the green body. This research on amorphous nano-size powders has the potential to decrease sintering temperature from approximately 1900 to 1400°C for fabrication of dense silicon nitride. Early diamond anvil cell results encouraged us to study the possibility of low-cost processing of silicon nitride on a larger scale which involves neither the use of sintering aids nor hot isostatic pressing (HIPing). The low-cost associated with this processing method results from a simplified approach to powder processing which includes shape forming by pressing followed by hot pressing/sintering for densification, but eliminates a number of other conventional steps.

Objectives

The objective of this research is to fabricate transparent Si_3N_4 from nano-size amorphous powders at much lower temperatures than the conventional sintering temperature of approximately 1800-1900°C without using sintering aids. This is a true nano-phase ceramic material rather than conventional ceramic material fabricated from nano-size powders. The primary goal of this research is production of a microstructure containing homogeneous pore distribution, a nano-scale grain size, nano-scale porosity, and improved mechanical properties. Specifically, the following are the major steps planned to accomplish this goal:

- * Design and fabrication of computer controlled equipment for investigation of the rheology of powder compaction under different lubricant conditions.
- * Production of transparent green compacts with 3 mm diameter and 0.3-1.0 mm thickness.

- * Optimization of random packing density by using cryogenic or other lubricant compaction techniques at pressures up to 2.5 GPa.
- * Increase of contact area at interfaces between particles by hot-pressing green compacts.
- * Low-temperature sintering (at 1400°C) to control surface solid-state diffusion along interfaces at the contact area.
- * Characterization of sintered (65-80 % theoretical density) ceramic for optical, mechanical, microstructural, and electrical properties.

It should be noted that a number of these steps are modified from those in the original proposal to effectively utilize the results obtained during the past two years.

Accomplishments

1. Equipment Design

A specially designed novel apparatus for powder compaction was developed to study rheology of fine powder. A hot-pressing device has been developed to generate high pressures and high temperatures that are necessary to produce improved densification.

Room temperature compaction of starting powders is a commonly used procedure in the processing of ceramic materials. The objective of the compaction procedure is to produce a dense green-state compact from the powder that subsequently can be sintered at high temperatures to form a dense ceramic piece. High density in the green-state after dry pressing is of primary importance for achieving high densities after sintering. Large voids, cracks, and other defects in the green-state are not easily eliminated during sintering and their presence in the sintered body reduces the strength of the ceramic. Partial removal of these unwanted large defects usually can be accomplished by using higher sintering temperatures or employing expensive techniques such as hot pressing or HIPing. An understanding of the compaction behavior and rheology of ceramic powders is desirable for the production of defect-free greenware in which particles are packed in the densest possible arrangement.

To study the cryogenic or other lubricant compaction process in more detail and for fabricating larger volume samples, we designed and constructed a novel apparatus. This apparatus [8] is capable of producing 3 mm diameter by 1 mm thick disk-shaped samples under vacuum or in a variety of controlled conditions such as surrounding gaseous or liquid environments and temperatures in the range from liquid nitrogen to room temperature, and pressures up to 3 GPa. The samples are used for measurements and analysis of density, hardness, fracture toughness, transparency, microstructure, and pore-size distribution. In addition, the equipment is used to provide detailed data for the compaction rheology of powders in the form of volume or density of the compacting sample as a function of the applied pressure, the rate of pressure application, processing temperature, and cover gas atmosphere. To provide complete information on the processing procedure, all of the above mentioned parameters are measured continuously and simultaneously.

The equipment is designed for two different modes of operation in the powder compaction experiments: (1) gas lubrication, and (2) liquid lubrication. In the first mode, a small amount

of lubricating cover gas is introduced into the environmental chamber under controlled pressure. The introduced gas condenses on the surface of particles and the powder is compacted under dry or semi-dry conditions. An alternative design of the experimental system is to study liquid lubrication-type compaction of powders as shown in Figure 1. Due to the large amount of data accumulated during an experimental run with this system, a real-time computer control of all measurements and experimental procedures is necessary. For more details, the reader is referred to a recent publication [8].

2. Powder Compaction

Cryogenic compaction was shown to provide significantly higher green density at pressures up to 2.5 GPa.

Forming densely-packed green compacts from nano-size particles is very difficult. Strong aggregation forces, such as the Van der Waals attraction, increase dramatically as particle size decreases. For nano-size particles, the Van der Waals attraction forces can prevent the particles from sliding past each other, and, thereby, promote agglomeration by diffusion across particle contact boundaries during compaction. As a result of aggregation and subsequent agglomeration of primary particles, compacts of nano-size particles usually have low densities after dry pressing. The low density is due to two factors: (1) the presence of large voids in the green-state and (2) inefficient packing of particles in the structure of the green-state. We now know that the use of suitable lubricants can improve the packing properties of the nano-size particles, but, in selecting a lubricant, one is severely limited because of the strongly reacting interface between the particles and the small size of voids in the resulting green microstructure formed by densely-packed nano-size particles. Generally speaking, the diameter of the channels which exist in the structure of densely-packed particles is approximately 1/10th that of the particle size. Therefore, for a 10 nm diameter particle, the channel diameter could be approximately 1 nm. Not many molecules can penetrate such small channels. Since the lubricant must be eliminated from the densely-packed structure prior to sintering, one must select a lubricant which consists of very small molecules. Moreover, the selected molecule must adsorb on the surface of the nano-size particles strongly enough to provide good sliding properties during compaction, but, it must also be eliminated easily after achieving a densely-packed structure. This could be accomplished by heating in vacuum for a reasonable length of time.

Figure 2 shows a maximum random packing density of approximately 64% for nano-size amorphous silicon nitride compacted at less than 2.5 GPa pressure under liquid nitrogen. At the same pressure, for a dry compaction of the same powder, the green density was about 57% of theoretical. These results indicate that compaction under liquid N₂ is an efficient technique to reduce compaction pressure to obtain high packing density and small-scale porosity. The green body produced by liquid nitrogen processing exhibits transparency under visible light which is an indication of nano-scale, uniform porosity. The highest green density of 64% for nano-size silicon nitride is much smaller than that achieved for gamma alumina powder using either liquid nitrogen or pentane as lubricants. The green densities for gamma alumina are in the range of 74% theoretical indicating that we have further possibilities for improving the performance of silicon nitride powder. We recognize the physical and

chemical differences between these powders that may contribute to their processing response [9]. However, the results of gamma alumina appear to indicate experimental conditions under which high green densities can be obtained. This aspect will be pursued in the next few months.

3. Pressureless Sintering and HIPing

The high density green compacts were studied by pressureless sintering and HIPing to achieve additional densification. As the studies progressed we found the need to study the merits of HIPing. The highest sintering temperature was 1400°C since this is expected to be the threshold temperature for initiation of phase transformation and grain growth.

In one series of tests, the green compacts were sintered without pressure at different temperatures from 1200-1400°C under flowing nitrogen gas. The transparency was lost at a sintering temperature of about 1500°C as expected because the phase transformation from amorphous to α -phase is accompanied by shrinkage and grain growth. One possible reason for loss of transparency could be that shrinkage in atomic distances is expected to result in physical shrinkage of the particle and, grain growth is expected to destroy the orderly porosity existing in the green body. The overall result is an increase in the scale of porosity which was estimated to be on the order of the wavelength of visible light (380 - 780 nm). The measurements of phase composition by x-ray diffraction, specific surface area by gas adsorption, and microstructure by TEM indicate that the phase transformation and grain growth initiate at approximately 1400°C. Also, it should be noted that the samples were not densified at this pressureless sintering temperature. The maximum Vicker's hardness of about 6 GPa was obtained at a compaction pressure of 2.8 GPa and 1400°C sintering temperature.

In another series of experiments, HIPing at 200 MPa using nitrogen gas as a pressure transmission medium was attempted to further densify high density green compacts of nano-size Si_3N_4 at 1200, 1300, and 1400°C. The samples were encapsulated in glass and embedded in BN, BN+ Si_3N_4 , or Si_3N_4 as packing powders. In one case, at 1300°C with BN+ Si_3N_4 as packing powders, some densification was observed.

Phase transformations from amorphous to α -phase and β -phase for pressureless sintered and HIPed compacts were studied by TEM and XRD. The transformation point for both processing methods is above 1400°C. In general, no significant densification was observed by HIPing.

In a series of experiments, we compacted a gamma alumina nano-powder at room temperature to investigate the influence of green density on sintering temperature and densification. The reason for using the alumina powder was the readily available sintering and densification data, which is not available for silicon nitride powder. These results presented in Figure 3 show the following:

- a. Low density green compacts cannot be densified appreciably by sintering below 1000°C
- b. High density green compacts were sintered to higher than 90% density at 1000°C. In other words, one can reduce the sintering temperature significantly by using high green density compacts.
- c. Higher compaction pressures resulted in a higher green density.

These results demonstrate that one can achieve a high degree of densification at a low sintering temperature of 1000°C, provided sufficiently high green density was obtained during compaction. These results clearly illustrate that the nano-size powders have to be compacted to a high green density before attempting to densify them at temperatures lower than the conventional.

4. Design of a Hot-Pressing Device with an Internal Tube Furnace

A "belt apparatus" to generate high pressure and temperatures for hot pressing green compacts has been designed and constructed.

As described above, experimental results indicate that phase transformation and grain growth at sintering temperature above 1400°C will generate larger scale porosity according to TEM analysis). The transformation from amorphous to α -phase causes a volume shrinkage of the original particle which destroys the packing in the green body and produces large-scale porosity. If the size of grain and porosity are controlled in the nano-scale, the sintering temperature must be in the range of 1400°C or lower.

To avoid phase transformation and grain growth, the new approach to fabricate transparent, true nano-phase Si_3N_4 is to hot press the densely-packed green compact before pressureless sintering. The contact area, ΔS divided by the total area S of a single particle, is given by

$$\frac{\Delta S}{S} = \frac{P_0}{\text{Hardness}} \quad (1)$$

where P_0 is pressure. Therefore, the contact area of particles can be increased by elevating either the hot-pressing temperature to reduce hardness of Si_3N_4 or the hot-pressing pressure. Since the solid-state surface diffusion is more likely to happen than the volume diffusion at low-temperature, the surface diffusion along contact areas can form a very strong bond between the particles if the contact area is large enough.

To accomplish this, it was necessary to design a special apparatus to generate high temperature and high pressure. A prototype of this apparatus, as shown in Figure 4, called belt apparatus, was designed and fabricated to carry out hot pressing of Si_3N_4 dense compacts. The belt apparatus may be described as a cross between a piston and cylinder and Bridgman anvils. It combines massive support for the pistons with a central die for the sample. The pressure generation is through the compression of a gasket material in the apparatus.

The belt apparatus evolved from Bridgman anvils by first incorporating a well in one of the anvils and compressing a pyrophyllite assembly. However, the pressure limit is quickly reached when the pyrophyllite compresses to its limiting volume and halts further compression inside the well. A major advance in the design was made with the realization that the conical profile has the compression of the cone for a given vertical compression stroke. Thus considerable compression of the sample may be obtained before the limiting volume of the gasket is reached. The final step was to double-end the apparatus producing a device consisting of two anvils or pistons compressing a sample restricted in a central die.

The piston and die are constructed from hardened tool steel for the prototype. The piston has a high pressure bearing face approximately 6.4 mm diameter. The contact diameter between the piston surface and the load-bearing surface is approximately 25 mm. The initial conical portion with a half-angle of 30° to the vertical extends for about 5 mm and then there is a smooth curve to the upper surface. The die has an internal profile such that the tapered piston can move freely into it.

The gaskets are constructed separately for the upper and lower halves of the apparatus. They consist of two conical pyrophyllite gaskets sandwiching a hollow steel cone. The use of an annealed steel cone is important because it prevents the thick pyrophyllite gaskets from breaking at low pressures and helps to lower the compressibility of the combination. A cylindrical region approximately 5 mm diameter and 8 mm high is left for the high pressure cell assembly which usually consists of samples and heaters inside a pyrophyllite cylinder.

This device is expected to generate pressures up to 1 GPa and temperatures up to 1000°C for study of sintering Si_3N_4 green compacts. Experiments are in progress on the study of densification behavior of Si_3N_4 at different temperatures and pressures using nano-size powder compacts with different green densities.

Future Plan

Based on the information developed in this project, it is necessary to maximize the random packing density of nano-size particles before firing. The packing density will directly influence the final mechanical and optical properties, as well as the scale of porosity and its distribution. Unfortunately, we have found that an increase of packing density in the green body will cause lamination inside the greenware for submicro-scale particles because of the weak Van der Waals forces. Therefore, on a relative scale, a strong bond cannot be formed between these particles. However, for nano-size powders, theoretical analysis and experimental results indicate that the Van der Waals force is strong enough to form a dense compact without lamination. This is an important and major difference between the behavior of micro- and nano-scale powders.

Hence, obtaining a dense green compact is one of the most important aspects of this project. We will explore additional possibilities for increasing the packing density using selective lubricants that have not been explored previously. Some examples of proposed lubricants are gases such as ammonia, hydrogen, and helium. In addition, we will study the application of

small organic molecules. The primary criteria for selection of a lubricant will be its ability to increase green density in excess of 65% of theoretical and ease of its removal by heating.

Since silicon nitride suffers from a small coefficient of self-diffusion, we would like to pursue compaction and densification of other nano-scale powders, especially oxides. The purpose of such a study would be to develop a fundamental understanding of the rheological and compaction behavior of nano-scale particles in the presence of gaseous and liquid lubricants. The results of these studies will enable us to not only understand fundamental limitations to nano-scale powder compaction, but also will likely lead to improvements in silicon nitride powder processing which is the major theme of this project.

To increase contact area between the particles, hot-pressing is necessary before pressureless sintering at 1400°C. This is one of the most feasible methods to elevate mechanical properties at low-sintering temperatures while keeping the ceramic transparent and maintaining true nano-phase properties. The pressure and temperature of hot pressing will be controlled below 1 GPa and 1000°C. Using the specially designed hot-pressing device, the green body (about 64% of theoretical density) will be further densified. The compact will be surrounded by boron nitride packing powder to enhance thermal conduction and isostatic pressure environment.

Pressureless sintering temperature is controlled to be less than that of the crystallization of the amorphous phase to α -phase and grain growth. Therefore, the sintering temperature is considered to be below 1400°C for the primary amorphous silicon nitride powder. The samples will be analyzed by various techniques to study the phase transformation and grain growth, and optimization of hot-pressing and pressureless sintering conditions.

We plan to emphasize characterization of pore morphology, pore size distribution, and changes in these parameters as the material undergoes transformation from a green compact to a sintered ceramic. The primary techniques to investigate porosity are SANS and TEM. This information will be a guide to densification studies.

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Figure Captions

- Figure 1. Configuration of liquid lubrication-type powder compaction assembly, (1) moving piston, (2) die, (3) cryogenic container, (4) powder, (5) fixed punch, (6) metal foil, (7) hardened steel frictional load transmission ring disk, (8) hardened steel load transmission disk, (9) heat insulating disk, (10) heat insulating ring disk, (11) hardened steel frictional load transmission ring disk, (12) frictional load cell, (13) positioning and heat conducting disk, (14) frictional load transmission pistons, (15) flange, and (16) plastic positioner.
- Figure 2. Green density of silicon nitride as a function of compaction pressure and lubrication for a nano-size silicon nitride powder.
- Figure 3. The density of green and sintered compacts of gamma-alumina as a function of sintering temperature and compaction pressure.
- Figure 4. Configuration of a hot-pressing device, called "belt apparatus".

Appendix- Paper published

- * [1] Wei Chen, Alexander Pechenik, S. J. Dapkunas, G. J. Piermarini and S. G. Malghan, *"Novel Equipment for the Study of the Compaction of Fine Powders"*, J. Am. Ceram. Soc., 77, 1005-1010 (1994).

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Figure 1.

Green Density vs. Pressure for Nanosize Si_3N_4

Figure 2.

Figure 3.

Figure 4.

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