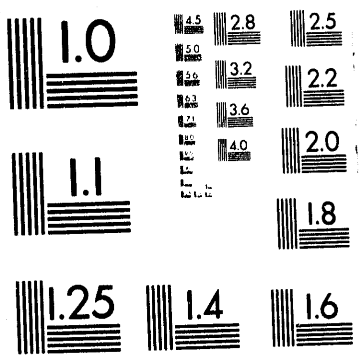
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DOE/ER/45286--6

Final Report on the project entitled

FABRICATION, PHASE TRANSFORMATION STUDIES, AND CHARACTERIZATION OF SiC-AlN-Al_2OC CERAMICS

(Grant No. DEFG0287ER45286)

Submitted to

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February 28, 1994

ABSTRACT

The attached document is the final report on the project entitled 'Fabrication, Phase Transformation Studies, and Characterization of SiC-AlN-Al_2OC Ceramics'. It is well recognized that structural ceramics based on refractory carbides and nitrides are potential candidates for both low and high temperatures applications. Structure property relations in many carbides and nitrides have been investigated mostly in the as-fabricated materials with the exception of Si_3N_4-based materials in which morphology and desired phases are achieved through post-sinter annealing treatments. However, phenomena such as precipitation within the matrix and the growth of second phases have generally not been investigated in most carbides and nitrides. This is primarily due to the fact that mass transport processes in many of these materials are very slow. The SiC-AlN-Al_2OC system is very interesting in this regard. It has been shown, for example, that SiC-AlN and AlN-Al_2OC pseudobinaries form homogeneous solid solutions at elevated temperatures (typically above 1900^oC) which undergo phase separation at lower temperatures. The morphology and the kinetics of phase separation are different in the two systems although it has been shown that phase separation can occur by the formation of modulations (or uniform, fine precipitates) as well as by cellular precipitation depending upon the composition/annealing temperature conditions.

While the two pseudobinaries exhibit some similarities, they also exhibit some key differences. For example, the kinetics are much faster in the AlN-Al_2OC system compared to the SiC-AlN system. Also, in the SiC-AlN system, it is seen that phase separation occurs by the formation of modulations. In the AlN-Al_2OC system, however, tiny precipitates form rather quickly. An important question is whether the phase separation in the AlN-Al_2OC system does or does not occur by the formation of modulations. It is possible, for example, that this is the case and the faster kinetics may have so far revealed only the later stages of phase separation. The other important question relates to the direction of modulations/orientation of precipitates and the effect of coherency strain energy. In the AlN-Al_2OC system, the coherent precipitates are disc-shaped with the axis along [0001]. In the SiC-AlN system, the modulations occur along a direction ~43^o off the c-axis. The presence of very fine precipitates suggests the possibility of nanocomposites.

I. INTRODUCTION

Much work has been done on Si_3N_4-based structural ceramics in which alloying with other constituents has been found to be beneficial for the ease of processing, the development of microstructure and generally improved properties (although the presence of low melting grain boundary phases are often detrimental). By comparison, relatively limited amount of work has been reported on the alloying of SiC-based materials with the notable exception of solid solutions with isostructural (2H) AlN and Al_2OC. The potential for incorporation of SiC-based structural ceramics in elevated temperature applications is well known. High creep resistance (low creep rates), high hardness and good oxidation resistance are the key attributes of SiC and it excels in these properties in comparison to Si_3N_4. At the same time, however, properties such as high toughness and lower coefficient of thermal expansion currently make Si_3N_4 the leading candidate for numerous applications. It is nonetheless generally accepted that for temperatures higher than about 1300°C, the material of choice may likely be SiC or materials containing SiC. SiC-based materials are important from an applications standpoint and interesting from a scientific standpoint. High creep resistance, high hardness, and good oxidation resistance are the reasons for practical interest. At the same time, tendency to form solid solutions with other isostructural materials at high temperatures and undergo phase separation at lower temperature is the source of the scientific interest.

Cutler and coworkers [1] were the first to report that the 2H crystallographic polymorph of α-SiC (lattice parameters a = 3.076 Å, c = 5.048 Å [2]) forms an extensive solid solution with isostructural AlN (a = 3.114 Å, c = 4.986 Å [3]) and Al_2OC (a = 3.19 Å, c = 5.09 Å [4]). In analogy with sialons, these solid solutions were termed sicalons. X-ray diffraction (XRD) was the only analytical tool employed their work. Later work by Rafaniello et. al. [5] showed that the relatively small differences in the lattice parameters of the three constituents makes it difficult to distinguish a solid solution from a multiphase mixture on the basis of XRD alone. Indeed, hotpressed samples were often inhomogeneous and samples had to be annealed at rather high temperatures before single phase solid solutions could be formed. Further, annealing at lower temperatures (≤ 2000°C) led to the formation of two phase mixtures, both in the SiC-AlN and AlN-Al_2OC pseudobinaries [5,6]. The SiC-AlN system has been studied in somewhat greater detail due primarily to its potential for application as a high temperature structural ceramic. Work of Rafaniello et. al. [5,7] and Zangvil & Ruh [8,9,10] demonstrated that: (1) Single phase solid solutions can be formed in the SiC-AlN system, and (2) Annealing at lower temperatures leads to the formation of a two phase mixture consisting of a SiC-rich and an AlN-rich solid solutions of the same structure. On the basis of this, it was concluded that a miscibility gap exists in the SiC-AlN pseudobinary system [7,8]. The formation of a solid solution at high temperatures (≥ 2000°C) and its decomposition to form varied microstructures at lower temperatures offers an excellent opportunity to investigate structure-property relationships and to explore the potential of these materials from an applications standpoint.

From a scientific standpoint, SiC-AlN as well as $AlN-Al_2OC$ systems are particularly attractive since they offer an opportunity to investigate several fundamental aspects of phase stability and phase transformations involving long range diffusion in these covalently bonded refractory materials. It is well known that diffusion in these covalently bonded materials is generally very sluggish implying that the kinetics of phase transformation must be very slow as well, as observed. This means annealing must be carried out for long periods of time. As a result, these materials exhibit excellent microstructural stability even at elevated temperatures, a key requirement for high temperature applications.

We have conducted research on a project entitled 'Fabrication, Phase Transformation Studies, and Characterization of $SiC-AlN-Al_2OC$ Ceramics' under DOE funding. The principal focus of this work has been on the study of phase transformation, microstructure development and elevated temperature creep, although some effort has been devoted towards exploring room temperature mechanical properties of selected materials. Fabrication has been largely conducted by hot pressing even though many of the compositions can be densified by pressureless sintering. The hot pressing route has been selected to ensure the attainment of full density with a fine microstructure. Most of the work has been conducted on the SiC-AlN and the $AlN-Al_2OC$ pseudobinaries. Over the past three years, the following refereed papers have been published, accepted for publication or submitted for publication.

List of Publications:

1) "High Temperature Creep and Cavitation of Aluminum Nitride", Z. C. Jou and Anil V. Virkar, *J. Am. Ceram. Soc.*, **73** [7] 1928-1935 (1990).

2) "Morphology of Phase Separation in $AlN-Al_2OC$ and SiC-AlN Ceramics", S. Y. Kuo and Anil V. Virkar, *J. Am. Ceram. Soc.*, **73** [9] 2640-2646 (1990).

3) "High Temperature Creep of SiC Densified using a Transient Liquid Phase", Z. C. Jou, A. V. Virkar, and R. A. Cutler, *J. Mater. Res.*, **6** [9] 1945-1949 (1991).

4) "Phase Separation in SiC-AlN Pseudobinary System: The Role of Coherency Strain Energy", J. Chen, Q. Tian, and A. V. Virkar, *J. Am. Ceram. Soc.*, **75** [4] 809-821 (1992).

5) "Damage-Resistant SiC-AlN Layered Composites with Surface Compressive Stresses", R. Sathyamoorthy, A. V. Virkar, and R. A. Cutler, *J. Am. Ceram. Soc.*, **75** [5] 1136-1141 (1992).

6) "Effect of Coherency Strains on Phase Separation in the $AlN-Al_2OC$ Pseudobinary System", J. Chen, Q. Tian, and A. V. Virkar, *J. Am. Ceram. Soc.*, **76** [10] 2419-2432 (1993).

7) "High Temperature Creep of Polycrystalline Silicon Carbide - Aluminum Nitride Ceramics", Z. C. Jou and A. V. Virkar, submitted to *J. Mater. Res.*, (1992).

Copies of the above publications are included as Appendices 1 through 6 in this report.

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7) W. Rafaniello, 'Fabrication and Characterization of Silicon Carbide Alloys: The Silicon Carbide - Aluminum Nitride System', Ph. D. Dissertation, University of Utah, June (1984).

8) A. Zangvil and R. Ruh, *J. Am. Ceram. Soc.*, **71** [10] 884-890 (1988).

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