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Publication Date
1978-08-01

To be Submitted to the
JOURNAL OF THE AMERICAN
CERAMIC SOCIETY

LBL-8029
Preprint c.2

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August 1978

Prepared for the U. S. Department of Energy
under Contract W-7405-ENG-48

GLASS-FREE GRAIN BOUNDARIES IN BeSiN CERAMICS

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The high temperature loss in strength of nitrogen ceramics is well-known and has been attributed to the formation of a continuous glassy film at grain boundaries during fabrication(1,2). Softening of the glass at high temperatures enables grain boundary sliding to occur, drastically reducing strength. Improvements in strength have been achieved by strict control of grain boundary chemistry either by the reduction of the impurity contents of the glass(3) or by the addition of more refractory glass forming elements such as yttrium(4). More dramatic improvements in high temperature strength would however be expected if nitrogen ceramics could be fabricated without the formation of an intergranular glassy film.

Observations made using dark field and lattice fringe imaging in the transmission electron microscope have shown that a continuous film of glassy phase does exist at boundaries in Si_3N_4 samples prepared with MgO(5) and Y_2O_3(6) additives. The purpose of this note is to report observations that indicate that in the Be_3N_2-Si_3N_4 system, nitrogen ceramics with glass-free boundaries can be produced.

Grain boundaries in two samples with compositions corresponding to $BeSiN_2$ and $Be_4Si_3N_{10}$(15R) along the Be_3N_2-Si_3N_4 tie line of the Be-Si-O-N system were examined using high resolution dark field imaging. The samples were prepared by hot presssing Si_3N_4, Be_3N_2 powder mixes as described by

Huseby, et al[7]. Thin sections of the bulk samples were prepared by the usual techniques of sectioning, grinding and ion beam thinning.

The dark field technique used to examine the grain boundaries (desribed elsewhere in detail)[8] consists of excluding all Bragg reflections from the objective aperture and allowing only diffuse intensity from any glassy film at the grain boundaries to contribute to the image. Boundaries imaged using these conditions will then appear as a bright line in the dark field image only if a glassy film exists at the boundary. Care was taken to maximize any contrast present by tilting the boundary until it was edge on and by recording images with several positions of the objective aperture. This type of contrast is seen in Fig. 1 in which a 15Å film between two Si_3N_4 grains in a sample in MgO sintered Si_3N_4 has been imaged. Corresponding images of boundaries in the BeSiN samples showed no such contrast in most cases, (Fig. 2), and only a very faint line at a few boundaries. Where faint contrast was observed estimations of the boundary width from the lines intensity indicated a width of less than 3Å. This suggests that the contrast arises from the disordered boundary structure and not from a distinct glassy phase.

These observations indicate that grain boundaries are free of any glassy phase. This result was further supported by the observations that no pockets of glassy phase had formed at three grain junctions and that the grains were epiaxed with curved boundaries forming equilibrium angles at three grain junctions.

Further work is required to understand how these ceramics are formed but these preliminary experiments clearly show that certain nitrogen ceramics can be densified without the formation of a glassy film at the grain boundaries. This in turn should lead to materials with better high temperature strengths.

ACKNOWLEDGEMENTS

We are grateful to Dr. L. J. Gaukler, Max-Planck Institut fur Metallforschung, Stuttgart, for providing the samples. This work was supported by the National Science Foundation and the Division of Materials Science, Office of Basic Energy Sciences, U. S. Department of Energy.

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FIGURES

Fig. 1. (a) Bright field, (b) dark field images of a 15Å film between two Si_3N_4 grains in a MgO sintered Si_3N_4.

Fig. 2. (a) Bright field, (b) dark field images of a grain boundary in $BeSiN_2$. Bright contrast is absent at the boundary indicating absence of any intergranular film.

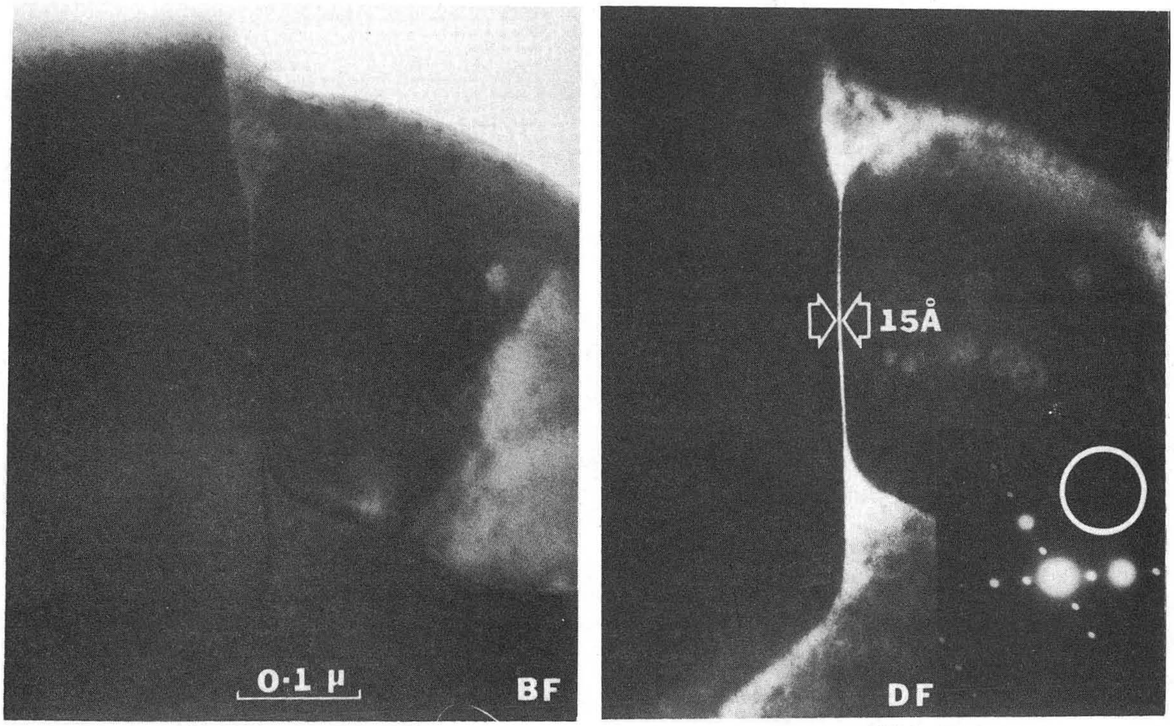

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Fig. 1

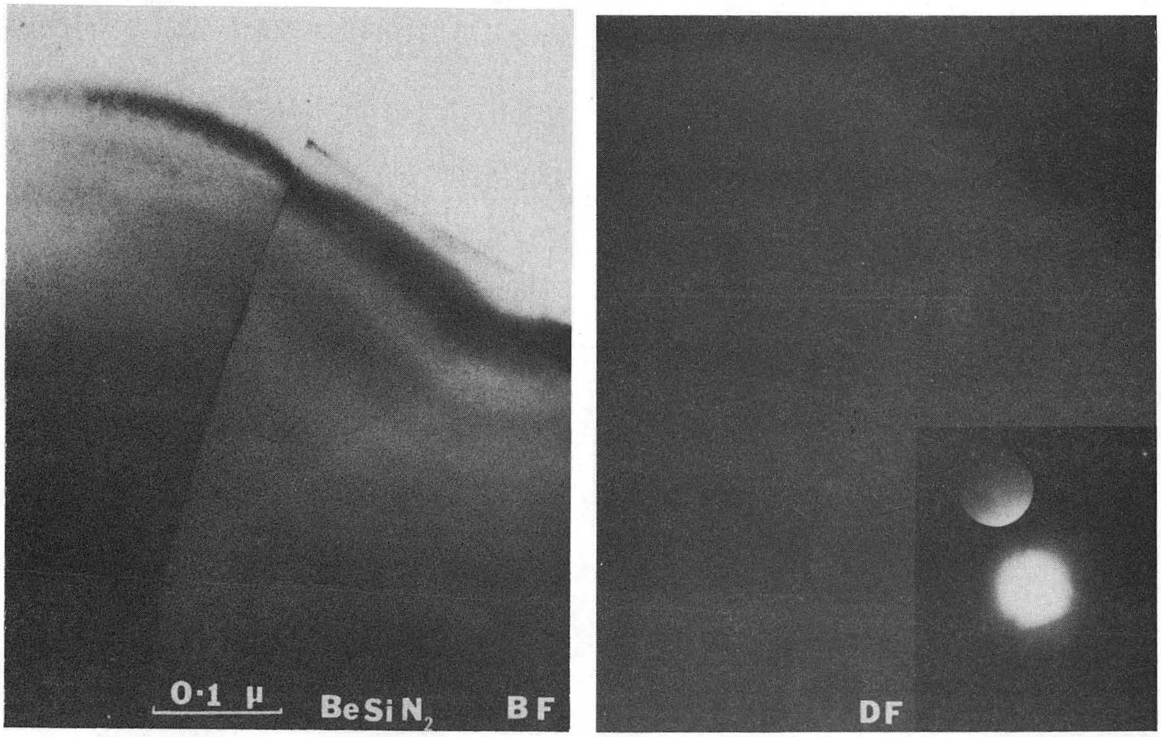

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Fig. 2

This report was done with support from the Department of Energy. Any conclusions or opinions expressed in this report represent solely those of the author(s) and not necessarily those of The Regents of the University of California, the Lawrence Berkeley Laboratory or the Department of Energy.

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