Ceramic Joining by Selective Beam Deposition

J. V. Tompkins, B. R. Birmingham, H. L. Marcus
Center for Materials Science and Engineering
The University of Texas at Austin
Austin, TX

Abstract

Current methods of joining of ceramic components may compromise the strength, chemical resistance, or high temperature properties of the resulting ceramic parts. A new method of joining, Ceramic Joining by Selective Beam Deposition, creates an all-ceramic joint between two or more ceramic components through selective decomposition of a gas precursor. An all-ceramic joint not only preserves the valuable properties of the ceramic materials joined, but may be tailored to match the coefficient of thermal expansion of the original material(s). The added material may be the same as one or both of the joined materials, or may be a composite material. This preliminary work explores the effect of scanning speed and precursor pressure on Selective Beam Deposition of silicon carbide using tetramethylsilane.

Introduction

The need to join ceramic components has driven the development of techniques such as brazing with metals or low temperature binders, ultrasonic joining of green pieces prior to firing, or friction welding. The following describes a new process, Ceramic Joining by Selective Beam Deposition, which involves placing ceramic objects together in a desired configuration within a reactive gas environment and scanning a laser beam over the contact area of the parts. The laser beam causes a gas reaction and produces new ceramic material in the area to be joined. Since gas reactions can produce advanced ceramics, the process eliminates the need for low temperature binder phases which often have low resistance to temperature, chemical corrosion, or abrasion. An all ceramic joint may also avoid problems with thermal expansion as well as wetting in the interface. Since gas reactions may occur well below melting or sintering temperatures the process minimizes thermal stress. It also works on ceramics which decompose below their melting point, such as silicon nitride. Additionally, the process can join materials with very different melting temperatures (for instance, a ceramic and a metal). Control of gas compositions allows deposition of composite materials within the joint, either to wet two different phases more effectively or to mitigate thermal expansion mismatch between joined materials.

The following work uses tetramethylsilane ($Si(CH_3)_4$) as the precursor gas . Pyrolysis of tetramethylsilane (TMS) occurs according to the following overall reaction [1]:

$$Si(CH_3)_4\ (g) \Rightarrow SiC + 3\ CH_4\ (g)$$

Previous work in Selective Area Laser Deposition (SALD) has demonstrated that deposition rates up to 6 mm/min may occur where a focused laser beam strikes a substrate surface. Recent SALD work has included deposition of pyrolytic carbon [2,3], silicon carbide, silicon nitride [4,5], titanium [6], and nickel [7]. In the case of silicon carbide, growth rates and hardness values indicated that selective deposition offers a viable method of joining advanced ceramic materials.

Procedure

The equipment used for evaluating the joining process is similar to that used for SALD [8] with Fig. 1 illustrating specific differences between the two. A rotating shaft holds sections of ceramic tube together within a vacuum chamber containing TMS vapor at the desired pressure. A focused beam from a laser strikes the adjacent surfaces of the tube sections, decomposing the TMS to produce silicon carbide in the joint. A flask containing liquid TMS provides vapor at pressures up to 600 Torr at room temperature. Auxiliary gas lines provide H_2 and N_2 as necessary. To provide forced convection at the deposition point, the chamber contains a small fan.

The chemical and thermal processes occurring at the joint during deposition resemble those shown in Fig. 2. TMS diffuses to the heated area at the surface where it decomposes to form silicon carbide and methane. At the same time, methane diffuses away from the surface. A bevel in the ceramic tubes at the contact area increases the area available for adhesion and allows a flush surface after the addition of silicon carbide.

Previous SALD work has shown that the growth rate and mechanical properties of deposited silicon carbide depend on variables such gas composition, scan rate, and laser power. Growth rate increases in a roughly linear fashion with TMS pressure [9]. With this in mind, the following work used TMS pressures of up to 350 Torr, as pressures over 350 Torr cause excessive formation of a solid by-product on the walls of the vacuum chamber. Earlier work with Selective Area Laser Deposition also revealed that scan rate affects the morphology of deposited material [10]. Finally, in the past growth rate has increased with increasing laser power. Actual process temperature measurements require a non-contact method which can distinguish a circular region approximately 0.5 mm in diameter, which the current system does not include. Consequently, we cite laser power in Watts and the calculated $1/e^2$ spot size in lieu of surface temperature at the deposition point.

Ceramic Joining by Selective Beam Deposition introduces parameters in addition to those listed above for SALD. Chief among these are the physical properties of the materials to be joined. The reflectivity and absorptivity of the joined materials in addition to those of the material(s) being deposited dictate the coupling of the laser beam to the deposition surface, and hence, the beam power required. Since these properties vary with the wavelength of laser light used, the materials selected may determine the type of laser which is appropriate. In this work, we considered alumina and silicon carbide objects for joining with deposited silicon carbide. Alumina possesses low intrinsic absorptivity for both the CO_2 and Nd-Yag lasers available, while silicon carbide showed high reflectivity for the CO_2 laser in preliminary trials. Both materials also have different thermal conductivities. To find a suitable material/laser combination, both materials were tested for deposition in a fixed spot as well as in a moving beam with both lasers. Also, the porosity of the material to be joined affects both the optical properties and the thermal conductivity. Therefore, substrate porosity was included as a variable in the above tests.

Another variable introduced by the joining aspect of the process is the geometry of the edges of the material to be joined. A simple 45° bevel was selected as a standard geometry for all trials. However, it is anticipated that varying bevel geometry will have an effect on beam absorption, heat conduction, mass transport, and adhesion of the deposited material.

After determining suitable conditions for joining experiments, the next goal was to examine the effect of TMS pressure and scan rate on the process while holding other variables constant. The Selective Beam Deposition process involves many transport, kinetic, and thermodynamic phenomena which make predicting the effect of deposition

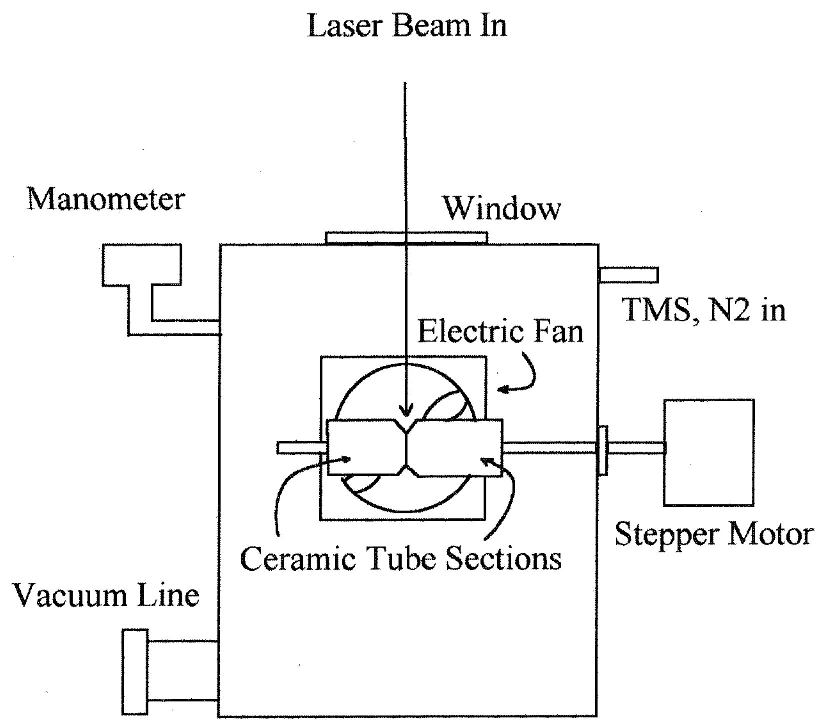

Fig 1. Schematic view of equipment used to evaluate Ceramic Joining by Selected Beam Deposition.

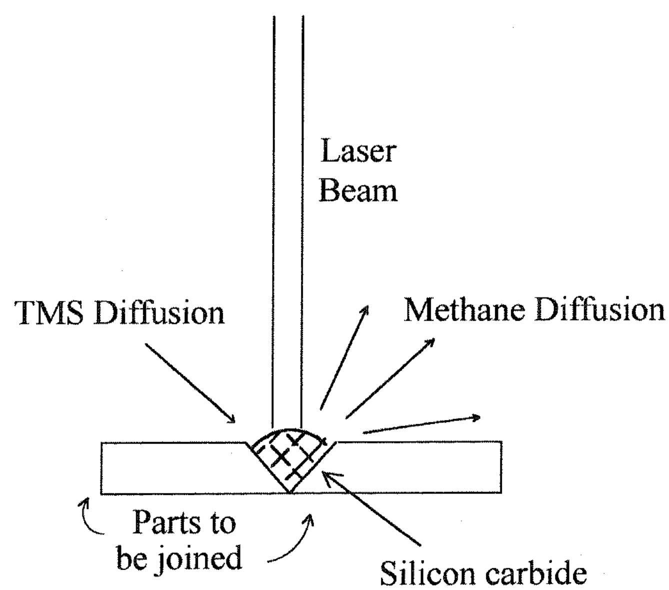

Fig. 2 Detail of deposition area during joining process.

conditions very difficult. In view of this, the experiment spanned a wide range of both gas composition and scan rate to find trends which might guide further experiments. An 18mm diameter alumina tube was placed in the chamber with an initial TMS pressure of 350 Torr. A 12 Watt beam from a Nd-Yag laser was focused to a 0.5 mm spot on the tube, and a stepper motor rotated the tube at 2 RPH for a period of 15 hours. As material formed on the tube, the effective diameter of the tube increased, leading to a proportional increase in scan speed as the deposit thickness increased. Simultaneously, a data acquisition system recorded the change in total pressure inside the chamber as the deposition progressed. The resulting structure after a number of hours would be a disk with many layers grown at different conditions. Each layer can be observed for growth rate and uniformity, and if desired, hardness. The radial position of a given layer indicates the scan rate and gas pressure during the growth of that material. Although the effects of gas composition and scan rate could not be evaluated independently, this technique provided an easy method to view a wide range of deposition conditions to evaluate deposition trends.

Results

Experiments performed with the two ceramics at various porosities with the two available lasers indicated that neither the CO_2 laser (30W) nor the Nd-Yag laser (150W) caused deposition on fully dense silicon carbide or alumina. Alumina with a porosity of 20% or greater absorbed sufficient heat from the Nd-Yag beam to cause deposition of silicon carbide, but did not work with the lower power CO_2 beam. Silicon carbide with 50% porosity worked with both lasers. With these results, 80% dense alumina tubes were selected for joining experiments with the Nd-Yag beam as shown in Fig. 1 above. However, attempts to join beveled tubes at gas pressures of 50 and 350 Torr did not produce integral parts. At 50 Torr, the joint showed good adhesion and uniformity, but suffered from a lack of deposited material in a longitudinal line down the center of the joint. At 350 Torr, the deposited material also showed good adhesion, but grew in an uneven, rod-like fashion. The lack of deposited material at lower pressures resembles a type of growth previously observed in SALD work, and may be due to depletion of the reactant in the center of the deposition.

The final experiment evaluated a wide range of pressures and scan rates to find a case where reactant concentration was sufficient to avoid depletion in the center of the deposit yet not high enough to produce rods or projections. The result was a disc of uniform thickness of approximately 2 mm with clearly distinguishable layers as shown in Figs. 3a and 3b. The initial layers show evidence of non-uniform, rod-like growth. Following layers become more uniform as the deposition time increased. TMS partial pressure is estimated by assuming the total pressure consists of unreacted TMS and the methane byproduct from the overall reaction $Si(CH_3)_4\ (g) \Rightarrow SiC + 3\ CH_4\ (g)$. Layer thickness vs. radial position was measured for three different radial sections. The decreasing thickness of layers over time initially appeared to result from both the decreased residence time as the radius increases and the decrease in reactant concentration. However, Fig. 4 shows the growth rate as a function of TMS pressure by dividing layer thickness by the residence time at a particular radius. The highest rates actually occurred several hours into the experiment after considerable depletion of TMS. Aside from the information available from the material produced at various conditions in the disk itself, the initial layers in the disk adhered to both ceramic tube sections, successfully bonding them together.

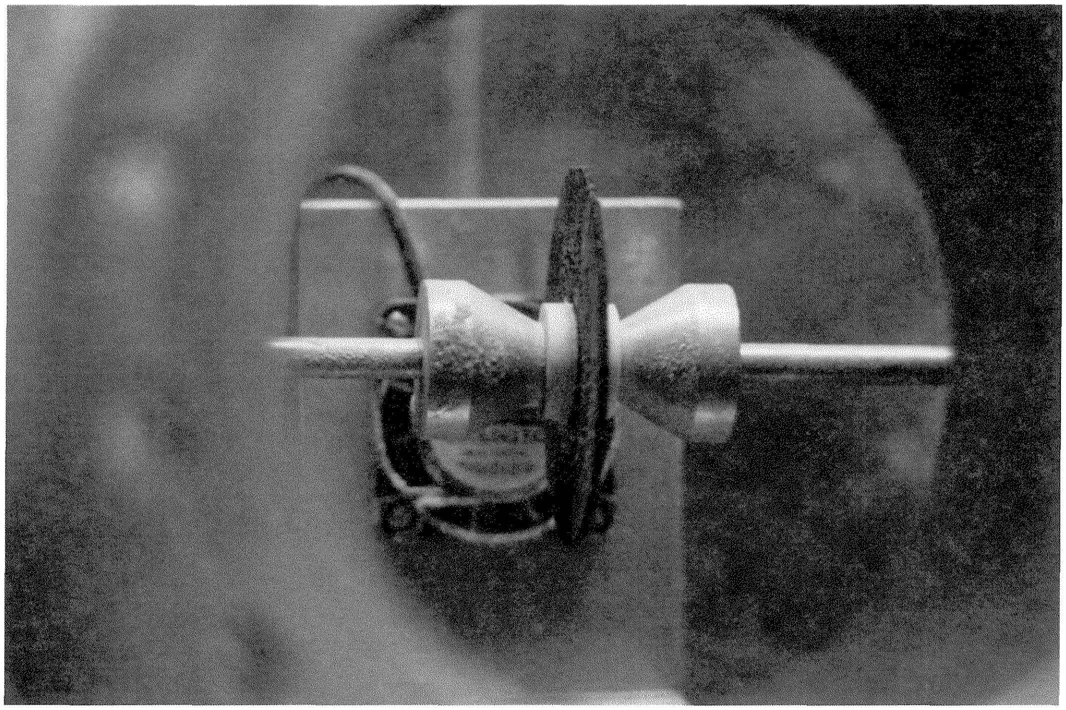

Fig. 3a Completed disk from extended deposition experiment before removal

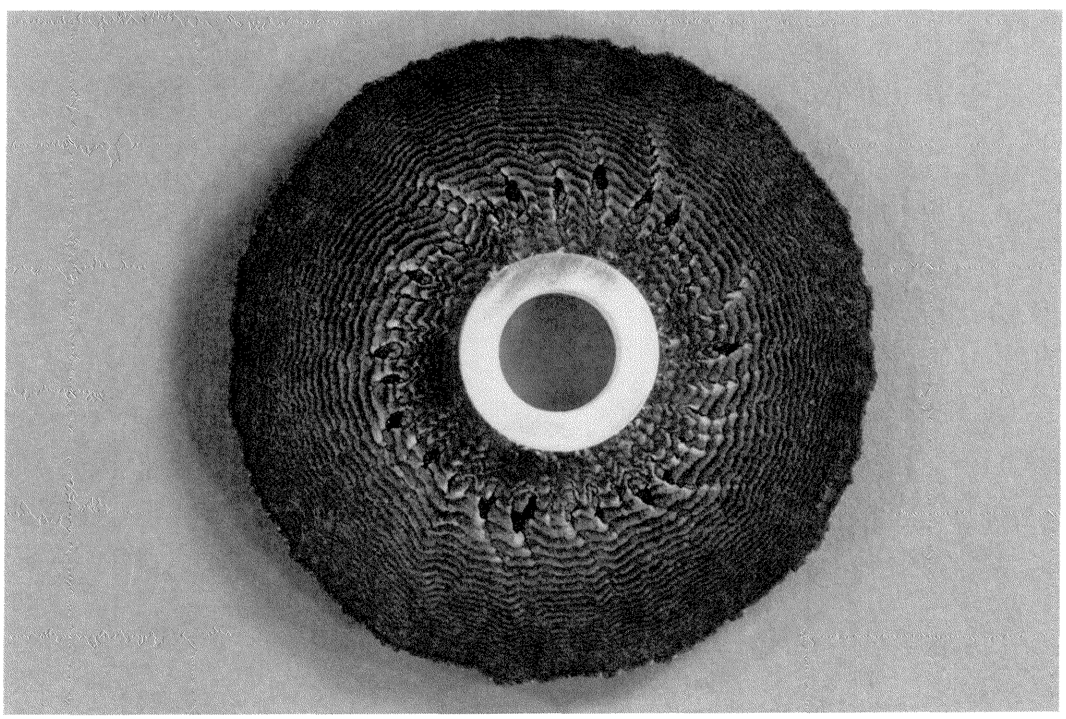

Fig. 3b Detailed photograph of above disk

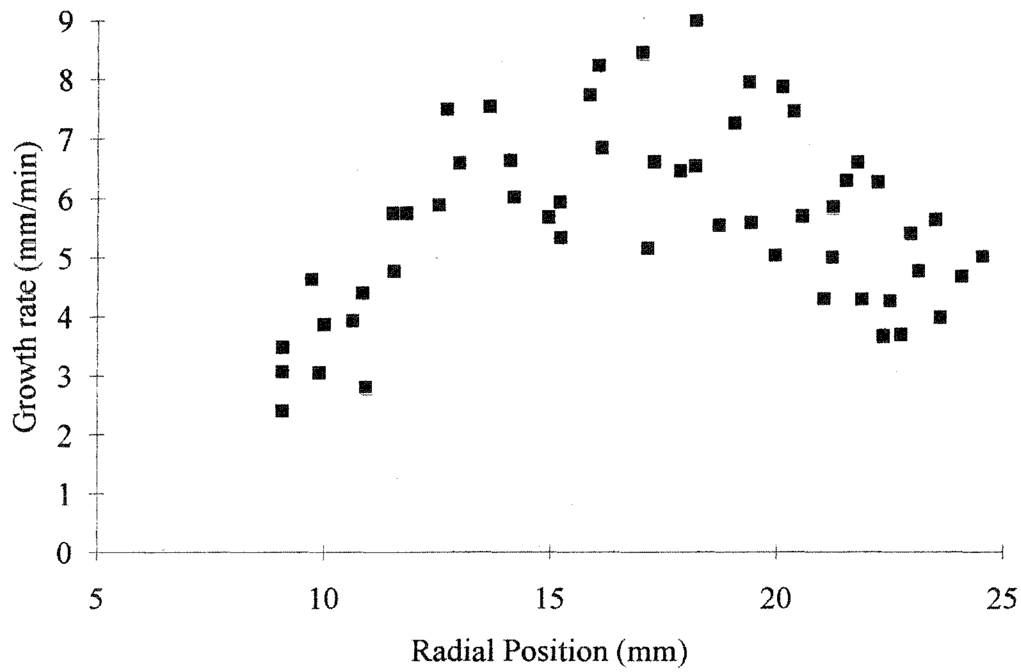

Fig. 4 Plot of growth rate vs. radial position.
(Combined data from three different radii)

Discussion

Control of deposition uniformity is the current limitation in the ability to perform Ceramic Joining by Selective Beam Deposition. Deposition rates for silicon carbide are already sufficient, and the material adheres strongly to substrates with 20% or more porosity. Uniformity suffers from an apparent starvation of reactant in the center of the deposition area at 50 Torr, and non-uniform growth at 300 Torr. The intermediate range may include conditions which avoid both problems. The disk produced by an extended deposition time investigates only certain combinations of scan rate and tetramethylsilane pressure. However, the ability to identify and examine material grown over a wide range of conditions provides a great deal of information in a single experiment. A continuously flowing gas reactor would avoid the accumulation of methane gas which occurs in the present system, but as a first order approximation we assume the effect of decreasing the activity of TMS on the formation of silicon carbide is independent of the presence of methane gas. Fig. 4 gives a summary of the phenomena observed: at the beginning of the experiment, the growth rate increases gradually even though the TMS concentration decreases and the methane concentration increases. After approximately eight hours, the growth rate peaked. The gas composition, assuming the overall reaction shown above, was 75 Torr TMS and 675 Torr methane. Layer uniformity continued to improve as the experiment progressed.

The increase in growth rate during a simultaneous decrease in TMS pressure and increase in methane pressure requires further study. One possible explanation is that the methane gas is producing pyrolytic carbon which is added to the silicon carbide growth. This possibility was not expected based on the lack of growth of pyrolitic carbon in earlier SALD experiments using methane [11]. It is possible that the presence of TMS acts to facilitate the growth of carbon. This explanation may be tested by examining the deposited material to establish the presence of a large fraction of pyrolitic carbon. If the increased growth rate is actually due to scan speed, scan speed may represent a useful parameter in decreasing the time required to join components, while at the same time increasing the surface uniformity of joint. A continuous flow reactor would help determine if this is an actual benefit by holding the gas composition constant as the radius of the disk increased, increasing the scan rate. Finally, one must consider that a phenomena not yet identified is responsible for the apparent increase in growth rate with respect to decreasing reactant concentration.

The tendency for the process to produce distinct layers indicates that interfaces between successive scans must be addressed to produces a structurally sound joints. In the past, the addition of hydrogen has improved interfaces in deposited silicon carbide [12].

Conclusion

The above suggests important implications for a gas-based joining process. In general, the process will require high growth rates and good uniformity. The physical properties of the materials joined impose additional constraints to those seen in earlier SALD processes. While investigating a wide range of deposition conditions in an extended experiment, two ceramic objects were successfully joined by the process for the first time.

Acknowledgements

This material is based in part upon work supported by the Texas Advanced Technology Program under grant No. CSME-ATPD-157 and the Office of Naval Research grant No. N00014-92-J-1514.

References

1. Figueras, A., S. Garelik, J. Santiso, R. Rodroguez-Clemente, B. Armas, C. Combescure, R. Berjoan, J.M. Saurel, R. Caplain, "Growth and Properties of CVD-SiC Layers Using Tetramethylsilane", Materials Science and Engineering, Volume B11, No. 1-4, January 15, 1992, pp 83-88.
2. Zong, G., J. V. Tompkins, W. R. Thissell, E. Sajot, and H. L. Marcus, "Processing Problems Associated With Gas Phase Solid Freeform Fabrication Using Pyrolytic Selective Area Laser Deposition", *Proceedings of the Solid Freeform Fabrication Symposium,* edited by Harris L. Marcus, Joeseph J. Beaman, David L. Bourell, and Richard H. Crawford, The University of Texas at Austin, Austin, Texas, August 12-14, 1991, p 271.
3. Pegna, Joeseph and James Maxwell, "Gas Phase Laser-Induced Pyrolysis of Tapered Microstructures", *Proceedings of the Solid Freeform Fabrication Symposium*, edited by Harris L. Marcus, J. J. Beaman, Joel W. Barlow, David L. Bourell, and Richard H. Crawford, ISSN 1053-2153, The University of Texas at Austin, Austin, Texas, August 7-9, 1995.
4. Tompkins, J. V, B. R. Birmingham, and H. L. Marcus, "Advances in Selective Area Laser Deposition of Silicon Carbide", *Proceedings of the Solid Freeform Fabrication Symposium*, edited by Harris L. Marcus, J. J. Beaman, Joel W. Barlow, David L. Bourell, and Richard H. Crawford, ISSN 1053-2153, The University of Texas at Austin, Austin, Texas, August, 1994.
5. Wallenberger, F. T., "Amorphous Silicon NitrideFibers Grown from the Vapor Phase", Journal of Materials Research, v 9, n 3, March, 1994, pp 527-530.

6. Jakubenas, K. and H. L. Marcus, "Selective Area Laser Deposition of Titanium", *Proceedings of the Solid Freeform Fabrication Symposium*, edited by Harris L. Marcus, J. J. Beaman, Joel W. Barlow, David L. Bourell, and Richard H. Crawford, ISSN 1053-2153, The University of Texas at Austin, Austin, Texas, August 7-9, 1995.

7. Pegna, Joeseph and James Maxwell, "Gas Phase Laser-Induced Pyrolysis of Tapered Microstructures", *Proceedings of the Solid Freeform Fabrication Symposium*, edited by Harris L. Marcus, J. J. Beaman, Joel W. Barlow, David L. Bourell, and Richard H. Crawford, ISSN 1053-2153, The University of Texas at Austin, Austin, Texas, August 7-9, 1995.

8. Tompkins, J. V, B. R. Birmingham, and H. L. Marcus, "Advances in Selective Area Laser Deposition of Silicon Carbide", *Proceedings of the Solid Freeform Fabrication Symposium*, edited by Harris L. Marcus, J. J. Beaman, Joel W. Barlow, David L. Bourell, and Richard H. Crawford, ISSN 1053-2153, The University of Texas at Austin, Austin, Texas, August, 1994.

9. *ibid*

10. *ibid*

11. Zong, G., J. V. Tompkins, W. R. Thissell, E. Sajot, and H. L. Marcus, "Processing Problems Associated With Gas Phase Solid Freeform Fabrication Using Pyrolytic Selective Area Laser Deposition", *Proceedings of the Solid Freeform Fabrication Symposium,* edited by Harris L. Marcus, Joeseph J. Beaman, David L. Bourell, and Richard H. Crawford, The University of Texas at Austin, Austin, Texas, August 12-14, 1991, p 271.

12. Tompkins, J. V, B. R. Birmingham, and H. L. Marcus, "Advances in Selective Area Laser Deposition of Silicon Carbide", *Proceedings of the Solid Freeform Fabrication Symposium*, edited by Harris L. Marcus, J. J. Beaman, Joel W. Barlow, David L. Bourell, and Richard H. Crawford, ISSN 1053-2153, The University of Texas at Austin, Austin, Texas, August, 1994.