**Characterization of Diamond Film Growth in a Combustion Flame**

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**ABSTRACT**

Significant progress has been made in producing large area, heteroepitaxial diamond films. Work has been directed at heteroepitaxial nucleation of diamond on nickel in a hot-filament chemical vapor deposition (CVD) reactor; and high rate, textured growth in a flat flame acetylene combustion reactor. In addition, a detailed kinetic model of the combustion reactor has been developed and validated by gas microprobe sampling. Comparisons of model predictions and experimental observations of growth rate and morphology have been used to develop a full understanding of the deposition process. Future studies will begin examining dopant effects on the diamond nucleation and growth process.

**SUBJECT TERMS**

Highly Oriented Diamond Film Growth, Combustion Flame
CHARACTERIZATION OF DIAMOND FILM GROWTH IN A COMBUSTION FLAME

FINAL REPORT

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Summary: This research has investigated the growth of diamond films in the combustion flame of an oxyacetylene torch. As a result of this research we now have a good understanding of the parameters that affect the growth of high quality diamond films and a better understanding of the nucleation process. Research resulting in the growth of highly textured, high quality diamond films on Si (100) substrates is reported.
This final report presents progress made during ARO contract #31720-MS, covering the period from 1 April 1993 - 31 March 1998.

(1) LIST OF MANUSCRIPTS:

Book Chapters:


Journal and Proceedings Publications:


9. "Control of Diamond Heteroepitaxy on Nickel by Optical Reflectance", P. Yang, R.


(2) SCIENTIFIC PERSONNEL:

Robert F. Davis - co-P.I. (1996-98)
Zlatko Sitar - co-P.I. (1996-98)
Jeffrey Glass - co-P.I. (1992-95)
Shane Trent - Masters (1993-95)
Peichun Yang - Ph.D. (1995-97)
Wei Liu - (1997-present)
Colin Wolden - Postdoctoral Student

(3) INVENTIONS: None

(4) SCIENTIFIC PROGRESS AND ACCOMPLISHMENTS

Program Objective: The purpose of this research is to provide an improved understanding of the early nucleation and growth of high quality diamond films deposited by combustion flame synthesis. The immediate program objectives have been twofold: 1) to identify surface features and chemical effects that control the formation of epitaxially oriented diamond nuclei on Ni substrates, and 2) to determine processing methods that promote the continued growth of diamond films into uniform single-crystal films with a minimum of defects and entrapped impurities, and improved physical (optical, thermal and electrical) properties.

Progress: Significant progress was achieved under this 5 year program toward the goal of producing large area, hetero-epitaxial diamond films. Work has been directed in two areas: heteroepitaxial nucleation of diamond on nickel in a hot-filament chemical vapor deposition (CVD) reactor; and high rate, textured growth in a combustion CVD reactor. The process we have developed to produce oriented diamond nuclei on Ni is very sensitive to the time-temperature history. To achieve better control, in-situ laser reflectometry was developed as a real time process monitor. It was found that the scattered light signal was sensitive to surface structures at the submicron level, and could be used as a process control parameter. A reproducible process for oriented diamond on nickel was developed in which experimental parameters were adjusted as a function of the scattered light signal. Subsequent optimization with this set up yielded an increase in nucleation density as shown in Figure 1, in which 90% of the nuclei demonstrate direct registry with the {100} Ni substrate. In addition, the mechanism underlying the process was extensively investigated using transmission electron microscopy (TEM) and X-ray diffraction analysis (XRD). Cross section TEM and selected area XRD studies identified the presence of a Ni4C interfacial layer that is believed to play an important role during nucleation.
A limitation of the hot-filament CVD system is that growth rates are relatively slow (< 1 m/hr). One would like to rapidly grow the oriented nuclei shown in Figure 1 into a coalesced film with properties that approach that of single crystal diamond. For this purpose a flat flame acetylene burner was constructed that produces uniform deposition at rates > 5 m/hr. In addition, the apparatus was equipped with the capability to monitor the gas phase composition at the growth surface using microprobe sampling and detection by quadrupole mass spectroscopy. Quantitative mole fractions were measured as a function of reactor conditions (reactant composition, pressure, flow rate, burner-substrate distance). A detailed kinetic model of the reactor was developed and validated by the experimental measurements. Comparisons of model predictions and experimental observations of growth rate and morphology were used to develop an understanding of the deposition process. Based on these insights, experimental conditions were determined that yield {100} textured growth. Using substrates that were pretreated by bias-enhanced nucleation, highly oriented {100} films were deposited on silicon as shown in Figure 2. The high growth rate of the low-pressure combustion reactor produced continuous films with low angle grain boundaries after only 3 hours of growth.

(5) TECHNOLOGY TRANSFER:

A poster presentation was made at the 20TH ARMY SCIENCE CONFERENCE (25-27 June 1996) entitled "Growth of Highly Oriented Polycrystalline Diamond Films in an Enclosed Combustion Flame System". This proved to be an excellent forum for bringing the research to the attention of the entire Army research community.

AWARDS:

Best Student Poster Award, November 1996. Materials Research Society, North Carolina Section Fall Meeting.

1997 Issai Lefkowitz Award (Army Research Office) to Dr. Prater
Figure 1: SEM micrograph of oriented diamond on (100) nickel produced after 6 hours of growth with the use of real time optical monitoring in a hot-filament CVD system.

Figure 2: SEM micrograph of continuous, highly-oriented diamond on (100) silicon produced in a low pressure combustion system after 3 hours of growth.