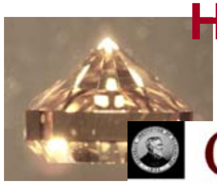

High-temperature/low-pressure (LPHT) annealing of SC-CVD diamond

Yu-fei Meng, Chih-shiue Yan, Joseph Lai, Szczesny Krasnicki, Haiyun Shu, Thomas Yu, Qi Liang,
Ho-kwang Mao, and Russell J. Hemley
Geophysical Laboratory, Carnegie Institution of Washington, Washington DC

Introduction

Diamonds produced by single crystal chemical vapor deposition (SC-CVD) at very high growth rate can be annealed at high temperature and low pressure conditions. High pressure has been considered necessary to prevent graphitization for high temperature annealing diamonds above 1600℃. However at below atmospheric pressure we successfully annealed single crystal diamonds up to 2200℃ by microwave plasma without graphitization. This LPHT annealing effectively improves the optical properties of nitrogen-doped brown CVD diamond.

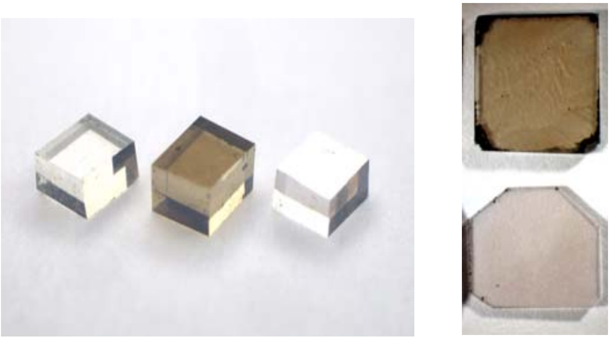

Figure 1. Left images: A brown CVD diamond cut into 3 segments. The middle piece is as-grown; the left is annealed to colorless; the right is annealed to pink. Right images: Pair of as-grown (brown, top) and annealed (pink, bottom) SC-CVD diamond crystals

Method

Our method: LPHT annealing
Technique: microwave plasma
Conditions: 1600~2200 ℃, <300 torr (outside diamond stability field), hydrogen environment
Sample: nitrogen-doped brown CVD diamond

Conventional method: HPHT annealing (Debeers & GE, 1999)
Conditions: 1800~2500℃, 5 GPA (to prevent graphitization)
Sample: brown diamond

Figure 2. MP-CVD instruments

This work was supported by NSF-EAR and DOE/NNSA (CDAC)

Optical enhancement

I. absorption spectroscopy

We observed a dramatic decrease in UV-visible absorption (Fig. 3) and a decreased absorption in near infrared region (Fig. 4) after the HTLP annealing .

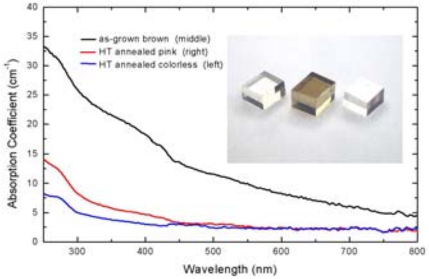

Fig. 3 UV-VIS absorption spectra of brown CVD diamond before & after LPHT annealing.

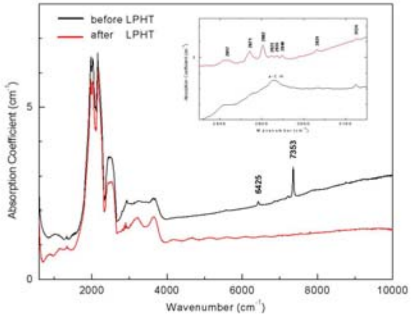

Fig. 4 UV-VIS absorption spectra of brown CVD diamond before & after LPHT annealing.

II. Photoluminescence

At annealing temperatures below 1700 °C or short annealing times, the PL of NV centers increases by factor up to 5 (Fig. 5, left). At temperatures over 1700 °C and longer annealing times, the PL of NV centers decreases (Fig. 5, right).

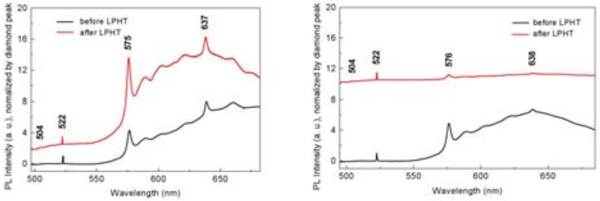

Fig. 5 PL spectra of brown CVD diamonds measured at 300K and excited by 488nm laser line.

Discussion

Characterization of CVD diamond by UV-VIS, PL and FTIR spectroscopy before and after LPHT annealing has advanced the understanding of the origins of brown and pink colors of our SC-CVD crystals and of the possible mechanism of annealing.

Fig. 6 Pink CVD diamond after LPHTannealing.

Conclusion

LPHT annealing of nitrogen doped brown SC-CVD diamond has been shown to be an effective optical properties enhancement process in such crystals. In contrast to the HPHT annealing technique, the LPHT method is not constrained by the size of crystals. The brown color of SC-CVD diamond may originate from the presence of NVH^- centers. The optical enhancement may be attributed to the disappearance of such NVH^- centers during the annealing. We suggest that the pink color of the annealed CVD diamond can be associated with the increased NV centers. As the spin associated with the NV^- complex may be a practical qubit and the number of NV^- complexes could be controlled by the LPHT annealing process, the SC-CVD diamond could be a promising host material for applications in quantum computing (Fig. 6).

Acknowledgments

We are grateful to Drs. Z. Liu, C. S Zha , and J. Wang for the IR, micro-absorption, & SIMS measurements, respectively. We thank Mr. J. Fox and Mr. T. Hall for help with the HPHT annealing.

References

[1] Yan C. S., Vohra Y. K., Mao H. K, and Hemley R. J., 2002 Proc. of. Nat. Acad. Sci. 99, R25-27.
[2] Peng M. S., Li X. Q., and Fu X. M., 1992 Human Geology, Supp., 17-21.
[3] Ho S.S., Yan C.S. Liu Z., Mao H.K., and Hemley R.J., 2006 Industrial Diamond Review, 1, 28-32.
[4] Yan C. S., Mao H. K., Li W., Qian J., Zhao Y., and Hemley R. J., 2004 Phys. Stat. Sol. (a), 201, R24-R27.
[5] Charles S. J., Butler J. E., Feygelson B. N., Newton M., Carroll D. L., Steeds J. W., Darwsh H., Yan C. S., Mao H. K., and Hemley R. J., 2004 Phys. Stat. Sol., 242, 2473-2485.