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Supporting information

Mass Spectrometric Investigation on the Roles of Several Chemical Intermediates in Diamond Synthesis

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Experimental details of diamond film growth

The experimental setup for diamond deposition using the CO_2 laser-assisted combustion flames is similar to the previous report.[S1] Combustion flames were produced by a gas mixture of C_2H_4, C_2H_2, and O_2 with a gas ratio of 1:1:2. A wavelength–tunable CO_2 laser (PRC Inc, 9.2 ~ 10.9 μm) was used to irradiate the $C_2H_4/C_2H_2/O_2$ combustion flames. The laser beam was normally projected through the flame and parallel to a tungsten carbide (WC) substrate, with a focused diameter of ~ 2 mm. The laser incident power was tuned to keep the absorbed power to be 20 W. A WC substrate (BS-6S, Basic Carbide Corp.) was placed on a water-cooled brass plate. The temperature of the substrate during the diamond deposition was maintained at 770 ~ 780 °C and monitored by a noncontact pyrometer (OS3752, Omega Engineering, Inc.). The deposition time was fixed at 1 hour.

Raman spectra of diamond films deposited as a function of the distance *h*

The bonding structures in the diamond films deposited at different *h* values were characterized using Raman spectroscopy as shown in Fig. S1. The Raman peak centred at 1337 cm^{-1} is a typical diamond peak. The band located at 1370 cm^{-1} (D-band) is attributed to the breathing modes of sp^2 atoms in rings, reflecting disordered carbon in the films. The broadband centred at 1550 cm^{-1} (G-band) is attributed to the bond stretching of all pairs of sp^2 atoms in both rings and chains and indicates graphite-like carbon contents in diamond matrix.[S2] In the sample deposited at an *h* value of 3.0 mm, a typical nano-diamond Raman feature is observed. As increasing *h* from 3.1 to 3.5, the diamond peak intensity increases and the G-band is suppressed, which indicates an improved diamond quality.

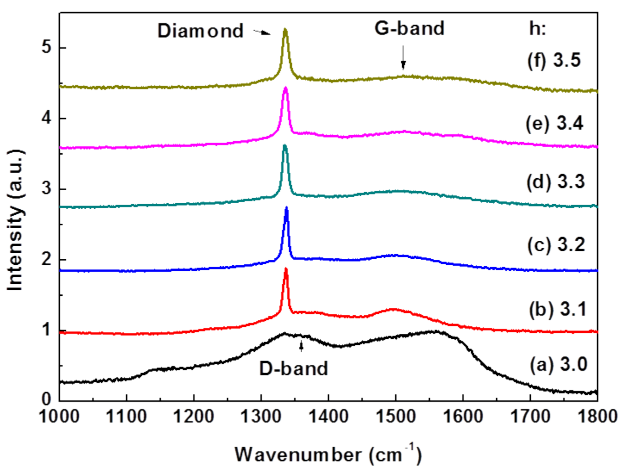

Figure S1. Raman spectra of diamond films deposited as a function of the distance *h* from substrates to the torch nozzle.

Raman spectra of diamond films deposited as a function of the gas composition *R*

The bonding structures in the diamond films deposited as a function of the gas composition *R* were characterized using Raman spectroscopy as shown in Fig. S2. The diamond obtained at an *R* value of 0.922 shows a typical nano-diamond feature. The diamond peak intensity increases with an increasing *R* value, suggesting an improved diamond quality.

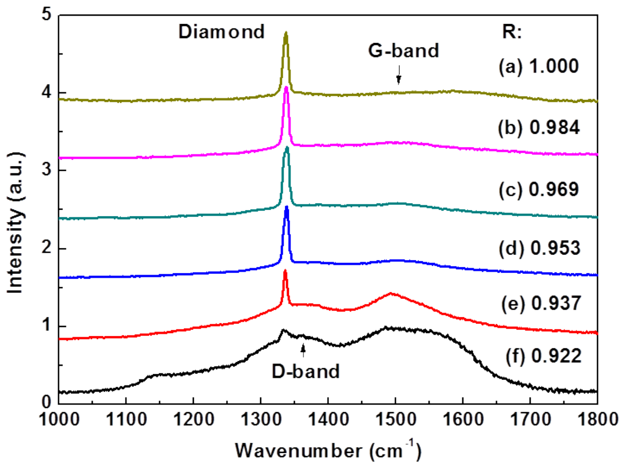

Figure S2. Raman spectra of diamond films deposited as a function of the gas composition *R*.

Notes and references

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2 A. C. Ferrari and J. Robertson, *Phil. Trans. R. Soc. Lond. A*, 2004, **362**, 2477.