**Supporting information**

**Laser-induced structure transition of diamond-like carbon coated on cemented carbide and formation of reduced graphene oxide**

Abdelrahman Zkria1,2,3,\*, Ariful Haque2, Mohamed Egiza1, Eslam Abubakr1, Koki Murasawa1,4, Tsuyoshi Yoshitake1, Jagdish Narayan2

1Department of Applied Science for Electronics and Materials, Kyushu University, Kasuga, Fukuoka 816-8580, Japan.

2Department of Materials Science and Engineering, North Carolina State University, Raleigh, NC 27695-7907, USA.

3Department of Physics, Faculty of Science, Aswan University, Aswan 81528, Egypt.

4OSG Corporation, 2-17 Shirakumo-cho, Toyokawa-shi, Aichi 442-0018, Japan.

\*Correspondence to: **abdelrahman\_zkria@kyudai.jp**

**ADDITIONAL INFORMATION**

**DLC films deposition and annealing experiments:** Cathodic arc ion plating method is employed for coating hydrogen-free DLC films onto cemented carbide (WC-Co) substrates. The films with thickness of 300nm are deposited at a deposition rate of 0.3 μm/hr, at substrate temperature of about 200 ºC. Hydrogen free DLC films prepared by cathodic arc ion plating method has the advantages of high hardness ~50 GPa and good adhesion on WC-Co substrates. Moreover, the hydrogen inflow found to increase the film’s internal stress that causes spontaneously films peeling off. In the cathodic arc ion plating technique, the distance from the target to the substrate is about 300 mm, and the plasma plume is large enough to entirely coat set of WC-Co substrates. In this work, DLC film entirely coated on the surface of WC-Co substrates. Practically, to enhance adhesion of DLC on the WC-Co substrates, higher substrate-temperature is applied due to the polished-like mirror substrate surface. The films show very low friction coefficient of 0.08 µm comparable with 0.16 µm of microcrystalline HFCVD diamond measured from pin on disk friction test in the environment air at the same conditions. After film’s deposition, the ultrafast ns-PLA process with a single pulse of ArF excimer laser (λ = 193 nm; FWHM = 20 ns) was applied on the deposited DLC films at different energy densities of 0.9, 1.2, 1.5, 1.7 J/cm2. The focal length of the converging lens is 50 cm. The spot size was selected based on the energy of the laser. As the energy of the laser is variable, so the distance between the lens and the sample is also variable. Usually, the separation was in between 35-40 cm. In this work, PLA experiments done in ambient.Schematic diagrams of the cathodic arc ion plating deposition method and ns-PLA process are illustrated in Fig.S1a and Fig.S1b, respectively.

***Raman spectra with laser annealing*:** Micro Raman analyses have been performed on the DLC/ WC–Co films, as deposited and after annealing. Figure S2 shows the reproduced experimental Raman spectra using three Gaussian peak fitting, at different energy densities of 0.9, 1.2, 1.5, 1.7 J/cm2. The fitting is performed with ﬁxed peak positions at 1140 cm-1 (T peak), 1332 cm-1 (D peak), and 1580 cm-1 (G peak), corresponding to Raman-active vibrational modes, using latest academic version of Origin-lab software (by NC State University). Obviously, Raman spectra illustrate the dependence of laser energy density. Compare to the non-irradiated film (before PLA), the laser annealed DLC films with 0.9 and 1.2 J/cm2 reveal blue shift of *G*- and *D* peaks, higher peak intensities and higher *sp3* content. The values of all parameters extracted from fitting and reproducing the measured Raman spectra before and after laser irradiation are summarized in table S1.

**FIGURES and TABLES**



**Figure S1.** Illustration of (a) cathodic arc ion plating method used for deposition of DLC films and (b) laser annealing process of DLC/WC-Co films by ultrafast ArF excimer laser (193 nm wavelength).

|  |
| --- |
|  |
| **Figure S2.** Raman spectra of DLC/WC-Co film, as-deposited and after pulsed laser annealing (PLA) at different pulsed laser energy densities (scatter), together with the best fitting results using the three Gaussian peaks (coloured lines). |

**Table S1.** The peak positions, *FWHM*, *sp3/sp2* ratio and *ID /IG* ratio, result from deconvolution of Raman spectra for DLC/WC-Co films shown in Fig.S2

