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REGULAR PAPER

Multigraphene growth on lead-pencil drawn sliver halide print paper irradiated by scanning femtosecond laser

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Drawings were made on various types of paper using lead pencils of grades from 4H through 10B. Raman spectroscopy verified both G and D peaks on all the drawings on PC print paper, PC photopaper, kent paper, and paper for silver halide print. After irradiation with a scanning femtosecond laser, silver halide paper drawn on with a 10B lead pencil maintained its surface flatness compared with the other types of paper. Raman spectroscopy on silver print paper showed a high-intensity G peak and a low-intensity D peak. After irradiating the scanning femtosecond laser on silver halide paper drawn on with a 10B lead pencil, Raman spectroscopy showed a high-intensity G peak and less intense of D peak together with a 2D peak around 2,700 cm^{-1} corresponding to the existence of multigraphene.

1. Introduction

After the discovery of graphene[1–3] prepared by peeling Scotch tape off graphite, graphene has been prepared by other methods such as thermal decomposition of SiC,[4–6] and chemical vapor deposition with catalysts such as Ni, Cu, and Fe.[7–11] In this study, we propose another method of preparing graphitic carbon: irradiating pencil-drawn print paper using a femtosecond laser.

Pulsed laser deposition is one of most versatile methods of growing metals,[12] oxides,[13–15] and multilayers[16,17] including graphene[18,19] and diamond.[20] In this technique, an incident laser beam irradiates a target in a vacuum chamber, and the vaporized materials from the target surface reach a substrate placed against the target, resulting in the growth of film. A femtosecond laser is often used as a laser source.[21,22] We drew on a sheet of printing paper with a lead pencil, and irradiated the drawn paper with a femtosecond laser in atmosphere at room temperature. Compared with conventional methods, this simple method of using pencil lead as a source material does not require either a high-vacuum atmosphere or high temperature. A vacuum chamber is not required and the scanned area is only dependent on the movement of the *X*–*Y* stage (sample holder), resulting in the large advantages of high productivity and large sample size.

The solid core of a pencil is made of graphite mixed with a clay binder, and is "harder" with more clay in the solid core and soft with more graphite. The ratio of graphite to clay determines the grade of the pencil from "H" for hardness to "B" for blackness, and an intermediate grade "F" for fine. A set of pencils ranging from a very hard, light-marking pencil to a very soft, dark-marking pencil usually ranges from hardest to softest with different amounts of carbon, clay, and wax,[23] as shown in Fig. 1. The grade 10B was not available until Mitsubishi Corp. produced the 10B lead pencil, which is available only in some regions of Japan.

Although paper and pencil are literally primitive materials, an interesting approach has been presented.[24–27] Lin et al. used pencil traces drawn on print paper as strain gauges and as chemiresistors. Graphite particle chains in a U-shaped pencil trace have varying resistance depending on the deformation of the paper. As shown in Fig. 1, the grade 10B with a high graphite content was mainly used for drawing on a sheet of paper in this study. Various types of paper were also prepared for this study: printing paper for a PC printer, photo print paper for a PC printer, kent paper, and print paper for silver halide prints. The paper for silver halide prints showed lower background fluorescence in Raman spectroscopy compared with other types of paper; thus, the grade 10B lead pencil was used for drawing on the back of the sliver halide print paper in this study.

2. Experimental procedure

The types of paper used in this study were (1) print paper for a PC printer (PC paper), (2) photo print paper for a PC printer (PC photo paper), (3) kent paper, and (4) paper for silver

Hardness	9H	8H	7H	6H	5H	4H	3H	2H	H	F	HB	B	2B	3B	4B	5B	6B	7B	8B	9B
Carbon (%)	41	44	47	50	52	55	58	60	63	66	68	71	74	76	79	82	84	87	90	93
Clay (%)	53	50	47	45	42	39	36	34	31	28	26	23	20	18	15	12	10	7	5	2
Wax (%)	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5

Fig. 1. Grades of lead pencils. "H" (for hardness) to "B" (for blackness), as well as "F" (fine), a letter arbitrarily chosen to indicate midway between HB and H.