# Supporting information

## Shear-force-dominated Dual-Drive Planetary Ball milling for Scalable Production of Graphene and Its Electrocatalytic Application with Pd nanostructures

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## 1. Dual-Drive Planetary Ball milling System

**Figure S1.** The Real image of Dual Drive Planetary Ball milling system used in this experiment.

The dual drive ball milling system employed in this work, shown in Figure S1. The shear force dominated dual drive milling provided two driven system, one is attached to the jar and other one with the disc. Both having different control system, which allow us to attain different transmission ratios (1/4:1 to 4:1) with precise control. The shear, frictional and impact forces can result the ball-to-wall and ball-to-ball collisions that can controlled by the optimization of rotation speed of the mill, grinding time, ratio of the sample to the ball filling as well as the material of the grinding parts.

This mill has a rotating shaft that sweeps a circle of diameter 580 mm with a horizontal axis of rotation. The two steel jars of 10 cm diameter (3800 ml each) rotate about their own axes, with again horizontal axis of rotation, and around the common axis of the main shaft. The instrument image is published for earlier work associate to the dispersion of nano-oxides in ODS ferritic steels.[1]

## 2. Energy Dispersive X-Ray Spectra (EDS) of Graphene

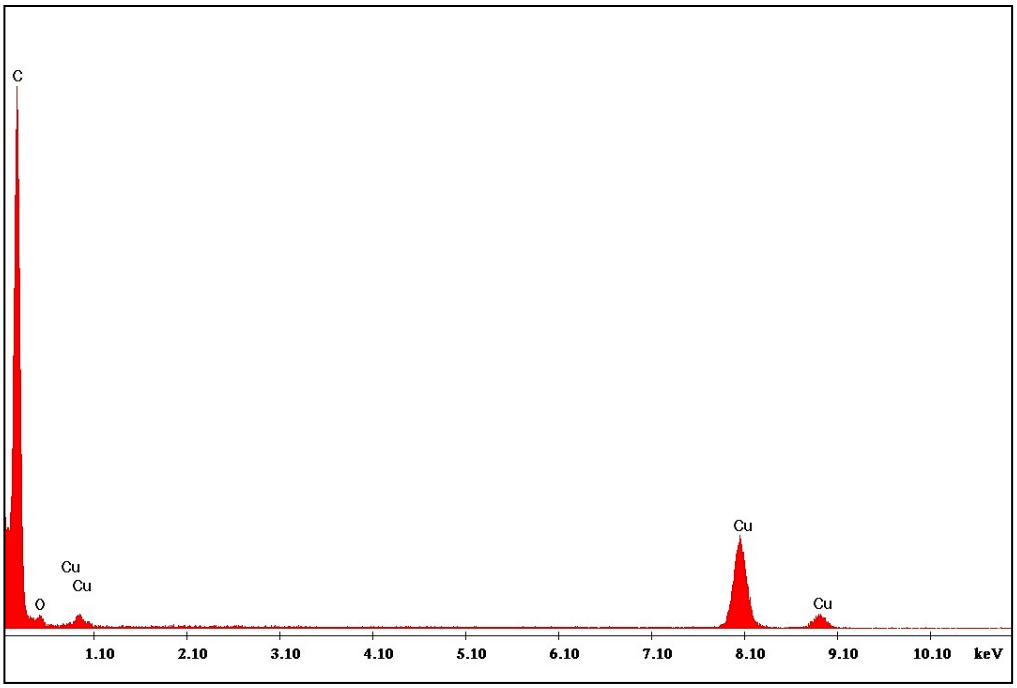


**Figure S2.** Energy Dispersive X-ray spectra of synthesized graphene sheets.

## 3. X-Ray Photoelectron Spectra of Graphite and Graphene

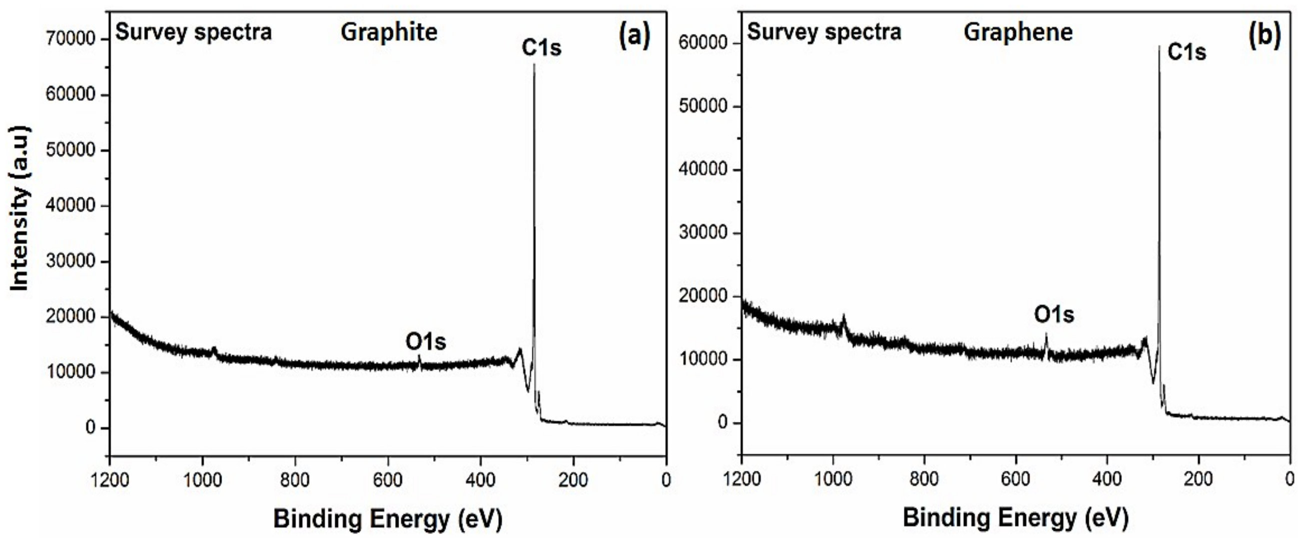


**Figure S3.** XPS Survey spectra of Graphite and Graphene

## 4. Critical Speed Calculation

**Critical speed calculation**

$$\%CS = \frac{N_2}{R \times N_1} \times 100$$

$N_1$ - Gyrating speed, $N_2$ is the Jar speed.

R –Distance between gyro and mill axis.

**Table 1 Critical Speed with respect to Gyro and Jar speed**

| Critical speed (in %) | Gyro speed (in RPM) | Jar speed (in RPM) |
|---|---|---|
| 40 | 100 | 100 |
| 70 | 150 | 300 |
| 98 | 100 | 265 |

## 5. Crystalline Size and Lattice Strain Calculation

The average crystallite size is calculated by using the Scherer's equation (1) follows:

$$D = \left( \frac{k\lambda}{\beta_{hkl} \cos\theta} \right) \qquad (1)$$

Where D is the volume weighted crystallite size, k is the shape factor (0.89), λ is the wavelength of $CuK_\alpha$ radiation, $\beta_{hkl}$ is full width at half maximum (FWHM) of the particular peak and θ is the Bragg's angle.[2]

**Table 2 Relative contribution of grain size and strain with respect to milling periods**

| Milling hours | Position | Crystalline size (nm) | Lattice Strain (%) |
|---|---|---|---|
| 0 | 26.50 | 35 | 0.45 |
| 10 | 26.45 | 33 | 0.48 |
| 20 | 26.65 | 31 | 0.50 |
| 600$^{o}$C | 26.48 | 36 | 0.53 |

## 6. Raman spectroscopic analysis

**Table 3 Purity and number of layers with respect to critical speed**

| Critical speed (in %) | Purity($I_D/I_G$) | Number of Layers ($I_{2D}/I_G$) |
|---|---|---|
| 40 | ~0.53 | ~4 |
| 70 | ~0.59 | ~6 |
| 98 | ~0.63 | ~9 |

## Reference


1. K. Jayasankar, A. Pandey, B. Mishra and S. Das, *Fusion Engineering and Design*, 2016, **102**, 14-20.
2. B. Culity and S. Stock, *Reading: Addition-Wesley*, 1978.