

## Electronic Supplementary Material

# Role of nanoparticles in achieving macroscale superlubricity of graphene/nano-SiO<sub>2</sub> particle composites

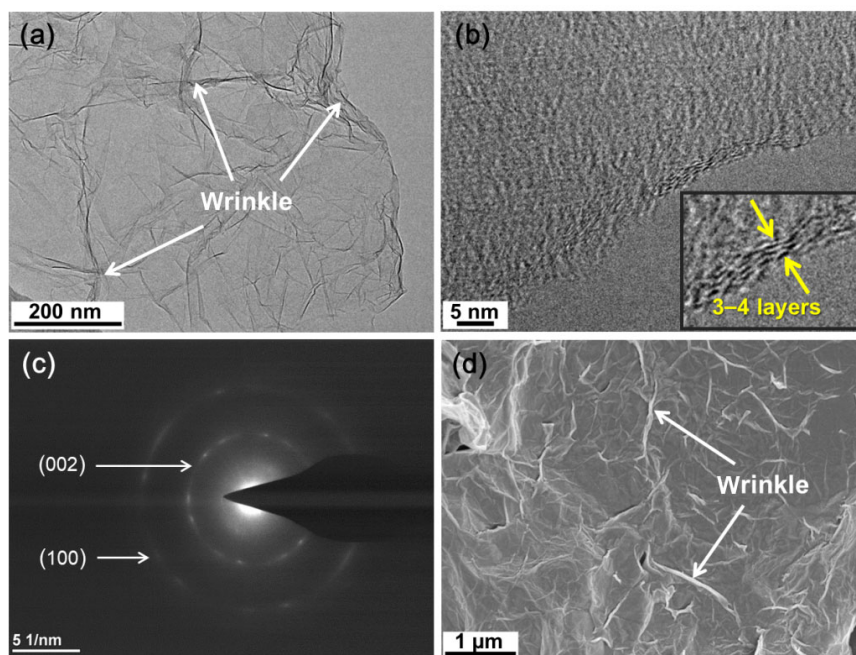
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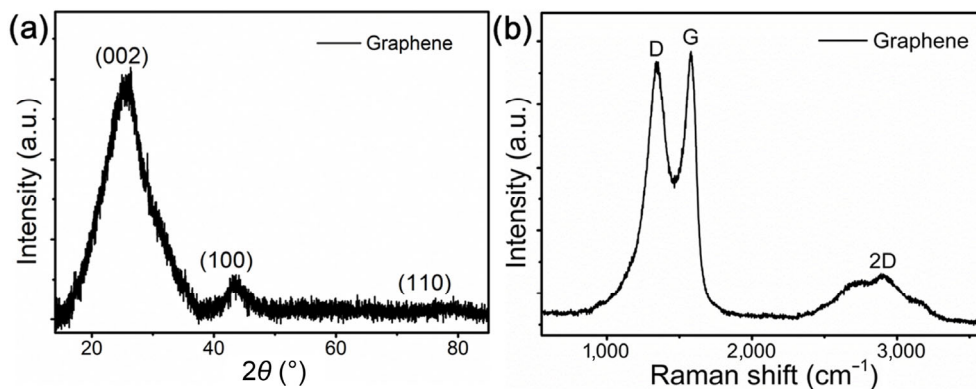
Supporting information to <https://doi.org/10.1007/s40544-021-0532-2>

### 1 Morphology and structure of original graphene nanosheets



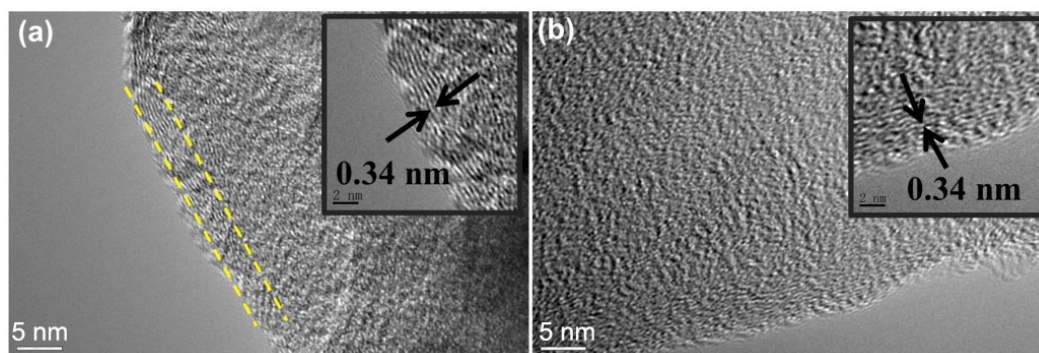
**Fig. S1** Morphologies and topographies of graphene nanosheets: (a) TEM image, (b) HRTEM image, (c) selected area electron diffraction pattern, and (d) SEM image.

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**Fig. S2** Original structures of graphene nanosheets: (a) XRD pattern and (b) Raman spectrum.

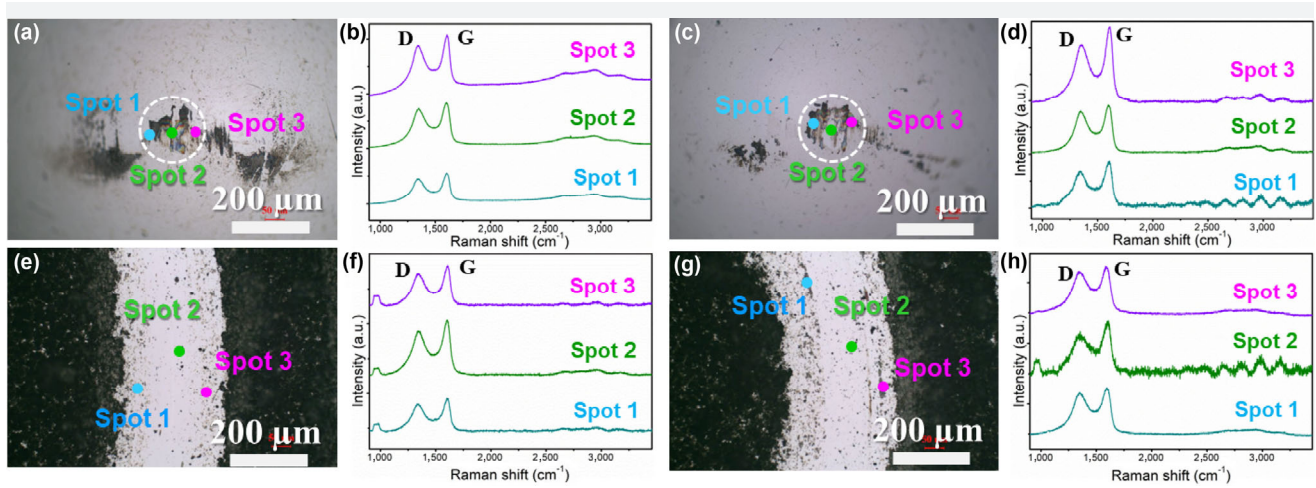
## 2 Ordered-slipping microstructure of pure graphene coating at sliding interface



**Fig. S3** HRTEM images of sliding interface of pure graphene coating after 30-min sliding under applied normal loads of (a) 0.5 N and (b) 2.0 N.

## 3 Typical morphologies and structure of wear scars and wear tracks

The morphologies of the wear scars on the counterparts and wear tracks on the substrate of the typical graphene/nano-SiO<sub>2</sub> particles composite coating in the stage that friction coefficient is stable (30 min) and the corresponding Raman spectra at labelled spots (Spot 1, Spot 2, and Spot 3) in the morphological images are shown in Fig. S4. It is indicated that the uniformly spread and compacted transferred film and lubricating film are formed on the friction pair both under low (0.5 N) or high load (2.0 N). The Raman spectra of the transferred film and the lubricating film at different positions (labelled Spot 1, Spot 2, and Spot 3 of the corresponding morphological images) show that obvious D peaks and G peaks of graphene exist at the location of the transferred film and the lubricating film, indicating that graphene nanosheets are formed on both counterpart ball and substrate at this time. There is almost no difference in the morphology of wear scars and wear tracks under low and high load. Therefore, the microstructure at the friction interface of the composite coating determines the friction coefficient of the composite coating.



**Fig. S4** Morphologies and Raman spectra of wear scars and wear tracks of the friction pair. (a) Optical images of wear scar of graphene/nano-SiO<sub>2</sub> particle composite coating after 30-min friction test under 0.5 N; (b) Raman spectra of the points in (a); (c) optical images of wear scar of graphene/nano-SiO<sub>2</sub> particles composite coating after 30-min friction test under 2.0 N; (d) Raman spectra of the points in (c); (e) optical images of wear track of graphene/nano-SiO<sub>2</sub> composite particles coating after 30-min friction test under 0.5 N; (f) Raman spectra of the points in (e); (g) optical images of wear scar of graphene/nano-SiO<sub>2</sub> composite particles coating after 30-min friction test under 2.0 N; (h) Raman spectra of the points in (g).