Supplementary

Asynchronous cracking with dissimilar paths

in multilayer graphene

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S1 Characterization of graphene specimens for fracture tests
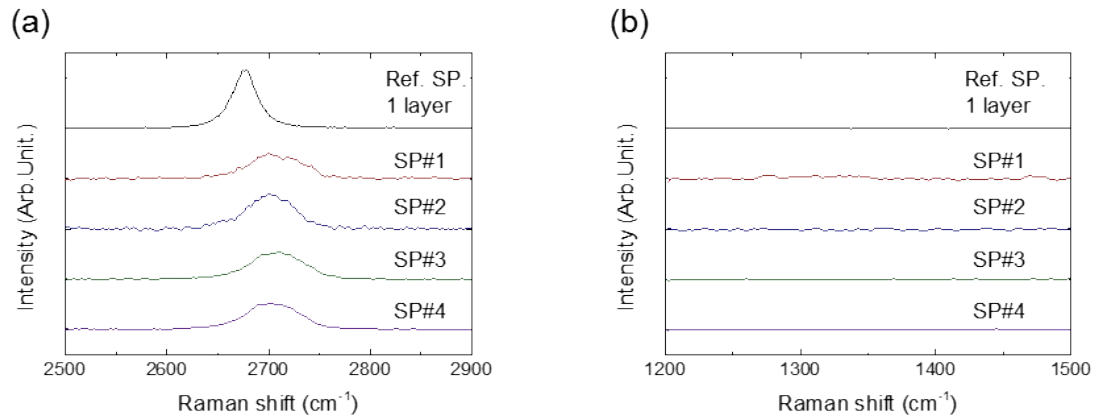
Figure S1 Raman spectrum of the specimens tested in this study. (a) 2D band and (b) D band. Shape of 2D band implies that the layers of the multilayer graphene specimens are stacked in Bernal configuration. Also, small D band explain that the specimens have few atomic disorders or defects.
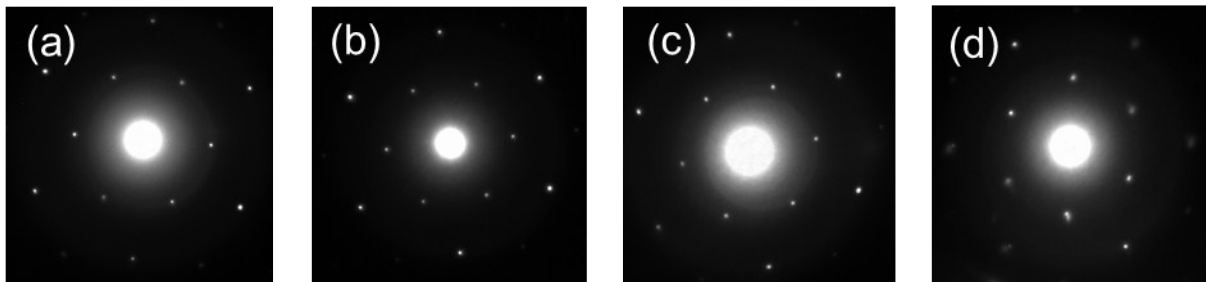

Figure S2 Diffraction patterns (a)–(d) of graphene tensile specimen (SP#1–SP#4) obtained by transmission electron microscope. This shows that the pristine graphene specimens are single crystal.
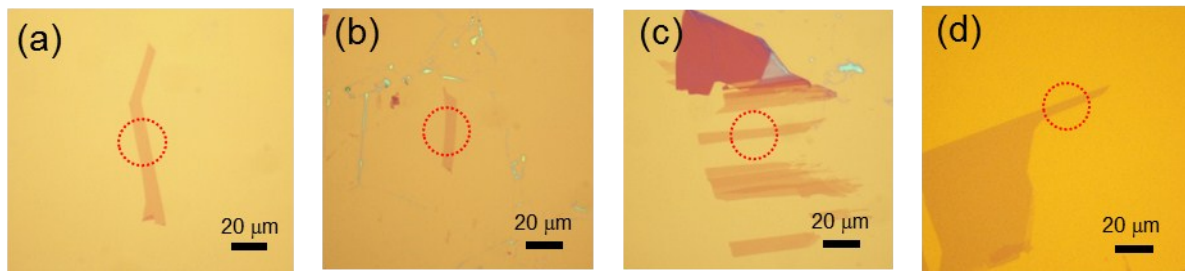
Figure S3 Optical microscope image (a)-(d) of the tested graphene specimen (SP#1-SP#4) exfoliated from highly oriented pyrolytic graphite. Red dotted circles indicate the specimen with strip shape. In the images, the number of the layers can be identified with the contrasts.[S1] However, they show uniform contrast in the entire region of the specimen. This means that the entire regions of the specimens are the same thickness.

S3 Crack propagation direction in multilayer structure
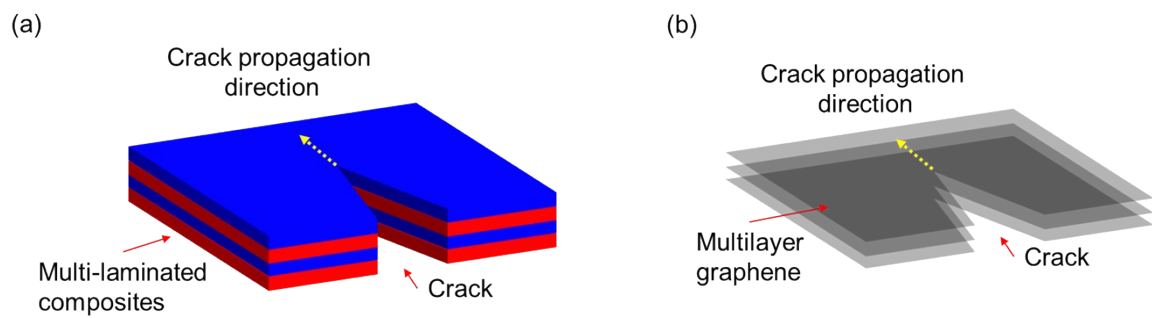
Figure S4. Crack divider orientation in multilayer structure (yellow dotted arrow). (a) Multi-laminated composites, (b) Multilayer graphene.

S3 *In-situ* movie clips of fracture testing

Suppliementary Movie 1 Movie clips taken by SEM during the *in-situ* fracture testing for SP#2.

References

S1 W. Quyang, X.-Z. Liu, Q. Li, Y. Zhang, J. Yang, Q. Zheng, *Nanotechnology*, 2013, **24**, 505701.