

Flash Graphene Morphologies—Supporting Information

Michael G. Stanford,¹ Ksenia V. Bets,² Duy X. Luong,¹ Paul A. Advincula,¹ Weiyin Chen,¹ John Tianci Li,¹ Zhe Wang,¹ Emily A. McHugh,¹ Wala A. Algozeeb,¹ Boris I. Yakobson,^{1,2, 3} and James M. Tour^{1,2,3,4*}*

¹Department of Chemistry, ²Department of Materials Science and NanoEngineering, ³Smalley-Curl Institute and the NanoCarbon Center, and ⁴Department of Computer Science, Rice University, 6100 Main Street, Houston, Texas 77005, United States

biy@rice.edu; tour@rice.edu

Schematic of flash Joule heating (FJH) system

Additional details and safety information on the FJH system can be found in a prior publication, and it is imperative that these safety recommendations be followed to mitigate accidental electrocution.ⁱ

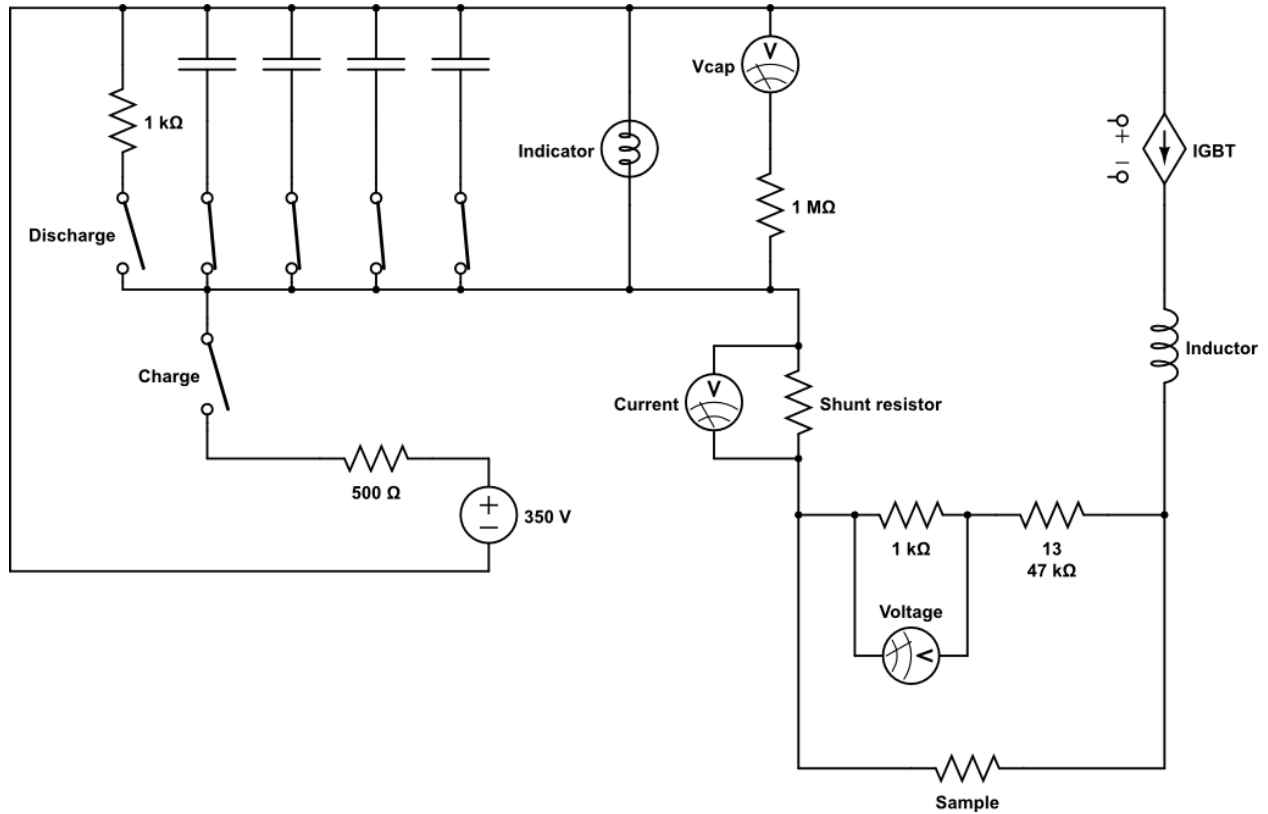


Figure S1. Schematic of the FJH system.

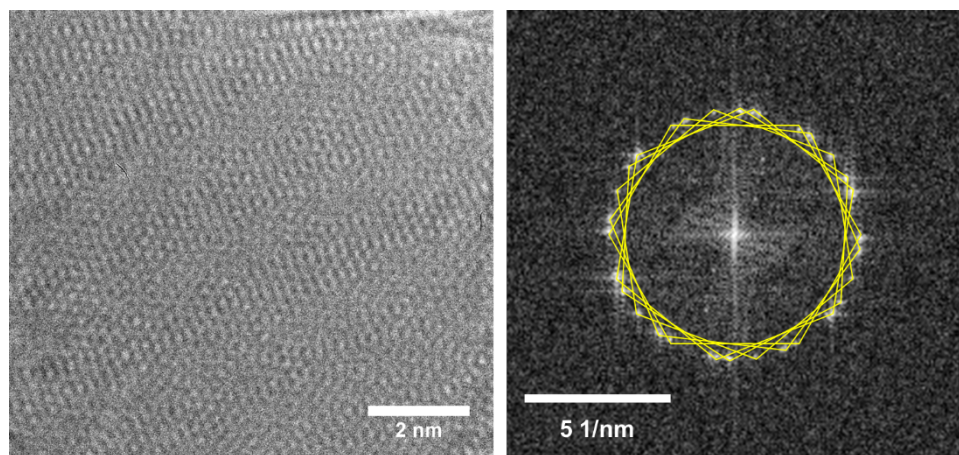


Figure S2. High-resolution TEM image of tFG sheet and FFT. FFT is composed of 5 sheets with 6-fold symmetry.

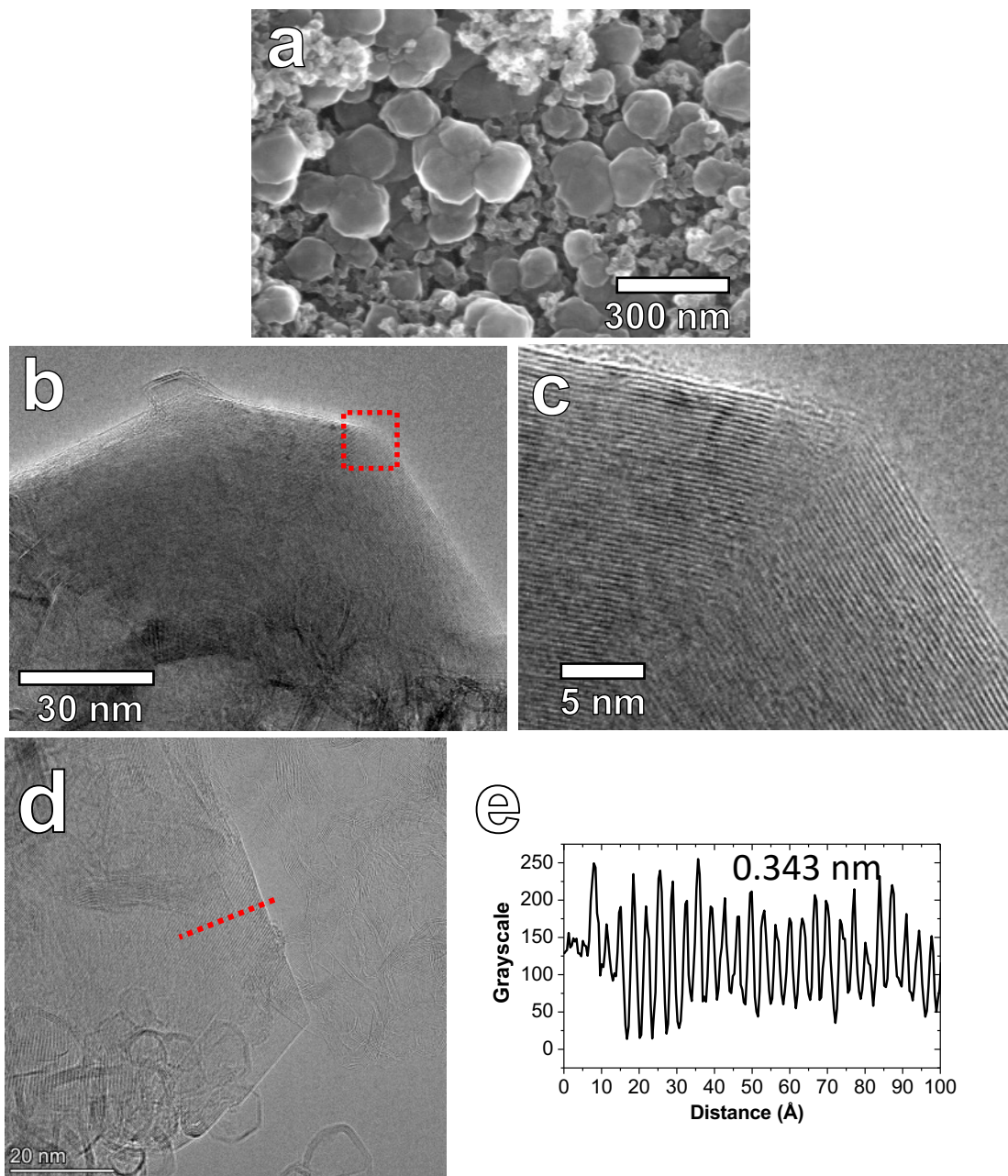


Figure S3. (a) SEM image of faceted tFG particles. (b-d) TEM images of particles. (e) Line scan showing interlayer spacing.

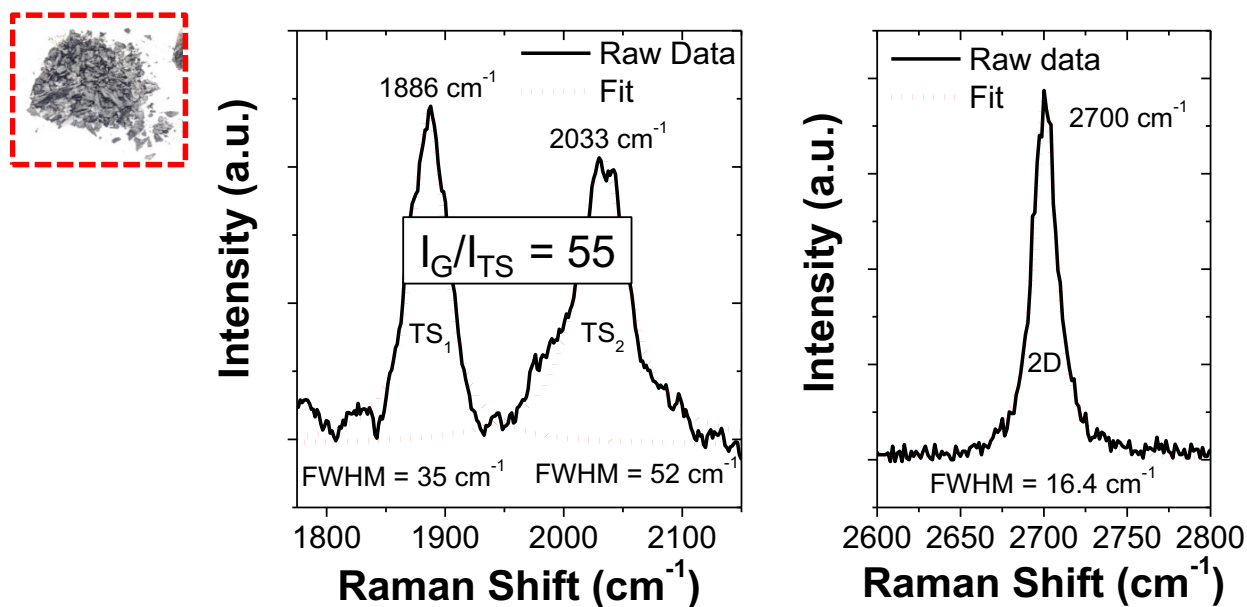


Figure S4. Raman spectra for TS1 and TS2 peaks as well as 2D peak for tFG sheets.

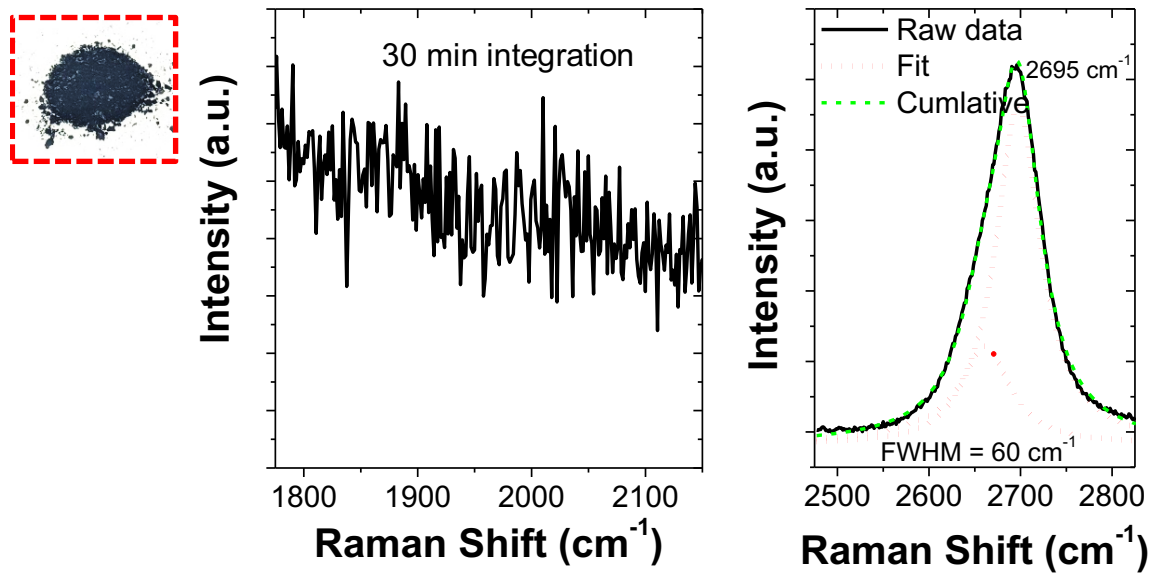


Figure S5. Raman spectra for TS1 and TS2 peaks as well as 2D peak for wrinkled graphene sheets.

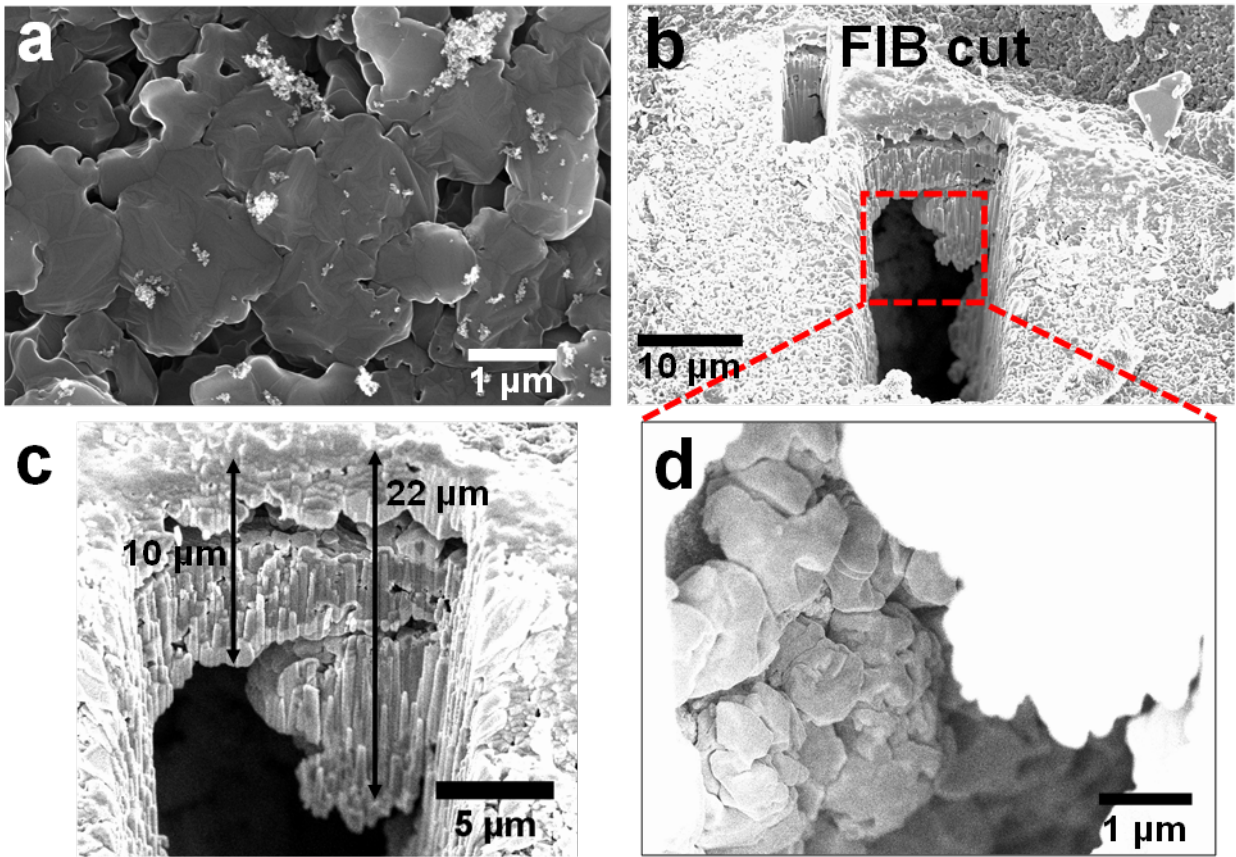


Figure S6. (a) SEM image of gray tFG particle from CB source. (b) SEM image of FIB cut into FG. (c) Measurement of tFG thickness. (d) SEM image inside of cut regions showing more turbostratic particles beneath cut.

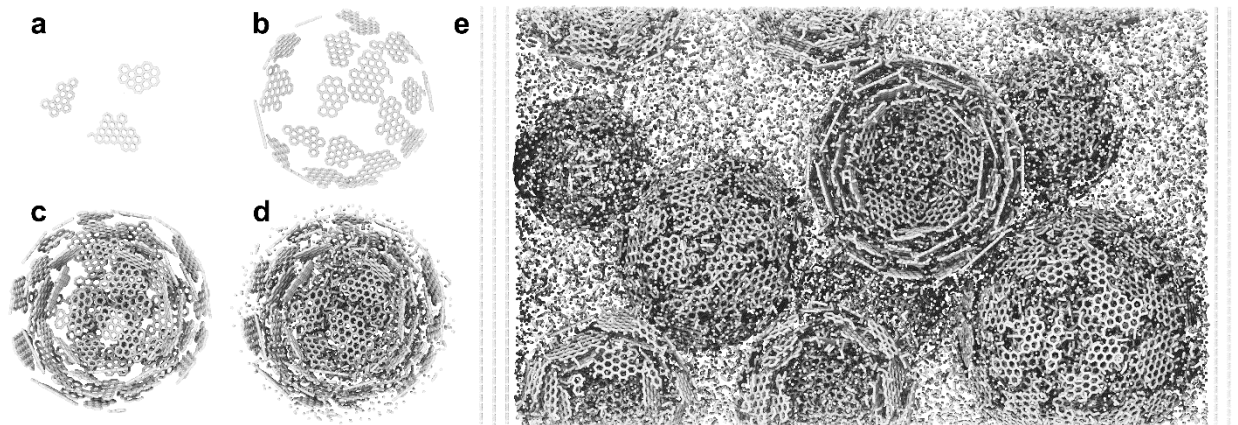


Figure S7. Formation of the initial configuration for atomistic simulations from (a) individual graphitic flakes arranged in (b) spherical shells that (c) form centroid particle with the (d) addition of individual carbon atoms. (e) The final configuration includes eight centroid particles, graphitic walls, and approximately 30% of individual randomly positioned carbon atoms.

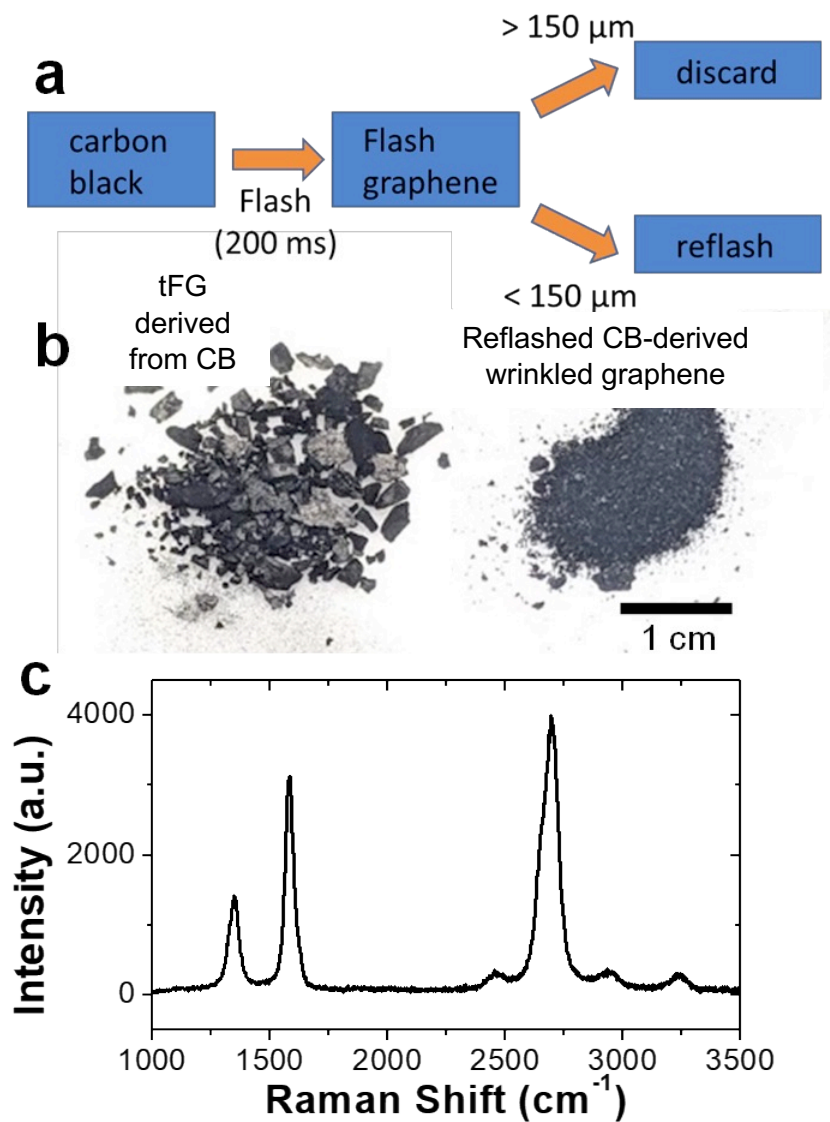


Figure S8. (a) Process flow showing the generation and reflashing of wrinkled graphene. (b) Optical image of a batch of tFG from raw CB (left) and reflashed wrinkled graphene (right) that had initially been made from CB. (c) Representative Raman spectra of reflashed CB-derived wrinkled graphene.

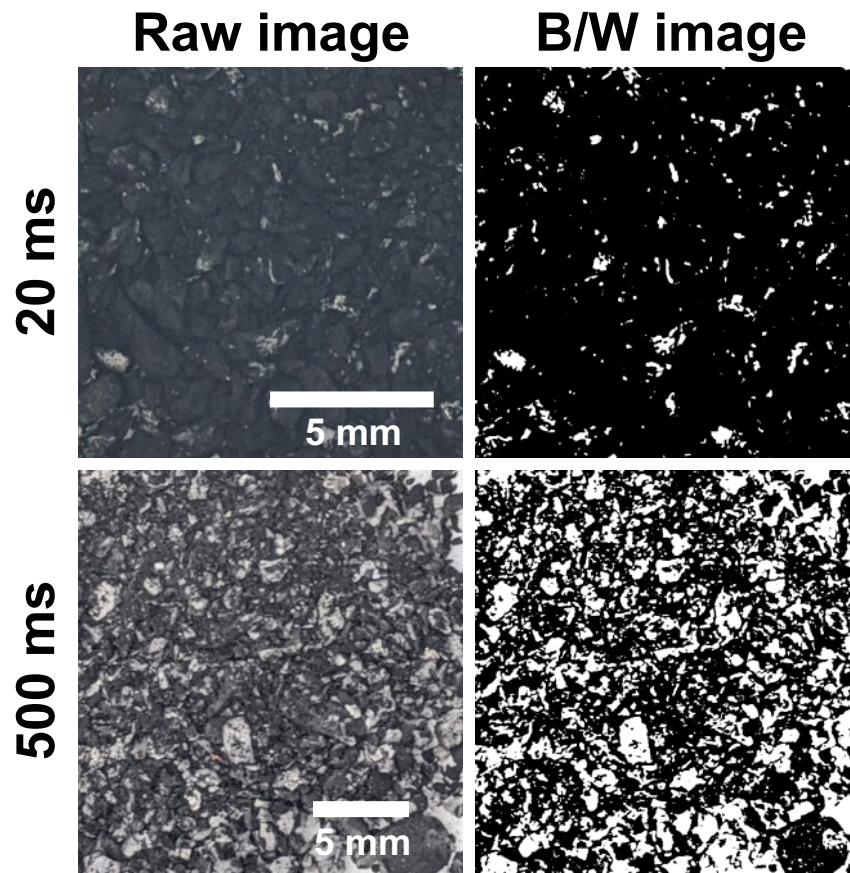


Figure S9. (left) Raw images and (right) processed images of FG at 20 and 500 ms flash duration.

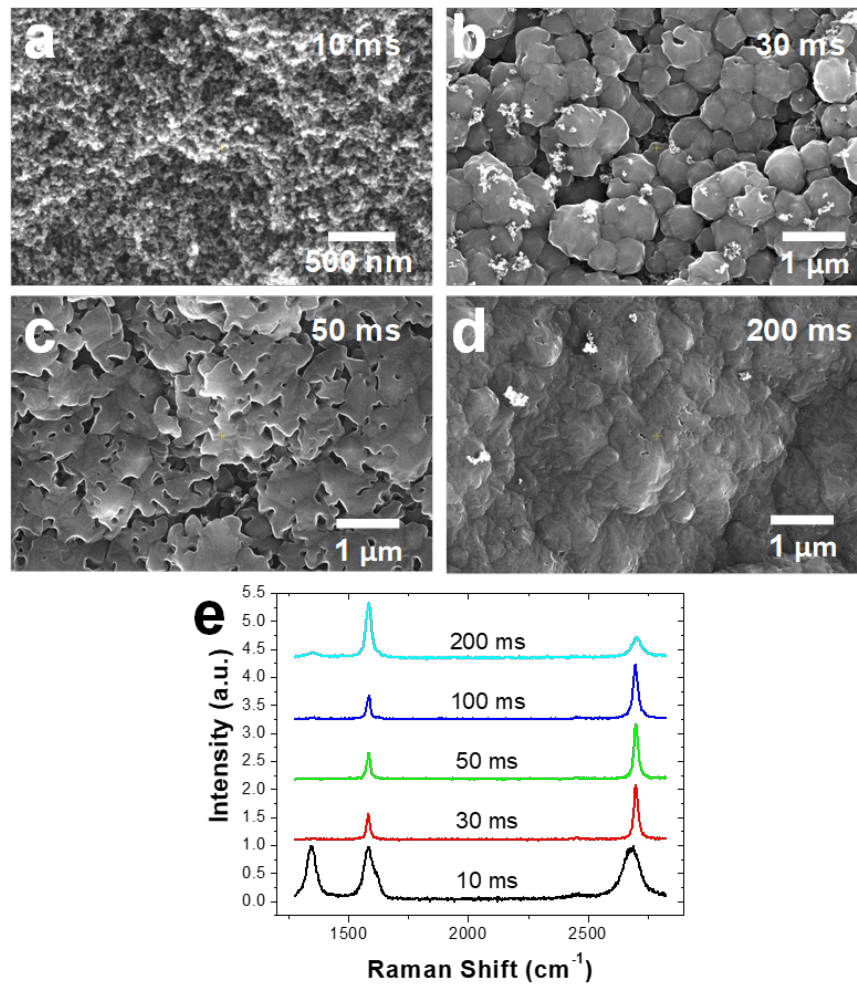


Figure S10. (a-d) Images of gray crystals, tFG sheets, as a function of flash duration. Flash voltage was 120 V. (e) Representative Raman spectra of gray particles.

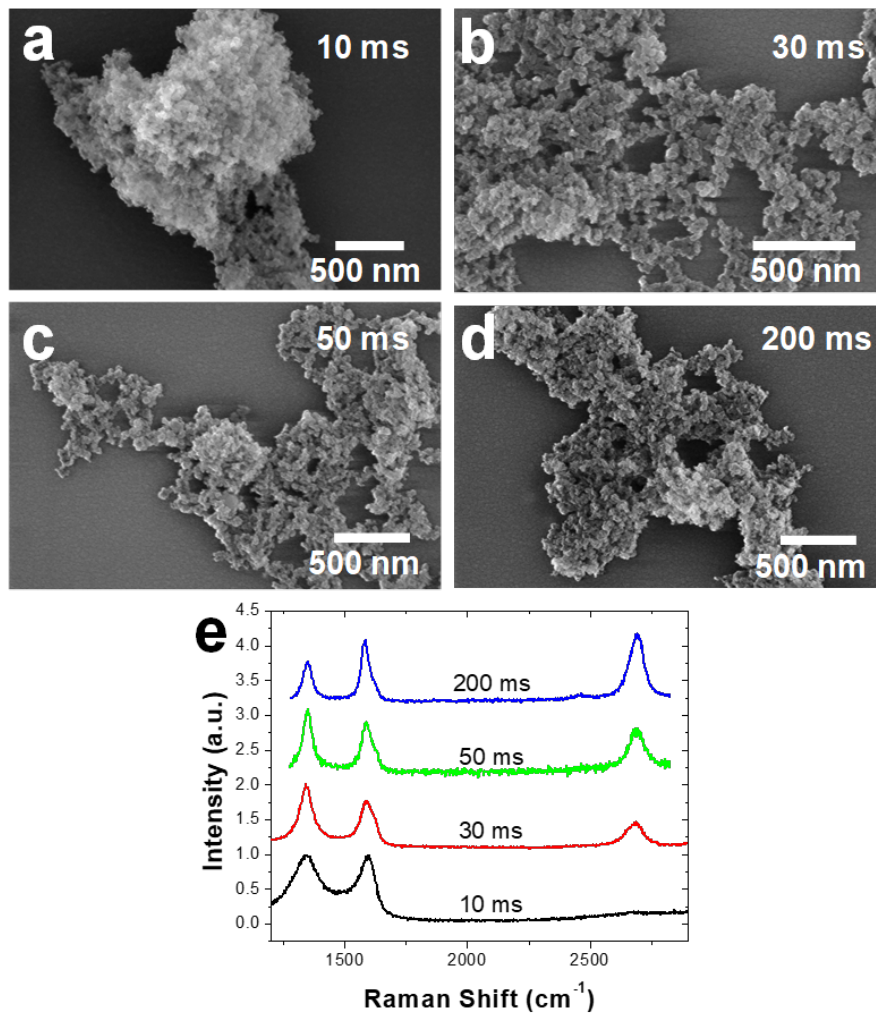


Figure S11. (a-d) Images of fine black FG powder (wrinkled) as a function of flash duration. Flash voltage was 120 V. (e) Representative Raman spectra of fine black FG powder.

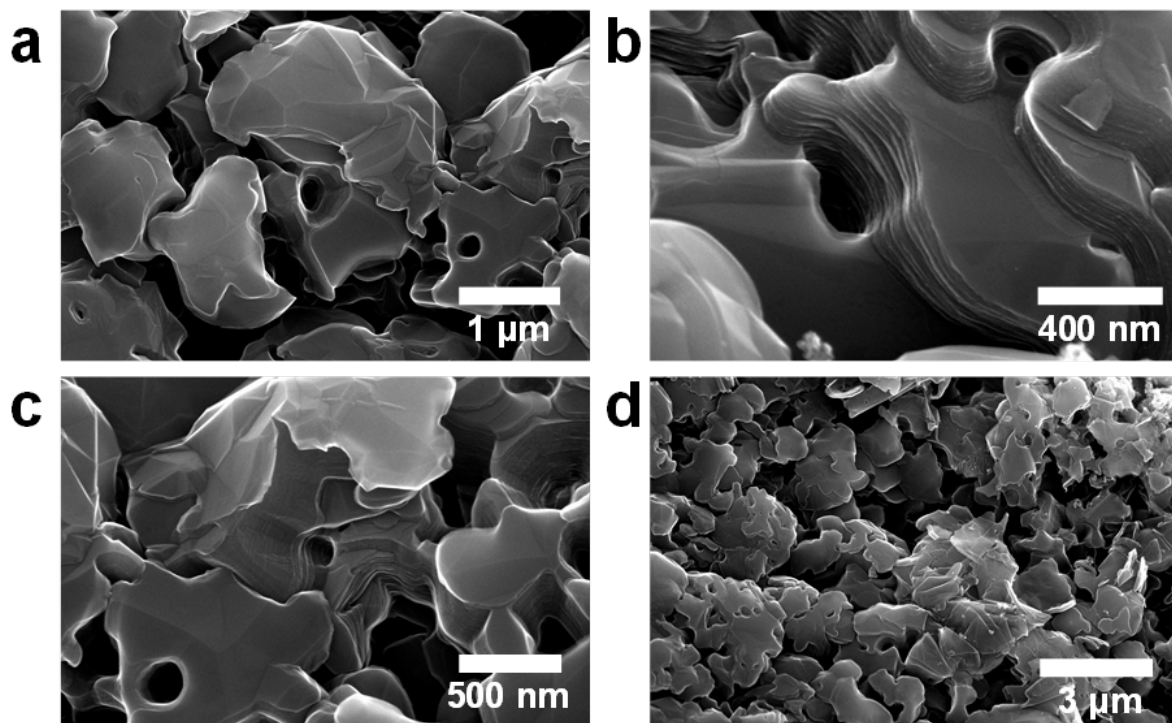


Figure S12. Various images of tFG sheets.

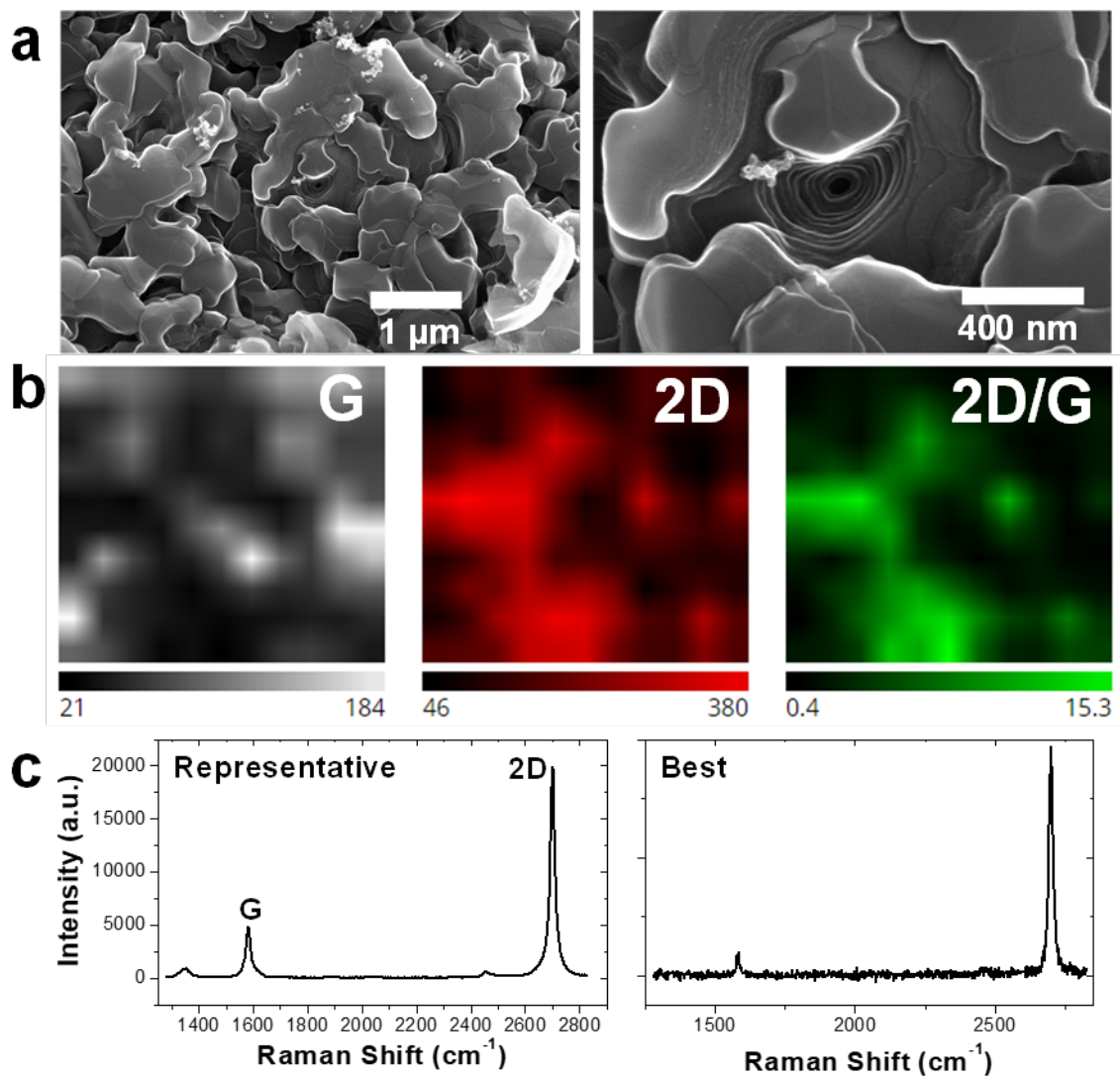


Figure S13. (a) SEM image of tFG. (b) Raman maps showing G, 2D, and 2D/G for a 20 x 20 μm area. (c) Raman spectra from representative and good regions.

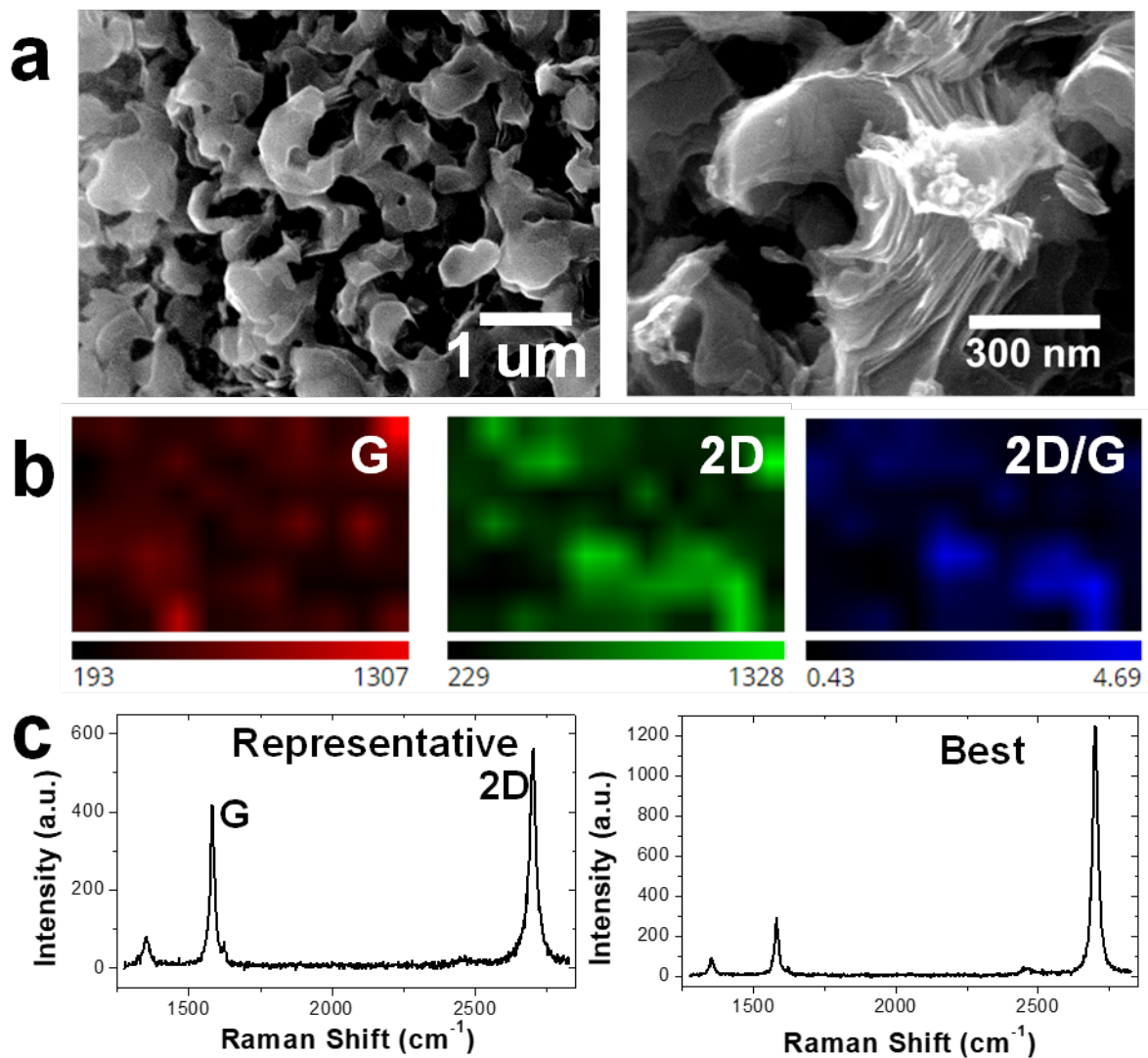


Figure S14. (a) SEM images of tFG from CB. (b) Raman mapping for a $20 \times 20 \mu\text{m}$ area. (c) Raman spectra for this material.

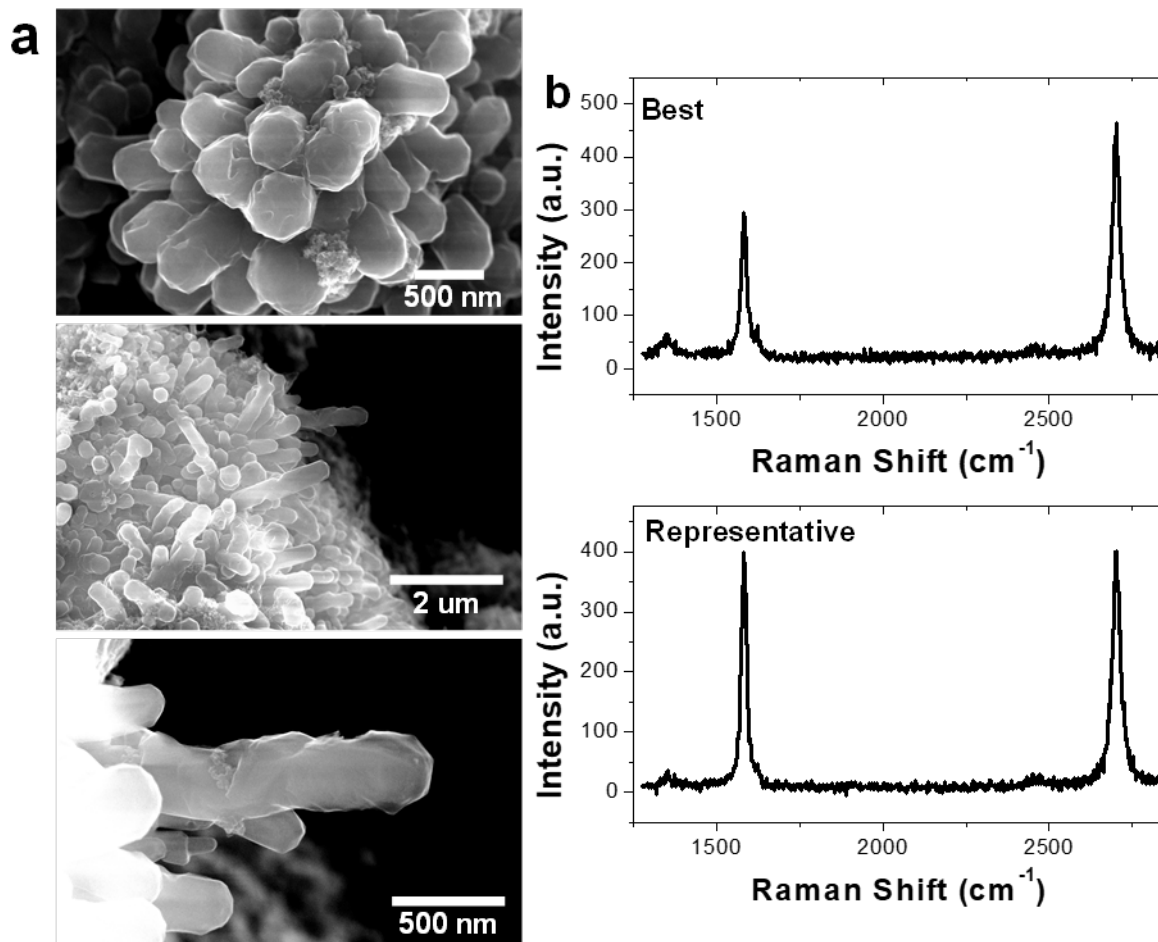


Figure S15. (a) SEM images of FG particles that exhibits rod-like structures with hexagonal faceted shape. (b) Typical Raman spectra for the FG particles.

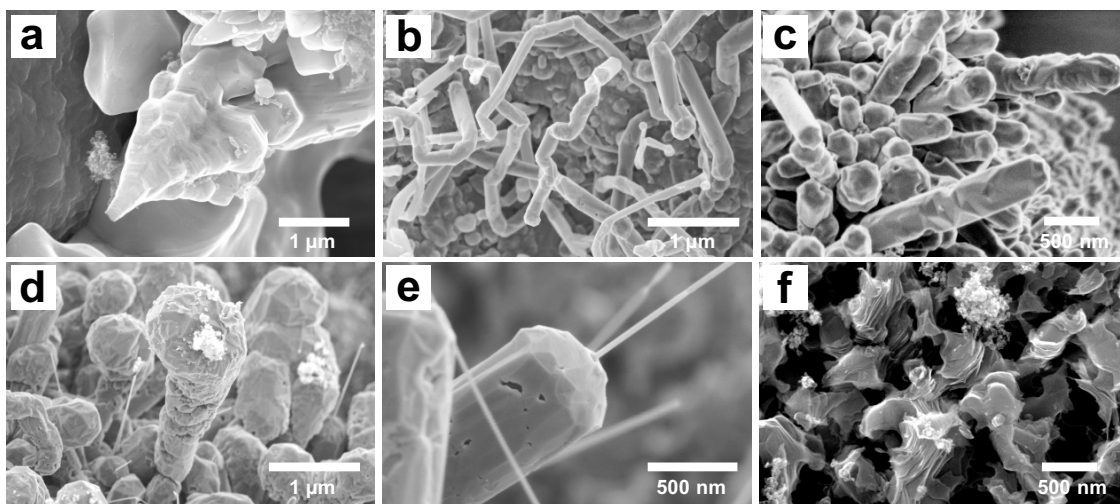


Figure S16. (a-e) SEM images of isolated graphite polyhedral crystals found on the surface of graphitized crystals. (f) SEM image of flakey, loosely bound FG.

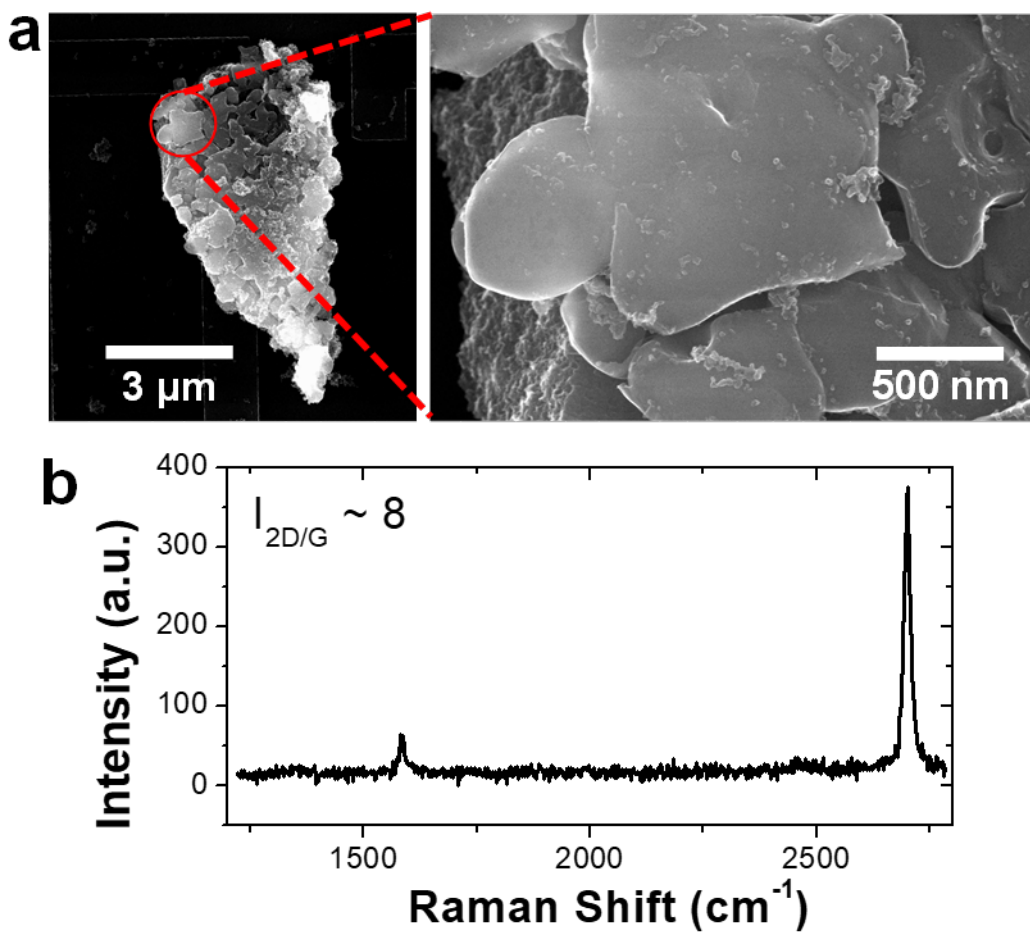


Figure S17. (a) SEM images of FG particle found with smooth morphology. Red circle indicates approximate location of 532 nm Raman laser. (b) Raman spectra for area shown in part a.

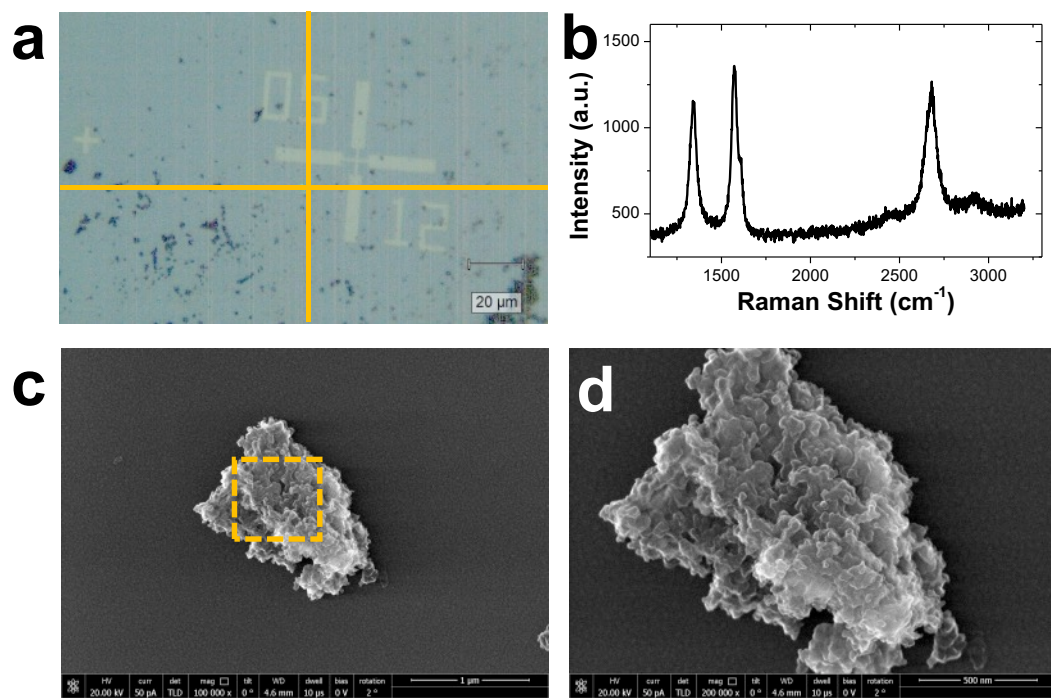


Figure S18. (a) Optical image showing FG location on alignment substrate. (b) Raman spectra of particle. (c-d) SEM images of FG particle.

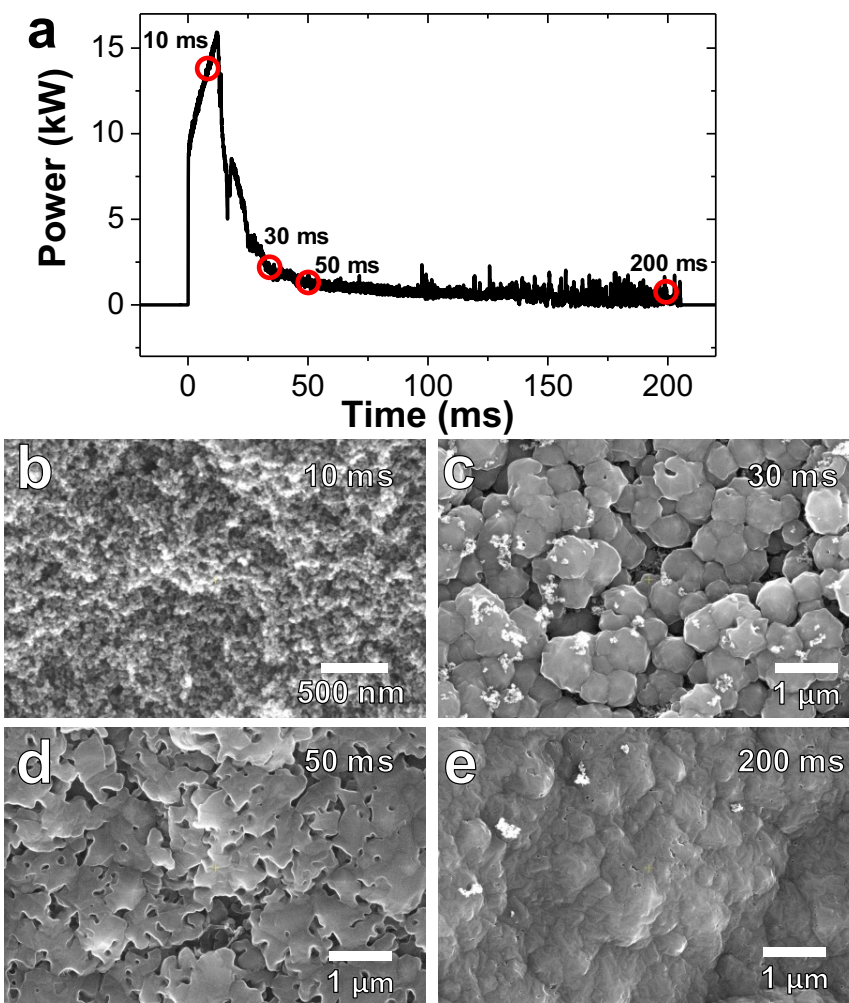


Figure S19. (a) Power dissipated in CB starting material as a function of flash duration. Representative SEM images of tFG crystals at (b) 10 ms, (c) 30 ms, (d) 50 ms, and (e) 200 ms.

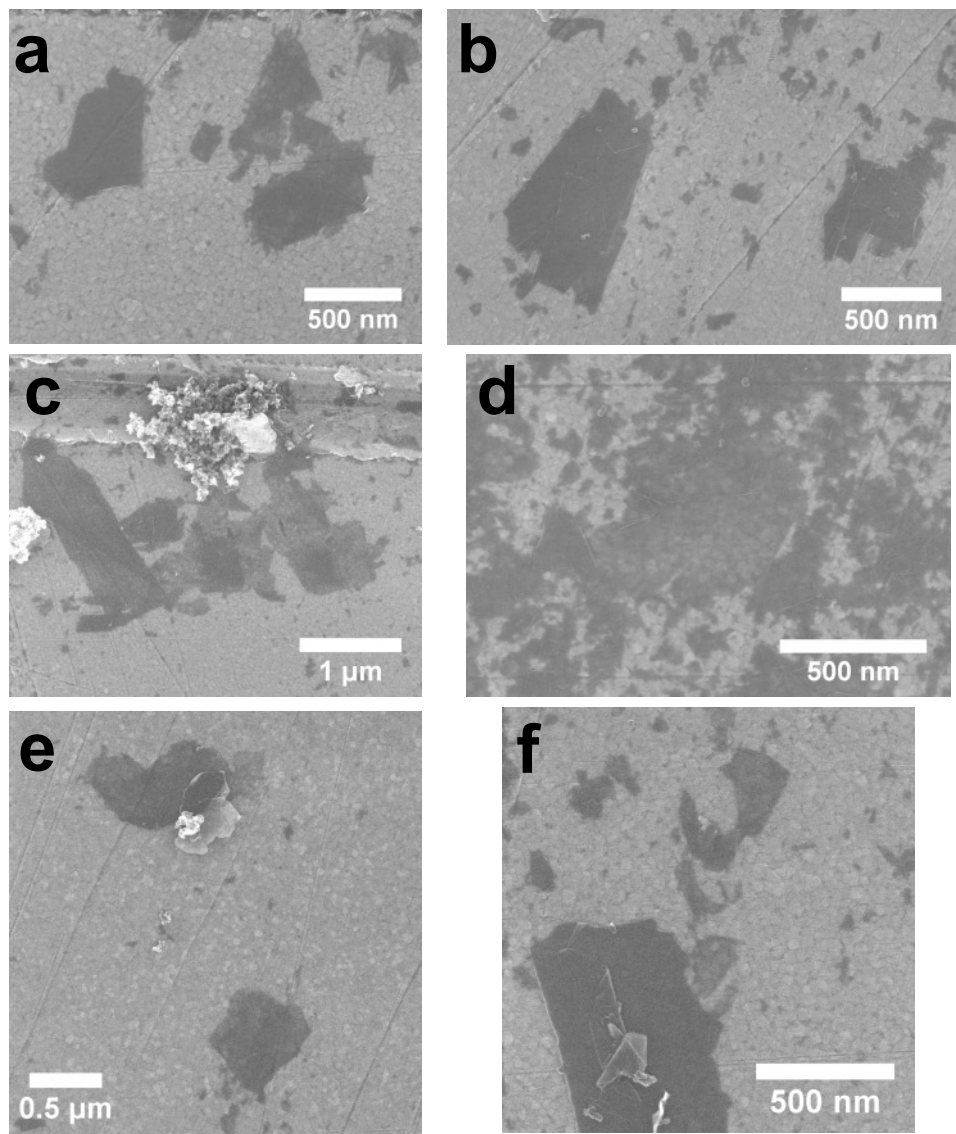


Figure S20. (a-f) Various SEM images of FG that were exfoliated onto gold.

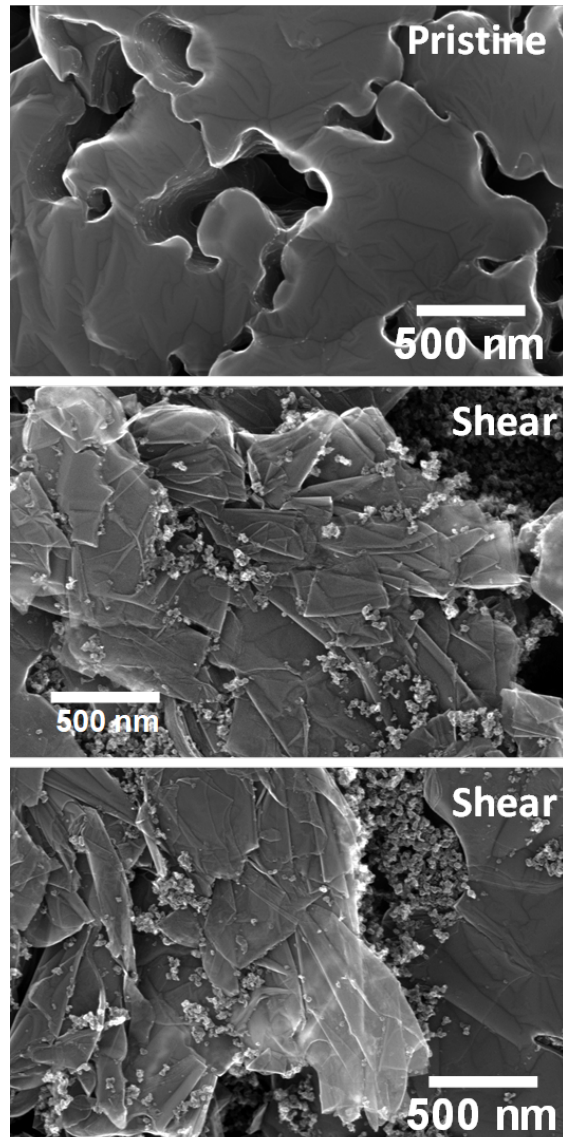


Figure S21. SEM images of FG that is (top) pristine and that has been exposed to (middle, bottom) mechanical shear.

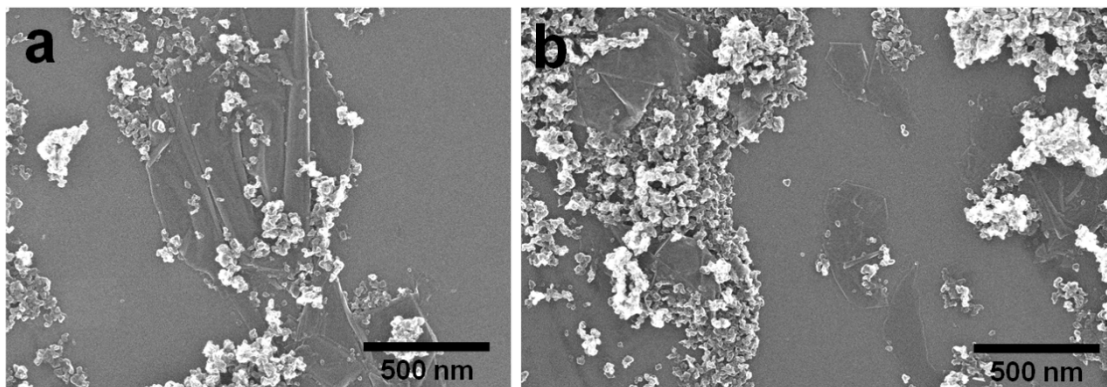


Figure S22. (a-d) SEM images of FG from CB that has been mechanically sheared between two silicon wafers.

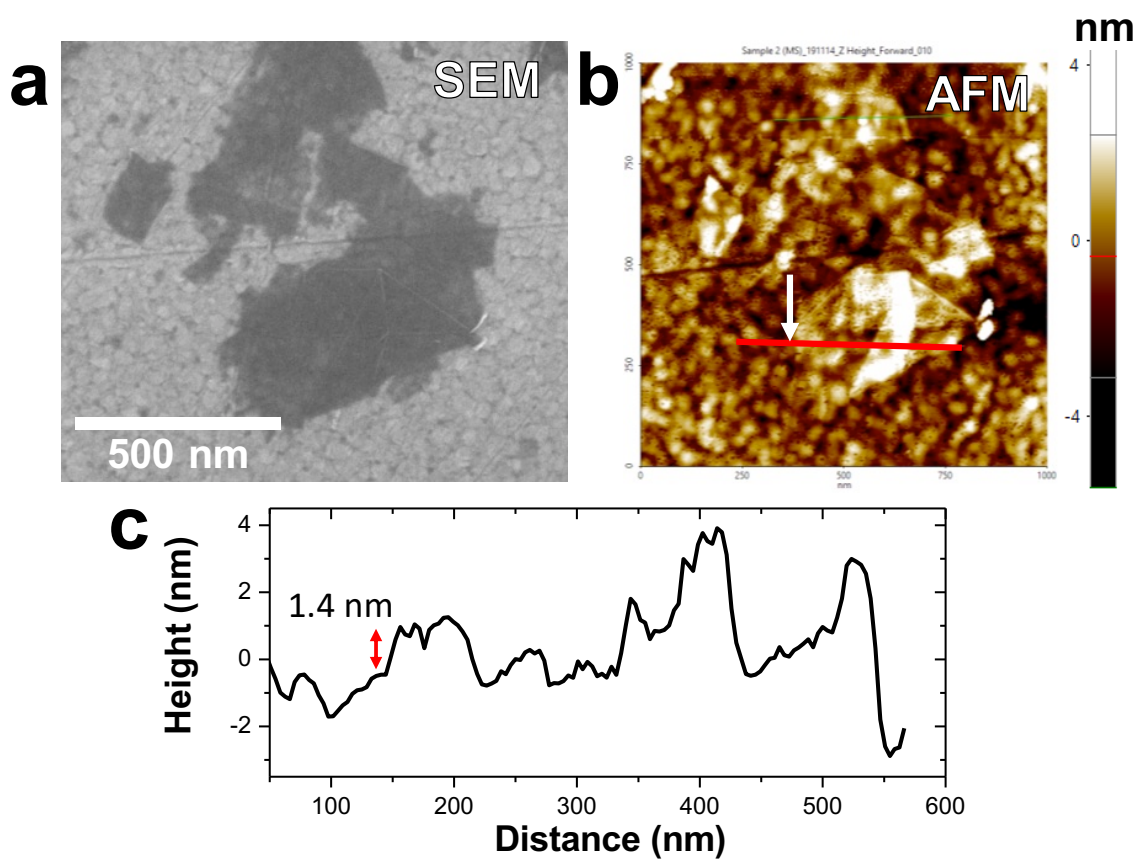


Figure S23. (a) SEM and (b) AFM image of a FG sheet. (c) Line scan from AFM showing sheet height of ~ 1.4 nm.

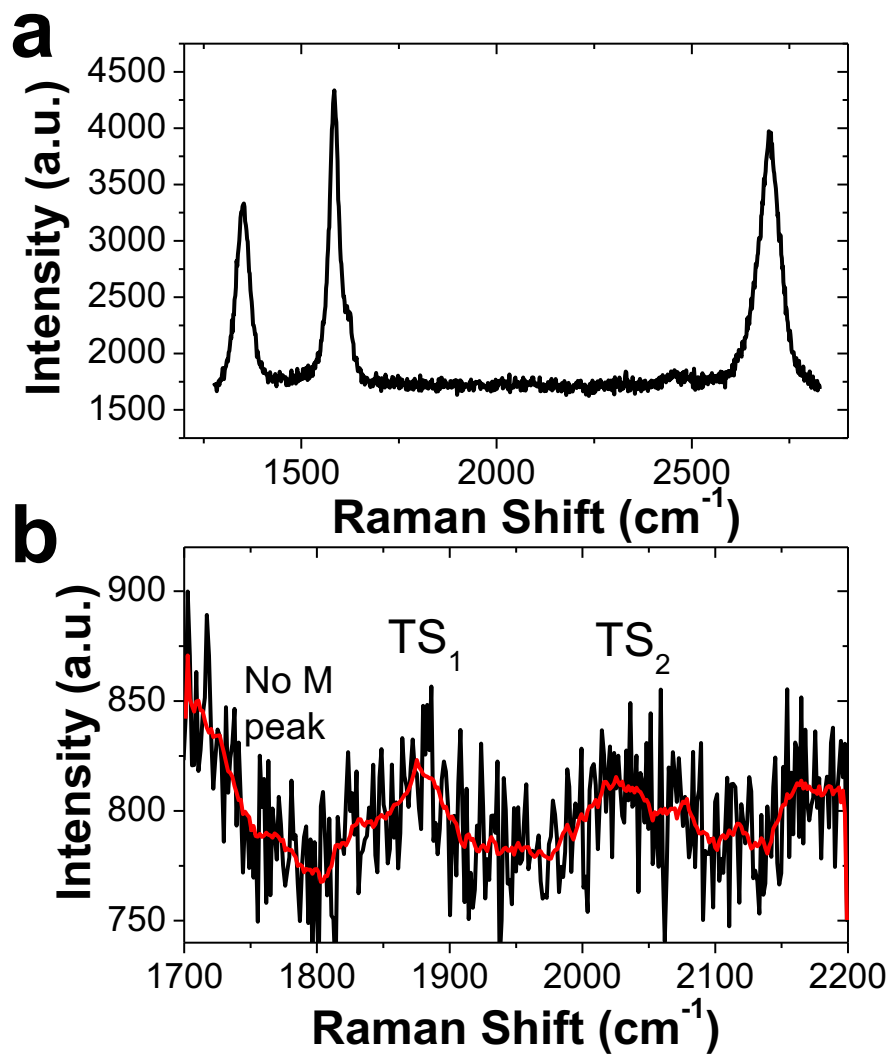


Figure S24. (a,b) Raman spectra of mechanically exfoliated FG on gold substrate.

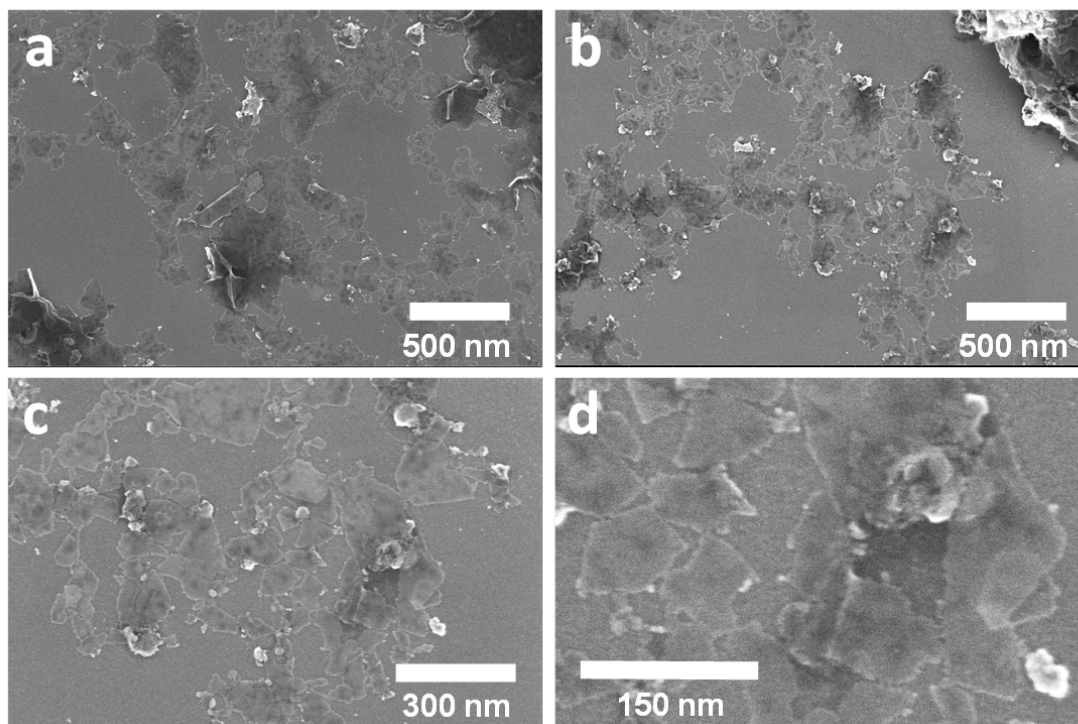


Figure S25. (a-d) SEM images of FG dispersed in oleum and dropcast onto Si substrate.

References

¹Luong, D. X.; Bets, K. V.; Algozeeb, W. A.; Stanford, M. G.; Kittrell, C.; Chen, W.; Salvatierra, R. V.; Ren, M.; McHugh, E. A.; Advincula, P. A.; Wang, Z.; Bhatt, M.; Guo, H.; Mancevski, V.; Shahsavari, R.; Yakobson, B. I.; Tour, J. M. Gram-Scale Bottom-Up Flash Graphene Synthesis. *Nature* **2020**, *577*, 647–651.