## Supplementary materials



**Fig.** A1 (color on line) Snapshots of MLGP (a-c) and SLGS (d-f). (a,d) Before equilibration, top view. Vacancies are high lighted in blue. (b,e) After equilibration, top view. (c,f) Side view of (b) and (e), respectively.



Fig. A2 (color on line) Mechanical properties of MLGP under tensile deformation, stretching along armchair direction. (a) Fracture strain, (b) ultimate strength and (c) Young's modulus with respect to defect coverage.



Fig. A3 (color on line) D moduli of MLGP and SLGS under (a) tensile and (b) shear deformations. The loads are applied along zigzag orientation.



**Fig. A4** (color on line) Snapshots of spatial atomic-stress distributions of MLGP-2.8% under in-plane shear deformations along the zigzag direction. (a,b) Per-atom stress distribution at the regime far from crack. (c,d) Stress distribution before bond breaking. Wrinkle structures can be observed, which resemble the wrinkle mode of monolayer graphene sheet under shear test. Stresses are accumulated mainly in the graphene plane, as indicated by the red sticks. (e,f) Beginning of the fracture. Plastic deformation is triggered from the weakest bond. Observable sheet tearing is initiated from one layer and the stresses are relieved within that layer, whereas the other layers remain nearly intact. This phenomenon suggests that the interlayer bonding in MLGP-2.8% is not strong. (g,h) Brittle fracture of the MLGP. The insets in (g) illustrate the tearing modes of two different layers in the MLGP-2.8%. The two sheets break from different regions. (a,c,d,g) Top views; (b,d,f,h) Side views.



**Fig. A5** (color on line) Stress contours of MLGP-11% under in-plane zigzag shear deformation. (a,b) Before deformation. Due to much stronger structural deformation and interlayer bonding, out-of-plane pleats are much more significant than that of MLGP-2.8%. (c,d) Within the elastic deformation region, before the fracture, stresses are accumulated, wrinkle structures can be observed in the graphene sheets. (e,f) Beyond the elastic region, plastic deformation is initiated from the local stress accumulated region, with significant bond breaking and sheet tearing. Note that the six layers break from the same region, resulting in an observable hole viewed from the top. (a,c,e) Top views; (b,d,f) Side views.



Fig. A6 (color on line) Mechanical properties of MLGP under in-plane armchair shear deformation. (a) Shear fracture strain, (b) shear strength and (c) shear modulus with respect to defect coverage.