Micromechanical Exfoliation of Graphene on the Atomistic Scale

— Supporting Information —

Robert C. Sinclair, James L. Suter, Peter V. Coveney

December 21, 2018

Centre for Computational Sciences - University College London, 20 Gordon Street, London, WC1H 0AJ, United Kingdom

In this supplementary information, we provide additional data from our exfoliation ensembles, and details on the videos we provide along with the manuscript.

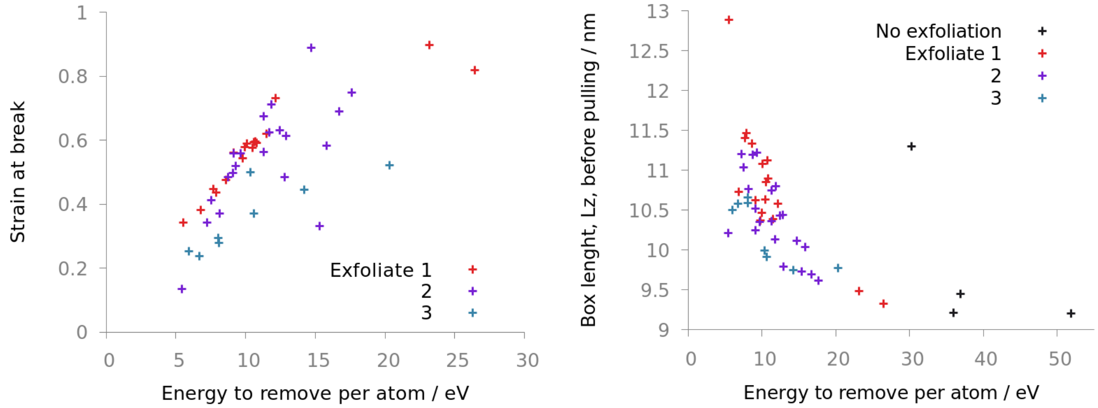
CONTENTS

I. Analysis of exfoliation simulation data 2

II. Accompanying Videos 2

I. ANALYSIS OF EXFOLIATION SIMULATION DATA

Here we display simulation data which breaks down simulations 2,4,5 by the energy required per atom to exfoliate the graphite, the strain when the stack broke and the height of the box when pulling started.



(a) (b)

FIG. 1: A break down of the exfoliation replicas taken from simulation 2 (see table 1). a) shows that exfoliation of graphene, red crosses, (apart from two exceptions) follows a smooth trend, due to the peeling mechanism discussed in the main text. Exfolation of 3 sheets follows a trend with a lower gradient, because more energy is required to bend 3 sheets than 1. b) shows the height of the box before the pulling simulation started; the distribution is quite broad and shows minimal correlation between compression and exfoliation outcome. Replicas that saw no exfoliation have a high stress because they often pulled polymers from the opposite tape with them, which requires a lot of rearrangement energy.

II. ACCOMPANYING VIDEOS

We provide two videos showing the exfoliation of 7 layer 12 nm diameter graphite by PMMA and PDMS. The videos show the same simulations from which the snapshots are taken in figure 2 in the main text. The periodic box begins at $20 \times 20 \times 10$ nm^3 and the simulation box is increased in the z direction by 10 ms^{-1}. The PMMA exfoliation lasts 1.4 ns seconds, the PDMS exfoliation lasts 1.1 ns.

The simulations differ in the mechanism of exfoliation. The PDMS is the more fluid polymer and rearranges to stay in contact with the graphite as the tapes are pulled apart, giving rise to a shearing mechanism. The PMMA does not stay in contact with the graphite

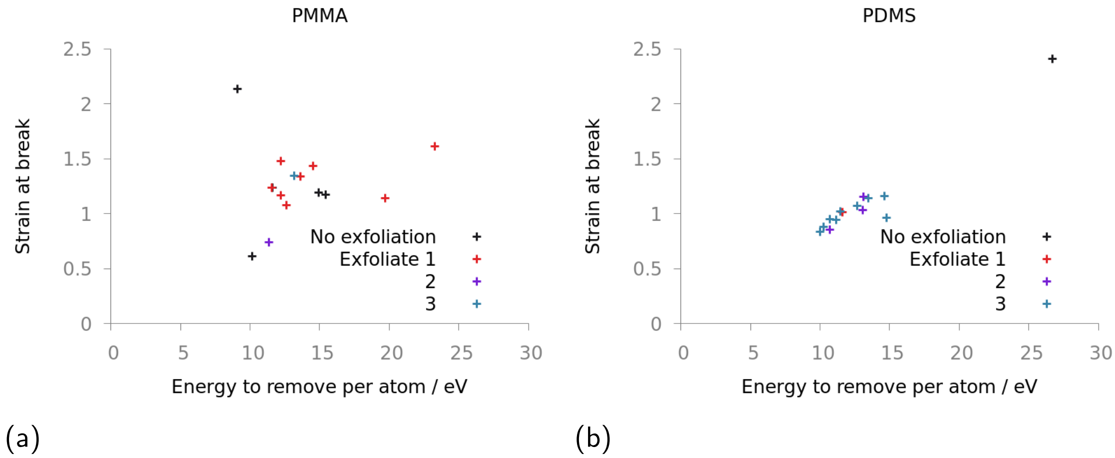


FIG. 2: Comparing the strain required to fracture simulations 4,5 (see table 1). PDMS produces a tight distribution compared to PMMA, the one exception being a simulation where no graphene was exfoliated.

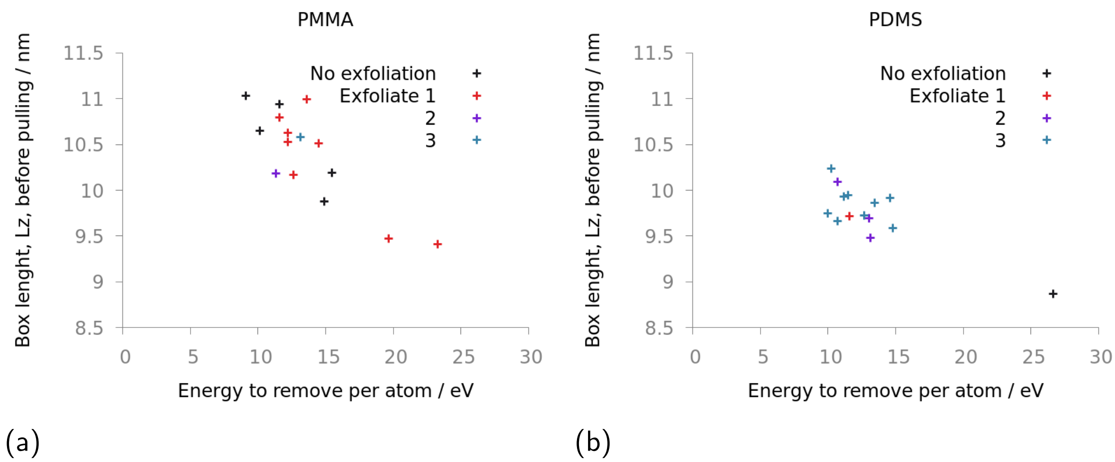


FIG. 3: Comparing the height of the simulation box before pulling of simulations 4,5 (see table 1). As discussed in the main text, PDMS is more fluid and therefore the polymer can deform around the graphite, the deformation makes the simulation boxes shorter after the compression stage.

for as long, and the outer layers are peeled apart from the main graphite.