

Supporting Information

Application of Soxhlet extractor for ultra-clean graphene transfer

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1. Optical and SEM Characterization.

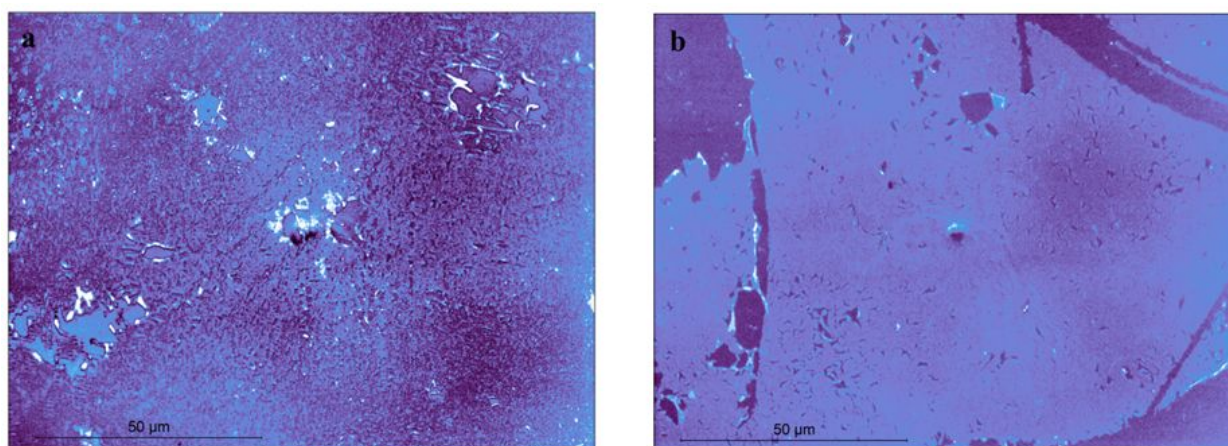


Figure S1. Optical microscopy images of graphene films from (left) conventional transfer and (right) Soxhlet assisted technique.

We did simple optical characterization of graphene films resulting from both conventional transfer (Fig. S1 left) and Soxhlet assisted techniques (Fig. S1 right) using Axio Imager Z2M upright Microscope. The initial optical scan of graphene films showed quite large area coverage of the substrate with graphene, however, the intermittent cracks seen on both techniques, although less pronounced in Soxhlet assisted method, were attributed to the damaging effect of PMMA during removal^{1,2}. It was also observed that the rough surface of growth substrate (i.e. Cu) left a distinctive pattern on the final graphene sheet transferred onto SiO₂/Si.

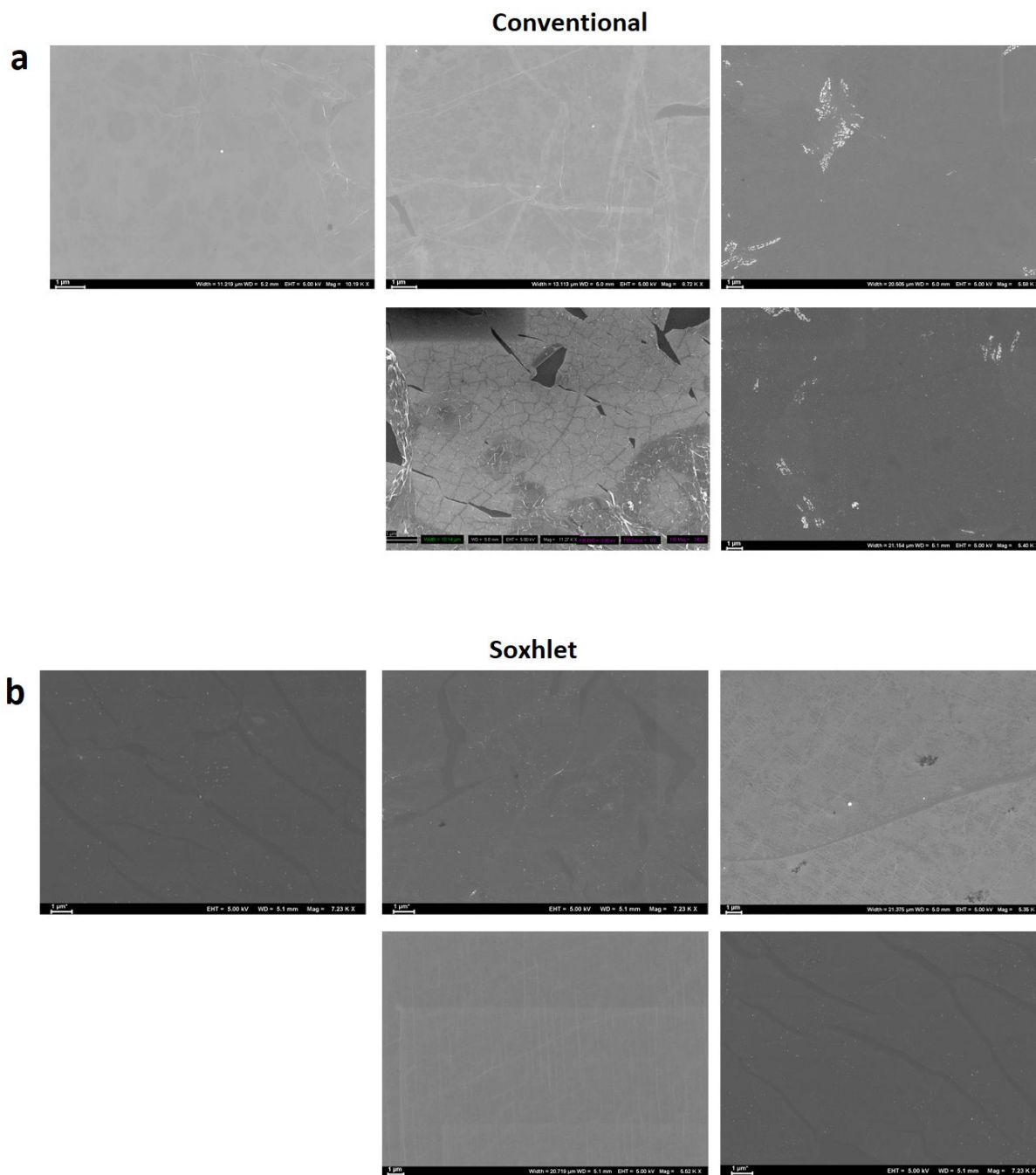


Figure S2. SEM images of repeated transfer process using the: (a) conventional transfer method, (b) Soxhlet assisted PMMA transfer method. A total of 5 different graphene transfer was carried out using under an identical condition thus eliminating sample-to-sample variability, further validating the outcome of the Soxhlet-enabled graphene process reported in this work.

2. Raman Characterization

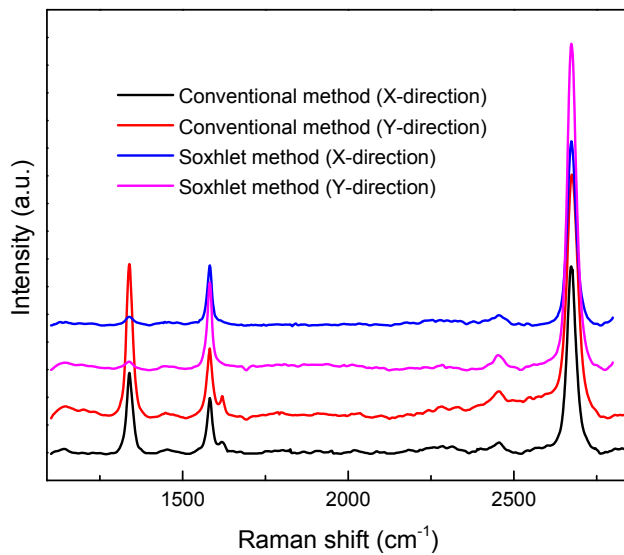


Figure S3. Micro-Raman spectroscopy scans in X- and Y-directions on the graphene transferred by conventional and Soxhlet methods.

To analyze sample quality we used characterization protocols developed by the leaders in the field^{3,4}. This analysis of statistical data extracted from Raman mapping is an established procedure for quantitative analysis of the transfer. Raman spectroscopy data has been obtained by using Horiba XploRA Raman Confocal Microscope. Fig S2 shows an initial large area scan in either X- or Y-direction on both samples revealed a significant defect in graphene transferred by the conventional method when compared with our Soxhlet method where the D-band is almost non-existent.

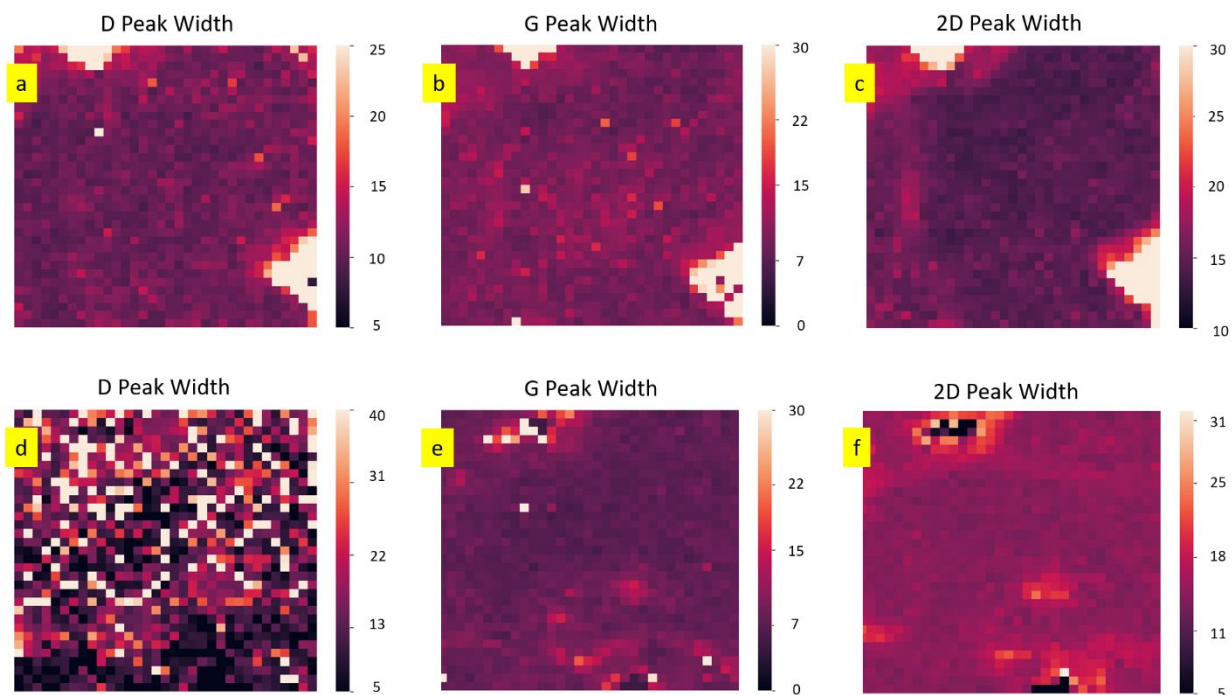


Figure S4. Width maps of fitted peaks obtained from Raman characterization ($10\times 10\ \mu\text{m}^2$) of graphene transferred with (a,b,c) conventional PMMA method; (d,e,f) Soxhlet assisted PMMA method.

Data fitting was achieved through use of Python. The primary package used was Lmfit, which allows for custom plots and constraints to be introduced. Additionally, a package named airPLS was used for background subtraction. Peaks were fit using a single Lorentzian peak each. Using this combination of packages allows for full width half maximum, center location, and amplitude for each peak to be referenced and used for creation of heatmaps.

Utilization of a custom program allows for easy importation, processing, and plotting of many different datasets with little input required. The downside to this method is the time required for each fitting; however, this can be counteracted by providing a more accurate range of potential values for the software to check. This method also allows for exportation of peak values in either text or Excel format for verification or plotting within different software.

For purposes of assessing quality, comparison between the two methods widths provides insight. All D, G, and 2D peaks are roughly the same width shown on the heatmap (Figure S4), especially the G and 2D peaks while D is more scattered in range. This is due to the low intensity of the D peak. With a low peak the fitting program begins to try and fit the background close, leading to a larger width. The Smaller widths for G and 2D go hand in hand with higher intensity quality of data acquisition.

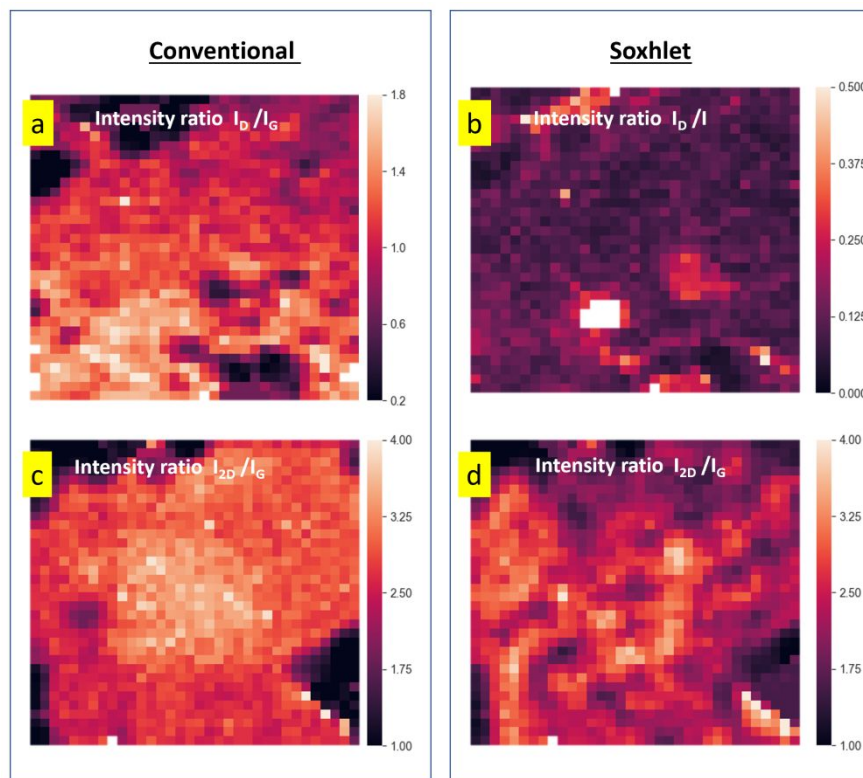


Figure S5. Correlation maps of the Raman D/G and 2D/G peak intensities of graphene transferred with conventional PMMA technique (a, c) and Soxhlet assisted PMMA technique (b, c); (b). Results for Soxhlet assisted PMMA transferred graphene clearly show that ID/IG is very close to 0.

References

- (1) Choi, W., Shehzad, M.A., Park, S. and Seo, Y., 2017. Influence of removing PMMA residues on surface of CVD graphene using a contact-mode atomic force microscope. *RSC advances*, 7(12), pp.6943-6949.
- (2) Shahzad, K., Jia, K., Zhao, C., Yan, X., Yadong, Z., Usman, M. and Luo, J., 2020. An improved rosin transfer process for the reduction of residue particles for graphene. *Nanoscale research letters*, 15(1), pp.1-11.
- (3) Banszerus, L., Janssen, H., Otto, M., Epping, A., Taniguchi, T., Watanabe, K., Beschoten, B., Neumaier, D. and Stampfer, C., 2017. Identifying suitable substrates for high-quality graphene-based heterostructures. *2D Materials*, 4(2), p.025030.
- (4) Ferrari, A.C., Meyer, J.C., Scardaci, V., Casiraghi, C., Lazzeri, M., Mauri, F., Piscanec, S., Jiang, D., Novoselov, K.S., Roth, S. and Geim, A.K., 2006. Raman spectrum of graphene and graphene layers. *Physical review letters*, 97(18), p.187401.