

Supporting Information

Roll-To-Roll Setup: The R2R transfer setup consists of three parts: a lamination unit, an electrochemical delamination unit and a rewind unit. The lamination unit is made up of two stainless steel cylinders (25 mm diameter, 65 mm length) that are filled with Teflon for thermal isolation and are mounted on aluminum shafts. The rollers are heated by two aluminum heat shoes with 100W cartridge heaters and thermocouples to control the temperature between 80 - 250°C. Clips are used to lightly press the heat shoes onto the steel rollers, which are slipping under the shoes as they rotate. The lamination speed can be altered between 0.7 – 5 mm/s by pulse-width-modulation controlled DC motor that is connected to the roller shafts by nylon gears. Additionally, the force with which the two rollers press together can be adjusted between 0 – 400 N by compressing the four springs that push the vertically movable bottom roller up against the clamped down top roller. The electrochemical delamination unit consists of a fixed Teflon roller that is immersed half way into an electrolyte filled Pyrex glass dish. Lastly, the rewind unit is responsible to pick up the delaminated film on separate spools. All spools are connected by gears and are powered another PWM controlled DC motor that can wind up films with a similar speed range of 0.7 – 5 mm/s.

Lamination onto Flexible Substrate: Polyethylene terephthalate (PET) films coated with ethylene-vinyl acetate (EVA) with total thickness of 75 μm (McMaster Product # 6927825) were used as flexible substrate. Unless noted otherwise a standard lamination process was performed as follows. Before lamination, two EVA coated PET film (19 mm x 89 mm) as well as one graphene-covered copper strip (12.7 mm x 75 mm) were rinsed in IPA and blow dried with a nitrogen blow gun. Both 50 mm wide rollers of the home-built lamination were heated to 135°C. The bottom roller was set to press against the top roller with 75 N. For lamination, the copper strip was sandwiched in between the two PET/EVA films and then placed in a paper sleeve (Aspen 30, 75 g/m²) to catch EVA that was oozing out from the sides. The speed of the lamination was set to 2 mm/s. After the lamination, the complete stack was trimmed with a paper guillotine to the dimensions of the copper strip.

Graphene Delamination: Unless otherwise noted in the text, the graphene was delaminated from the copper film as follows. Before performing the delamination the bottom EVA/PET film, top EVA/PET film as well as the copper and were extended by a PET and a copper strip, respectively, and then connected rewind rollers shown in **Figure 1b**) in the back of the picture. The plastic/Gr/Cu/Gr/plastic stack was delaminated with 1.3 mm/s in a 1 mol/l sodium hydroxide solution (NaOH, Macron Chemicals Product 7708-12). A voltage of 3.5 V was applied to the copper as cathode and a platinum gauze (25 mm x 25 mm, Alfa Aesar 10283) as anode, which resulted in a DC current of 100 mA for a typical delamination process.

Graphene Synthesis: The graphene used in this work was grown on 38 mm x 150 mm copper films (25 μ m thick, Alfa Aesar Product # 13882). Before growth, the surface of a copper strip was lightly etched in nickel etchant (Transene, Nickel Etchant TFB) for 90 s followed by a thorough rinse under deionized water. Next, it was bent along the long side in order to fit in a 25mm quartz tube furnace (Thermo Scientific, Lindberg Blue TF55035A-1). For growing the graphene, the copper was first gradually heated from room temperature (RT) to 1000°C in 20 min and was then annealed for 30 min while flowing 8 sccm of hydrogen (Airgas, HY UHP300) at a pressure of 0.38 Torr (Varian, SH-110). For growing the graphene layer, the gas flow was increased to 60 sccm of hydrogen and 3.5 sccm of methane (Airgas, ME UHP300) was added for 30min (pressure 1.95 Torr). Finally, the copper strip was cooled down to 600°C within 25 min while leaving the furnace closed. From there, it was cooled down to 100°C temperature within 15 min with a fan.

Hexagonal Boron Nitride Synthesis: The hexagonal boron nitride (hBN) used in this work was grown on 12 mm x 25 mm platinum foils (99.99 % pure, 50 μ m thick, Alfa Aesar Product # 42456) by chemical vapor deposition. The foil was placed in a 25mm quartz tube furnace (Thermo Scientific, Lindberg Blue TF55035A-1). For growing hBN, the platinum was first gradually heated from room temperature (RT) to 1050°C in 40 min and was then annealed for 30 min while flowing 300 sccm of hydrogen (Airgas, HY UHP300) at low pressure using a vacuum pump (Varian, SH-110). For forming the hBN layer, 0.1 sccm of hydrogen flowing through a borazine bubbler set to -4°C was added and maintained for 30 min. After that and while maintaining a hydrogen and borazine-rich gas flow, the temperature was slowly reduced to 700°C over the course of 20 min. Finally, all gases were shut off, the clam shell furnace was opened and cooled down to room temperature within 15 min with a fan.

Graphene Wet Transfer onto SiO₂/Si: A standard graphene wet transfer was performed as follows. The as-grown graphene on copper was spin-coated with PMMA A6 950 for 60 seconds at 2500 rpms and baked for 15 min in a 130°C oven. For the etch-based method the backside was etched for 10 sec in an oxygen plasma to remove the graphene there. For the bubble transfer method, the backside was also coated with PMMA using the same parameters. Two ways were used to remove the copper substrate:

1. For etch-based approach, the PMMA/Gr/Cu stack was placed in copper etchant (Transene CE 100) for 1 h. After fully removing the copper substrate, the floating PMMA/Gr membrane was transferred with a microscope glass slide into two consecutive DI water baths (5 min each) to clean the graphene surface. The membrane was then placed in a 10% hydrochloric acid bath for 5 min before being rinsed in two more DI water baths for another 5 min each to further clean remaining iron chloride residue.
2. For the bubble transfer approach, the PMMA/Gr/Cu/Gr/PMMA stack was first soaked in a 1mol/L solution of NaOH for 1h. To set up the bubble transfer, the Cu foil was attached to a clip connected to the minus terminal of a power supply and mounted on a scissor jack that holds the foil above the 1mol/L solution of NaOH. A platinum electrode was placed in the electrolyte solution and connected to the plus terminal of the power supply. To delaminate the PMMA/Gr stack, the foil

was slowly lowered into the electrolyte solution over a span of 1-2 min using the scissor jack. The current of the electrochemical lamination was kept between 100-200 mA, which corresponds to a voltage of 3.5 - 4.5 V. After the PMMA/Gr membranes were fully delaminated, they were rinsed in two consecutive DI water baths for 5 min each.

For the actual wet transfer, a piece of a silicon wafer with 300 nm thermal oxide was used to “scoop out” the floating PMMA/Gr on DI water. Subsequently, the membrane was blow-dried on the SiO₂/Si sample for approximately 30 sec using a nitrogen blow gun. Two bakes were used to drive out remaining water at the SiO₂/Gr interface: a 15 min bake at 80°C was well as a 15 min bake at 130°C. Lastly, the temporary PMMA transfer layer was cleaned off by submersing the sample in an acetone bath for 5 min followed by an isopropanol bath for 5 min and blow drying it thereafter.

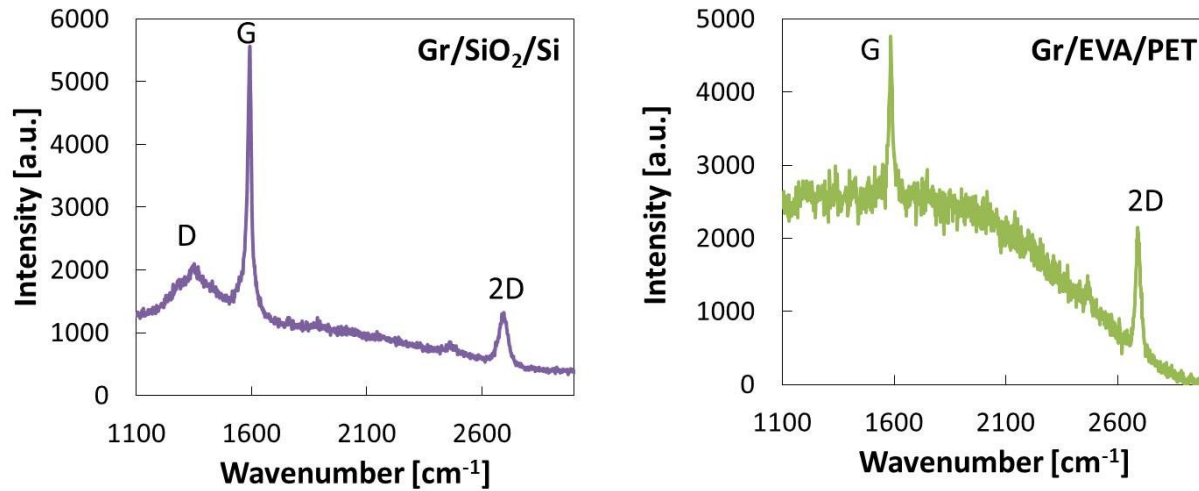
Characterization: After being delaminated both Gr/EVA/PET strips were rinsed under deionized water and adhered to a glass slide by double-sided tape (3M, 666). On the glass slide the strip was cut into 4 pieces measuring 1 cm x 1 cm each. Silver paint (TED PELLA, Leitsilber 200, Prod # 16035) was used for contacting all for corners. All electrical measurements were done using a home-built 4-probe hall measurement setup in conjunction with a HP 4156A Precision Semiconductor Parameter Analyzer. Atomic force microscopy scans were taken using a Digital Instruments Nanoscope IIIa. Optical micrographs were taken using a Zeiss Axio Scope A.1 in bright field mode. Scanning electron microscope images were obtained using a Zeiss Supra 40.

Lamination Temperature Dependence: The lamination temperature for the EVA/PET substrate used in this work was calibrated as follows. Two strips of EVA/PET film were placed together (with the EVA sides facing each other) in a paper sleeve (Aspen 30, 75 g/m²) to catch EVA that was oozing out from the sides. Subsequently, 5 different temperatures at a roller pressure of 50 N and lamination speed of 2 mm/s were tested: 80°C, 100°C, 110°C, 120°C, and 130°C with the optical results shown in **SI Figure 1**. It shows that the EVA film completely melts at 120°C as indicated by the film turning completely transparent. To have additional temperature margin, the standard lamination temperature was chosen to 130°C.



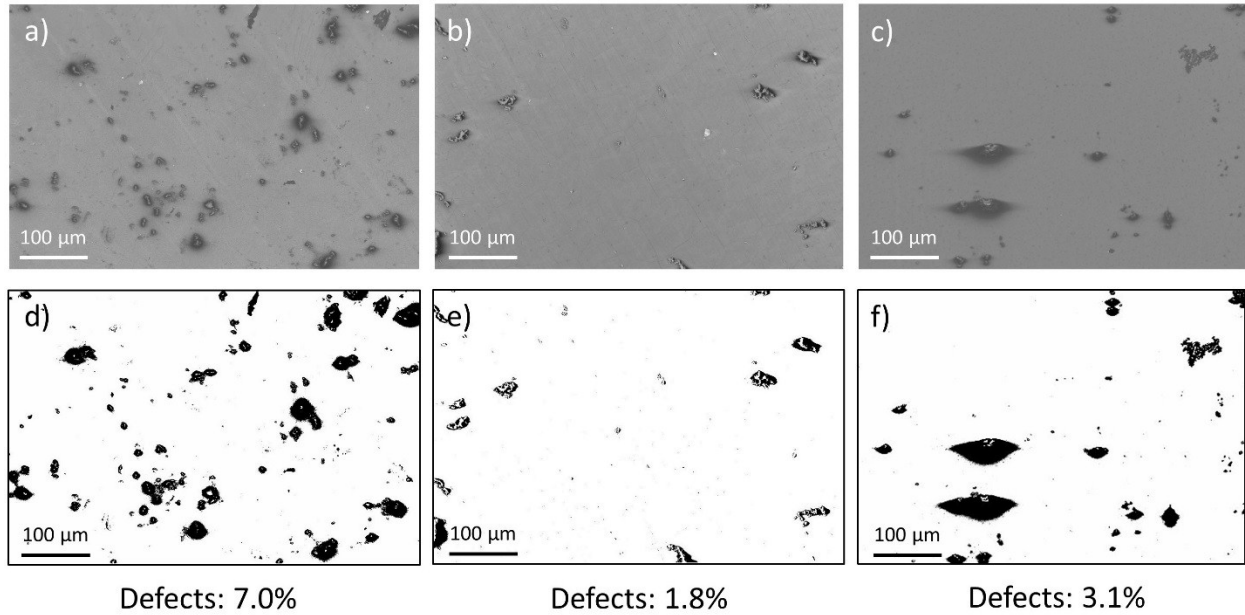
SI Figure 1: Determining minimum viable lamination temperature for EVA/PET film. Temperatures from 80-130°C were tested at a roller force of 50 N and a lamination speed of 2 mm/s

Raman Spectroscopy of Gr/EVA/PET: The graphene transferred onto EVA/PET was also characterized by Raman spectroscopy, see below. The Raman spectrum on EVA is averaged over 5 spectra and the background spectrum of EVA/PET was subtracted.



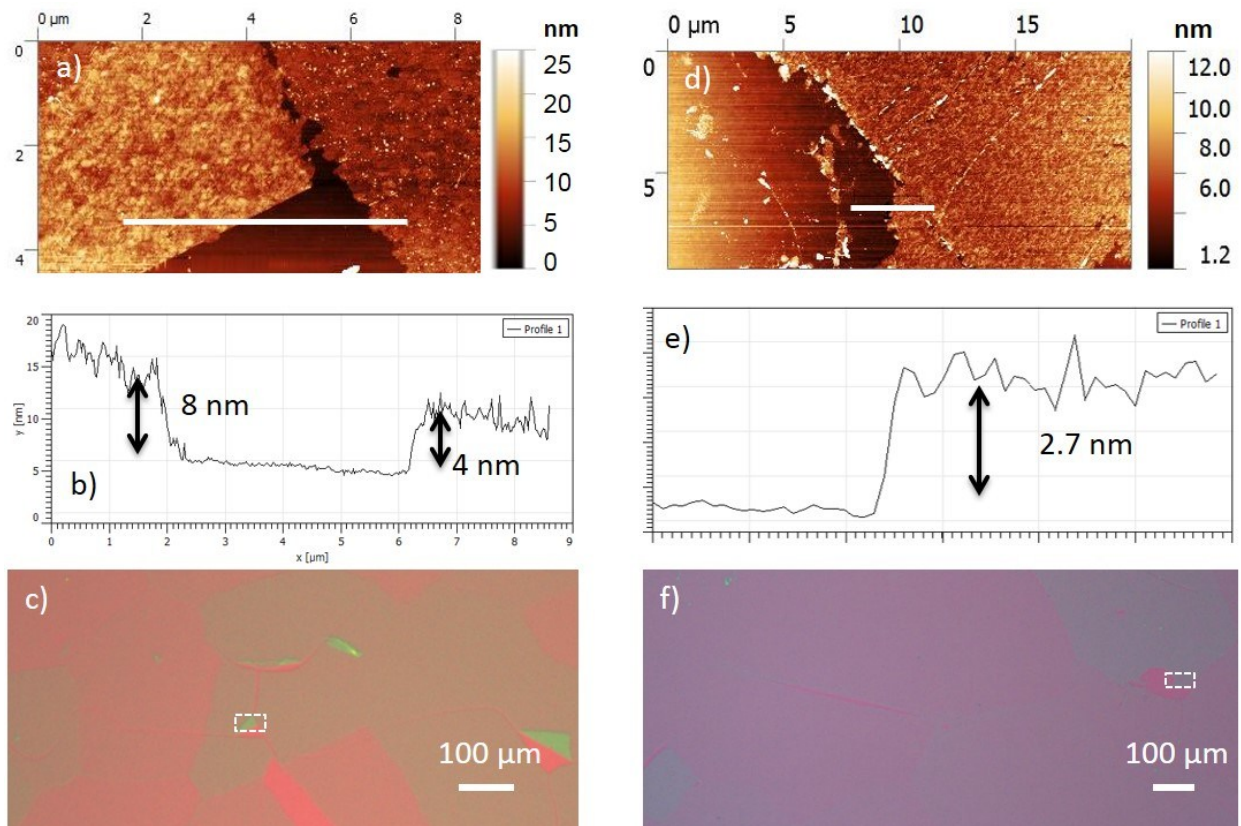
SI Figure 2: Raman spectrum of graphene on SiO₂/Si (left) and graphene on EVA/PET right.

Crack Density Analysis of Gr/EVA/PET: SEM images of Gr/EVA/PET samples were analyzed to roughly estimate the defect and crack density in the transferred films. Three examples of typical graphene transfer results on EVA/PET are depicted in **SI Figure 3 a-c)**. Defects in the film can be easily spotted by the darker regions in the images. The dark areas come about from charging effects of the non-conductive EVA/PET substrate. In areas where graphene is missing the electrons accumulate in the EVA layer and cannot flow off. As a result, the SEM image is darker in these areas. To get a conservative estimation of defect density, the image contrast was increased to 100% and the color resolution was reduced to 4 colors, see **SI Figure 3 d-f)**. The resulting white area A_{white} in these images was interpreted as undamaged graphene. Hence the area of damaged graphene was calculated as $A_{\text{damage}} = 100\% - A_{\text{white}}$. The average cracked and damaged graphene area is roughly 5% in the evaluated samples.



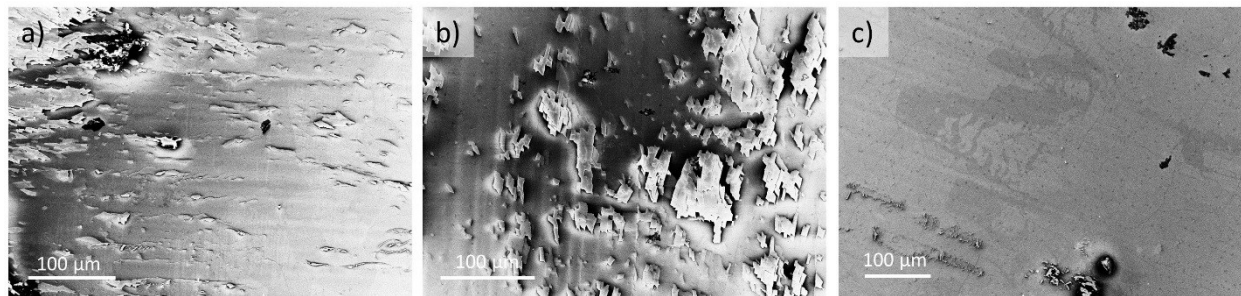
SI Figure 3: Crack and defect density analysis of Gr/EVA/PET. a-c) SEM images of Gr/EVA/PET. d-f) Fully increased contrast of images a-c).

AFM results of hBN reference sample: **SI Figure 4 a-b)** shows AFM scans of two sites on hBN reference samples on SiO₂. **SI Figure 4 c-d)** represents the corresponding cross-section profiles along the white lines in a) and b). Lastly, **SI Figure 4 e-f)** shows optical micrographs of the larger vicinity around the AFM scan areas with the black, dashed boxes indicating the exact AFM scan location.



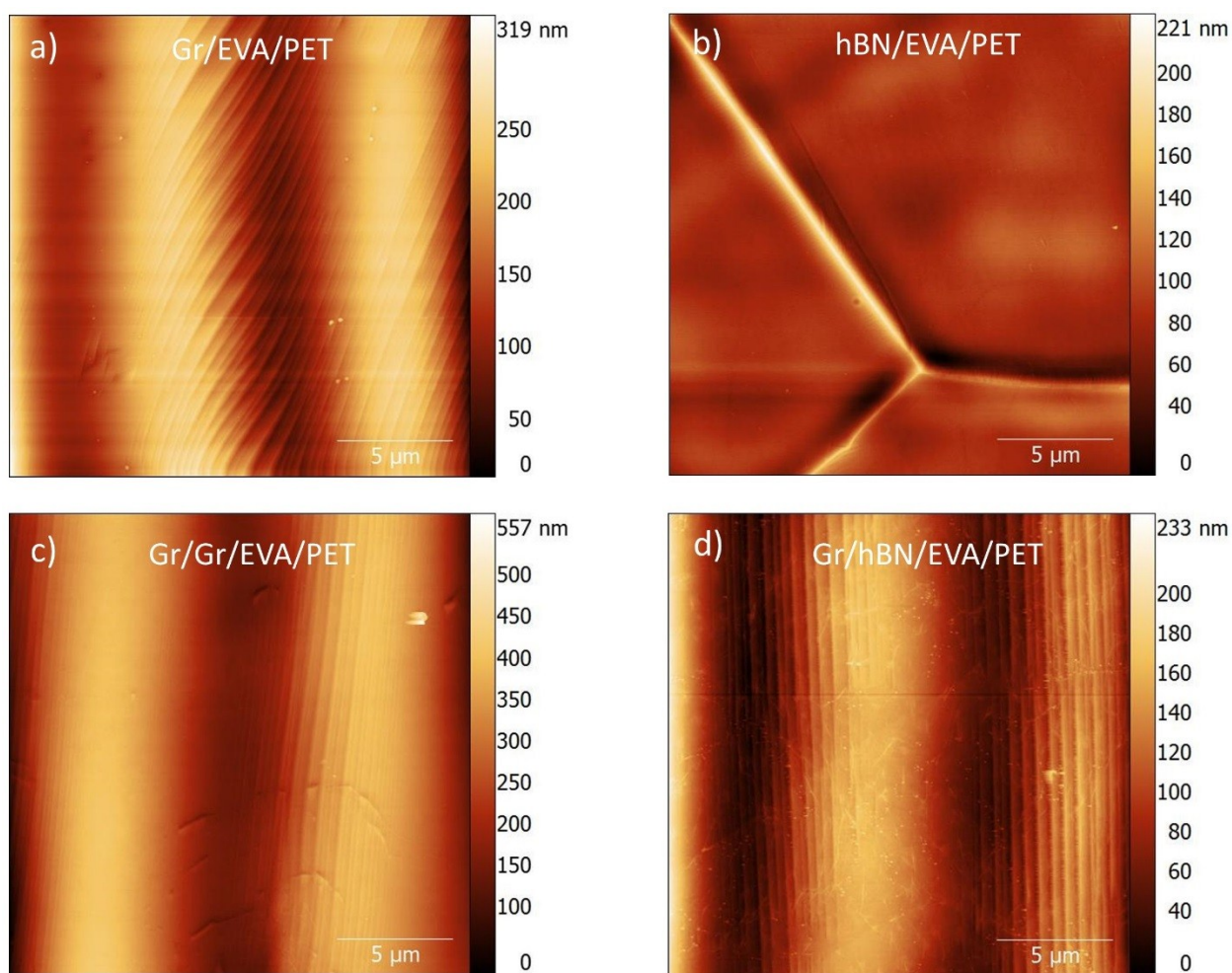
SI Figure 4: a-b) AFM scans of hBN on from two separate sites. c-d) corresponding cross-section profiles along the white lines in a) and b), e-f) optical micrographs of site 1 and 2 with dashed box indicating scan area.

SEM Images of Gr/hBN/EVA/PET: Further SEM images of Gr/hBN/EVA/PET were acquired from different samples, see **SI Figure 5 a-c)**. They show a many charging artifacts that are indicative of cracks in the graphene layer.



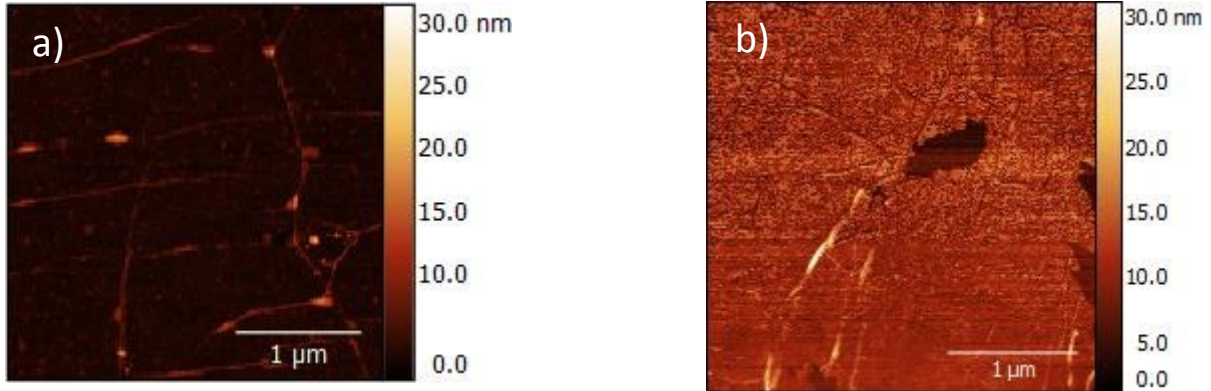
SI Figure 5: a-c) Large-area SEM images of different Gr/hBN/EVA/PET samples showing charging artifacts that indicate graphene defects.

Comparison of Gr/hBN/EVA/PET and Gr/Gr/EVA/PET by AFM: To better understand the differences in transfer quality of graphene onto Gr/EVA/PET compared to graphene onto hBN/EVA/PET, AFM images were acquired before and after transfer of the second lamination. **SI Figure 6 a-b)** shows the surfaces of Gr/EVA/PET and hBN/EVA/PET, respectively. It can be seen that the surface morphology strongly differs. While, the surface of Gr/EVA/PET is wavy with many small step terraces, as discussed in the manuscript, the surface of hBN/EVA/PET is generally smooth with large connected ridges that arise from the negative imprint of the Pt grain boundaries. The AFM results after graphene transfer are shown in **SI Figure 6 c-d).** Regardless of their previous surface topology, both the Gr/Gr/EVA/PET as well as Gr/hBN/EVA/PET now show the characteristic waviness and step terraces of the copper foil, at least down to a nanometer scale. Hence, it can be concluded that regardless of the initial morphology, both substrates conform similarly well to the copper topology after the second lamination process.



SI Figure 6: a-b) AFM images of Gr/EVA/PET and hBN/EVA/PET showing significant differences in morphology features. c-d) AFM images of Gr/Gr/EVA/PET and Gr/hBN/EVA/PET both having the typical morphology of annealed copper foil.

Surface Roughness Comparison of Graphene and hBN on SiO₂: Both hBN and graphene were wet transferred onto SiO₂ using a bubble delamination (as described above). Both surfaces were imaged with AFM and their average surface roughness was measured. **SI Figure 7 a-b)** shows two representative scans for graphene and hBN on SiO₂. It shall be noted that the surface roughness for hBN is more variable than graphene. Thicker regions have a higher surface roughness. The average surface roughness for Gr/SiO₂ and hBN/SiO₂ are 0.4 nm and 1.2 nm, respectively.



SI Figure 7: a-b) AFM images of Gr/SiO₂ and hBN/SiO₂ to determine surface roughness.

Conversion of Graphene-hBN Binding Energy to Adhesion Energy: Sachs et. al. [1] simulated the binding of graphene to hBN. They calculated the energy difference of 5 different stacking configurations of a graphene/hBN unit cell compared to the layers being far apart. These energy values are: -62 meV, -65 meV, 65 meV, -83 meV, -71 meV and -70 meV with an average of **-69.3 meV** for a unit cell of 2 carbon atoms, 1 nitrogen atom and 1 boron atom. Using a lattice constant of 0.25 nm and a unit cell angle of 60° for hBN [2], the number of unit cells U per square meter is:

$$U = \frac{1}{0.25 \text{ nm} * 0.25 \text{ nm} * \sin(60^\circ)} = 1.8475 * 10^{19} \text{ m}^{-2}$$

The adhesion energy A of graphene on hBN can be estimated as average binding energy multiplied by the number of unit cells per meter.

$$A = 69.3 \text{ meV} * 1.8475 * 10^{19} \text{ m}^{-2} = 0.205 \text{ J/m}^2$$

References:

1. Sachs, B., et al. (2011). "Adhesion and electronic structure of graphene on hexagonal boron nitride substrates." *Physical Review B* **84**(19).
2. Ooi, N., et al. (2006). "Structural properties of hexagonal boron nitride." *Modelling and Simulation in Materials Science and Engineering* **14**(3): 515-535.