Supporting Information

Ultra-large suspended graphene as highly elastic membrane for capacitive pressure sensor

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S1. The following figures and tables are information on the comprehensive growth conditions and characterizations for the optimization of single crystalline graphene domains.

In this study prior to the CVD growth process, Cu foil(Alfa Aesar 99.8%; 25μm) was subjected to a surface flattening process by electrochemical polishing, where the Cu foil was placed in the anode and a Cu plate was the cathode with a separation of 10 cm in an electrolyte mixture of 70 wt% H3PO4(Nihon Shiyaku 85%) with polyethylene glycol(Alfa Aesar,Polyethylene Glycol 6,000, PEG6,000) as additives. The setup of this process in shown in FigureS1-1 (a). Figure S1-1(b) shows the microscope images of as-received and polished Cu substrates. By testing various polishing times of 3, 6, 9, 12 min, the surface roughness was measured and listed in Table S1-1. The optimized polishing time was found to be 9 min, where the surface roughness was 89 nm. It can be noted that the Cu foil thickness should not be less than 16 μm because pin-holes can occur during the graphene growth process. Therefore, the optimal condition for the substrate is polishing for 9 min.

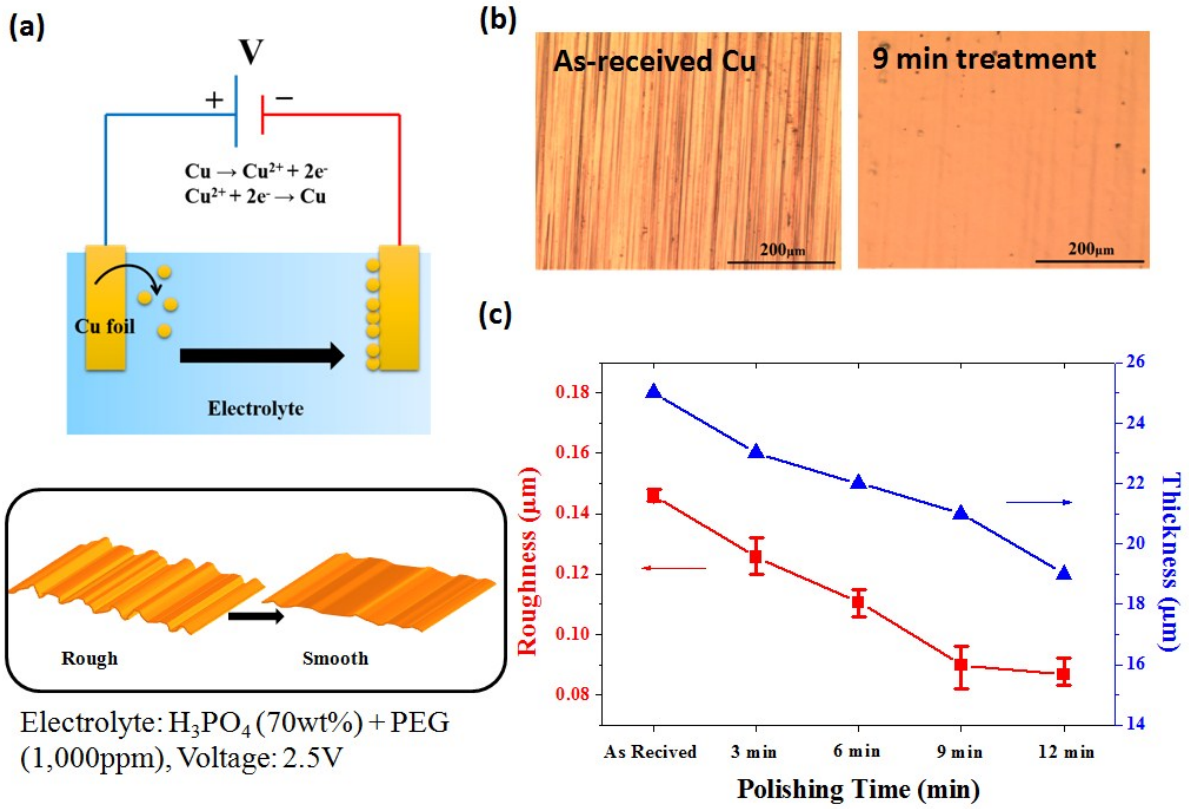
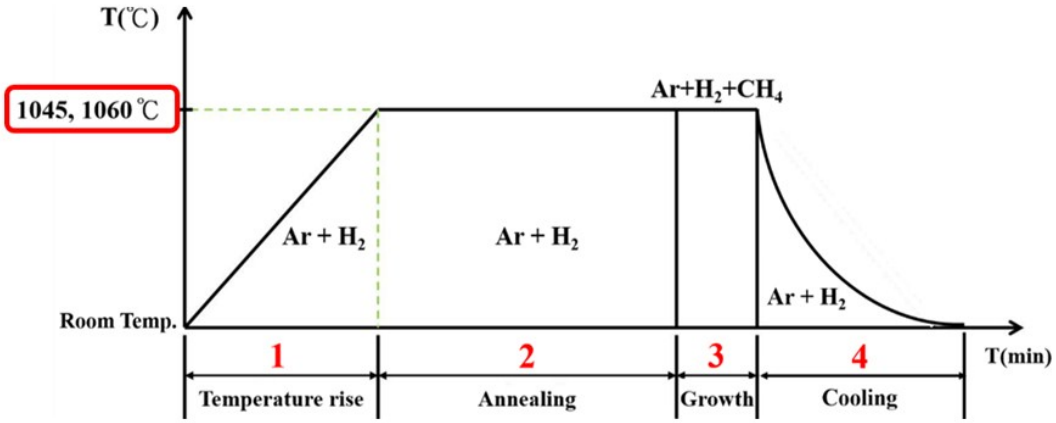


Figure S1-1. The surface flatten process by electrochemical polishing method. (a) Experimental setup for this process.(b) The optical images for the polished samples (c) The correlation between surface-roughness/ substrate thickness and polishing time.

Table S1-2. Surface roughness of the copper substrate with various polishing times.

EP time	Average roughness, Ra (μm)	Maximum(μm)	Minimum(μm)
As-received	0.1458	0.148	0.144
3 min	0.1256	0.132	0.12
6 min	0.1106	0.115	0.106
9 min	0.0898	0.096	0.082
12 min	0.0868	0.092	0.083



Gas flow\ Step	1	2	3	4
H_2 (sccm)	20	10,20,30,40,50	10,20,30,40,50	10
Ar (sccm)	80	1,000	1,000	1,000
CH_4(sccm)	0	0	0.5	0
Pressure	540mTorr	760Torr	760Torr	760Torr

Figure S1-2.Procedures for the CVD grown graphene on the Cu substrate.

Table S1-2 Comprehensive study on the growth conditions of CVD for optimizing the large single crystalline domains of graphene. Note that the pressure was fixed at 760 Torr.

Temperature		G-band	2D-band			
	H_2 flow rate (sccm)	**FWHM(cm^{-1})**	**FWHM(cm^{-1})**	**I_D/I_G**	**I_{2D}/I_G**	**Average domain size(μm)**
1045°C	10	22.33	39.82	0.029	0.991	8.67
	20	24.51	39.74	0.055	1.321	52
	30	24.5	38.7	0.049	1.501	22.9
	40	22.83	42.06	0.044	0.822	16.36
	50	25.24	41.1	0.064	1.399	24.36
1060°C	10	25.24	38.46	0.064	1.590	7.27
	20	22.99	37.66	0.053	1.607	34.87
	30	20.47	38.24	0.015	1.217	54.28
	40	22.43	36.05	0.061	1.868	14.8
	50	23.63	45.43	0.092	1.040	5.8

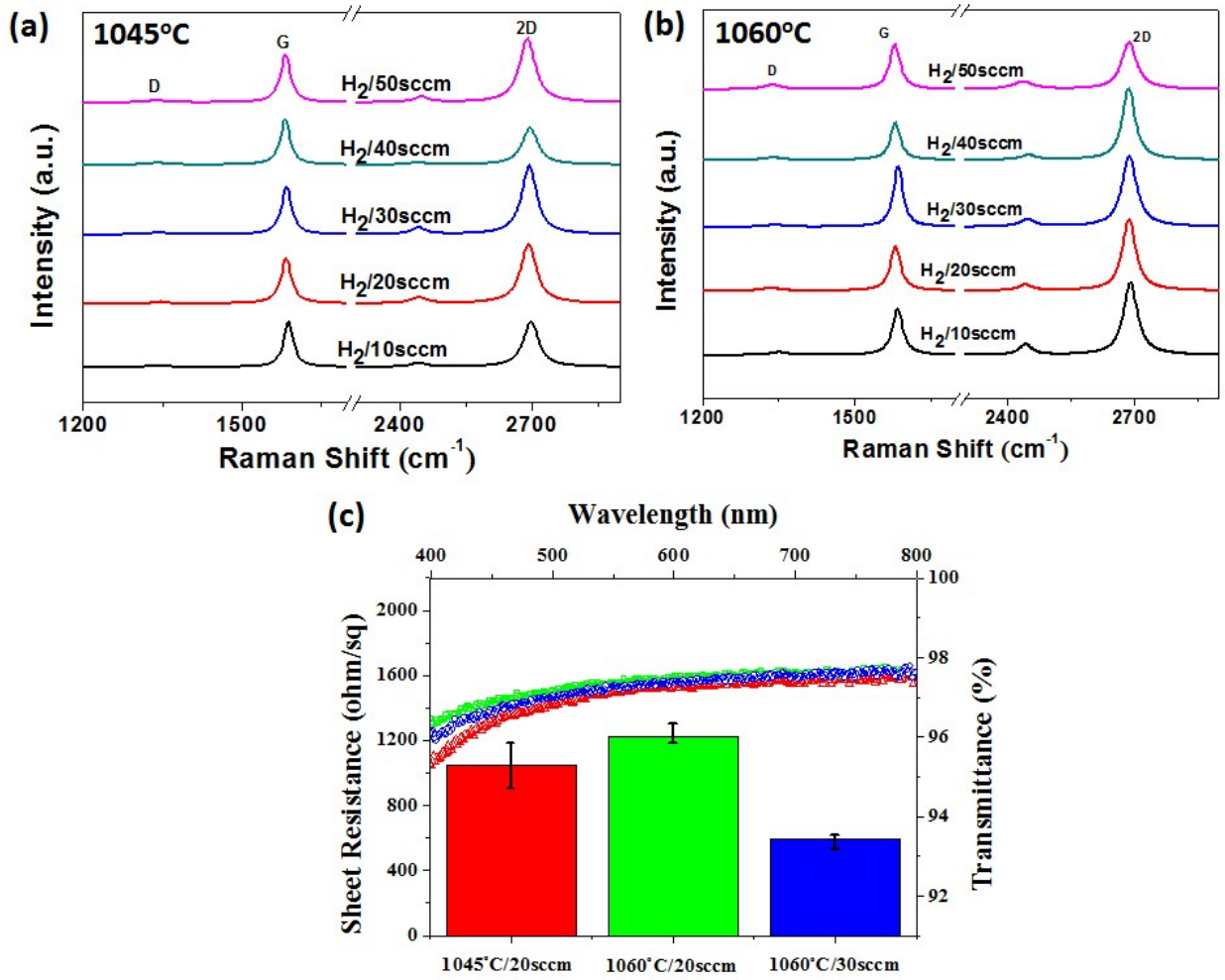


Figure S1-3. Raman spectra of the samples carried out with various growth conditions at temperatures of (a)1045°C and (b)1060°C. (c) The correlation of sheet resistances and optical transmittances for three different samples. The results were detailed in Table S1-3.

Table S1-3 Summary of the structure, sheet resistance, and the optical transmittance for three growth conditions. The results indicate that the case of 1060°C/H_2 30 sccm yielded the best graphene quality for this study.

Temperature	H_2 flow rate (sccm)	I_D/I_G	I_{2D}/I_G	G-band FWHM (cm^{-1})	2D-band FWHM (cm^{-1})	Sheet resistance (ohm/ sq.)	Transmittance (%)
1045℃	20	0.025	1.552	24.23	40.85	1044 ± 140.5	97.21
1060℃	20	0.029	1.518	25.62	43.65	1221 ± 62.5	97.37
1060℃	30	Tiny*	1.464	20.03	33.21	592 ± 45.5	97.27

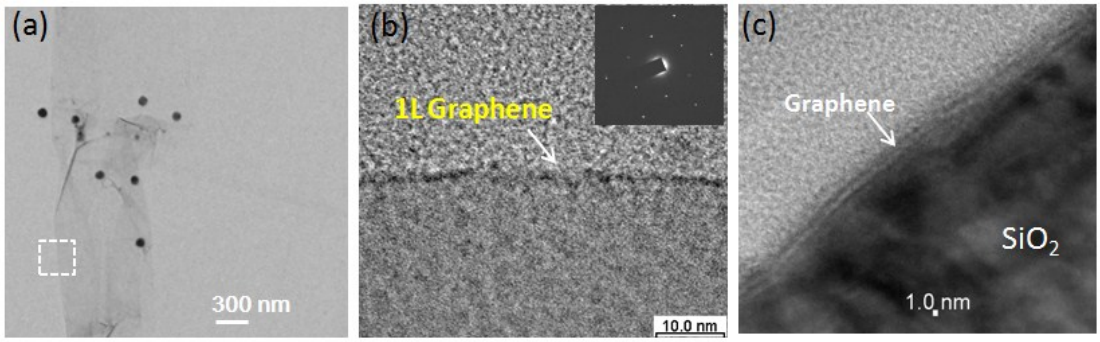


Figure S2. The cross-sectional TEM images on folded region of (a)(b)a transferred graphene film on copper grid and (c) the another one on SiO2 substrate.

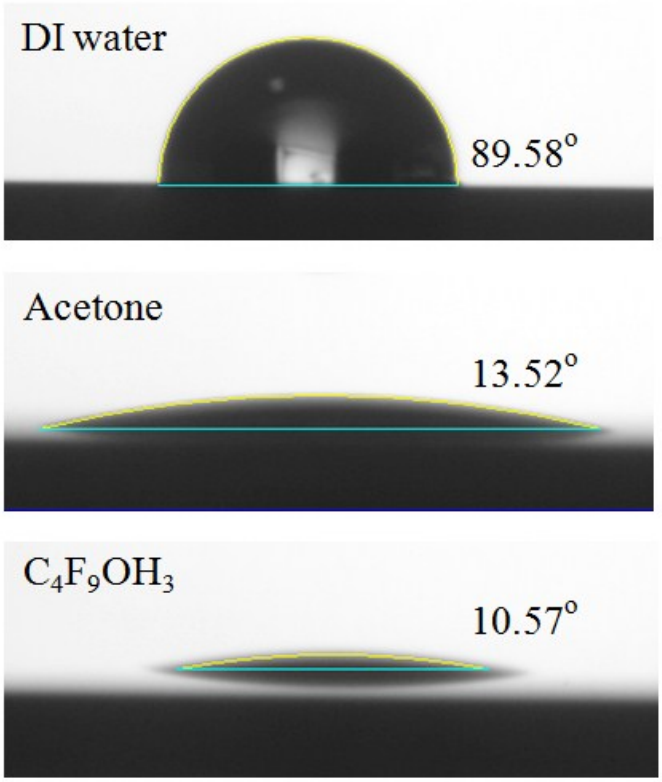


Figure S3. Contact angle measurements for graphene on Si/SiO_2, suggesting a low surface tension for the solvent of methoxynonaflurorbutane ($C_4F_9OH_3$).

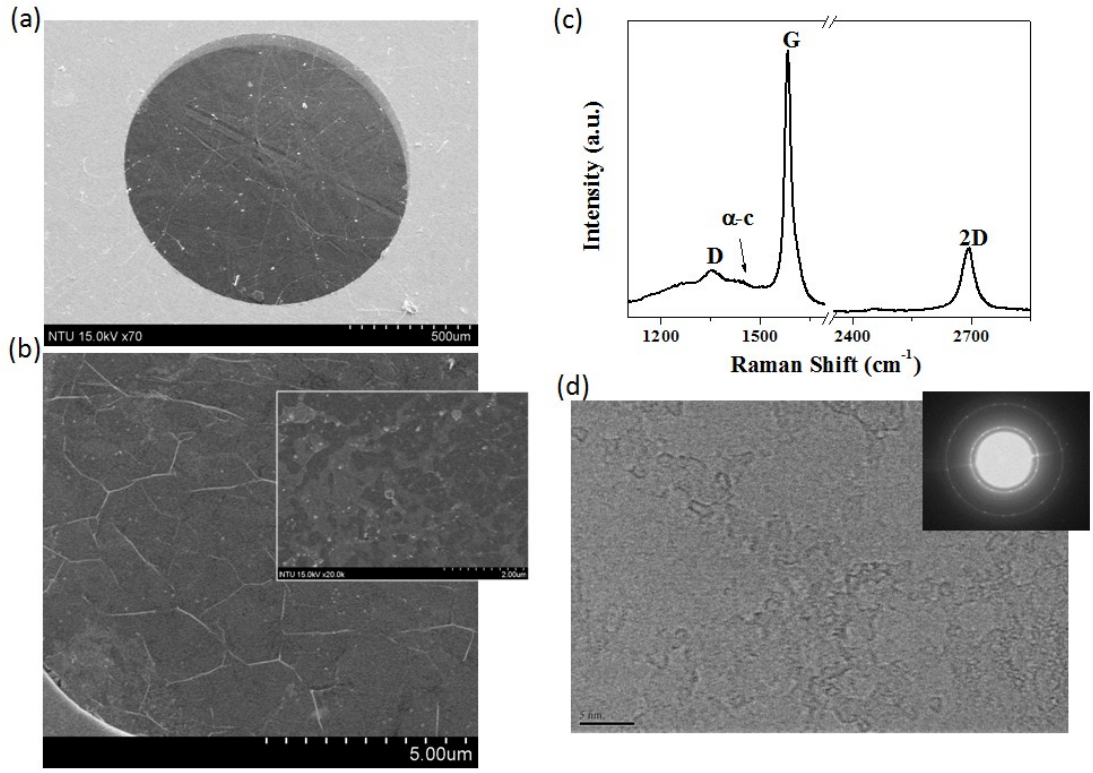


Figure S4. Suspended graphene sample prepared by stacking 5L graphene and using the thermal decomposition method. (a) Low magnification SEM image and (b) high magnification image. (c) Raman spectrum for the graphene membrane. (d) HRTEM image of the nanostructure of the graphene membrane, indicating the amorphous phase of carbon structure distributed over the surface.

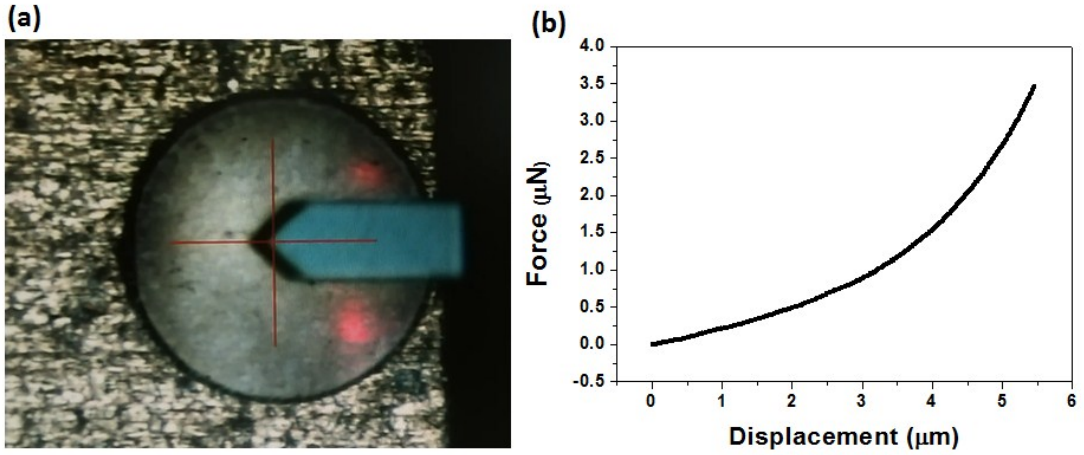


Figure S5. (a) The force curve for the suspended graphene membrane(500 μm hole) recorded by AFM (the elastic properties can be observed in the video clip). (b) The correlation of applied force(μN) with the displacement(μm).

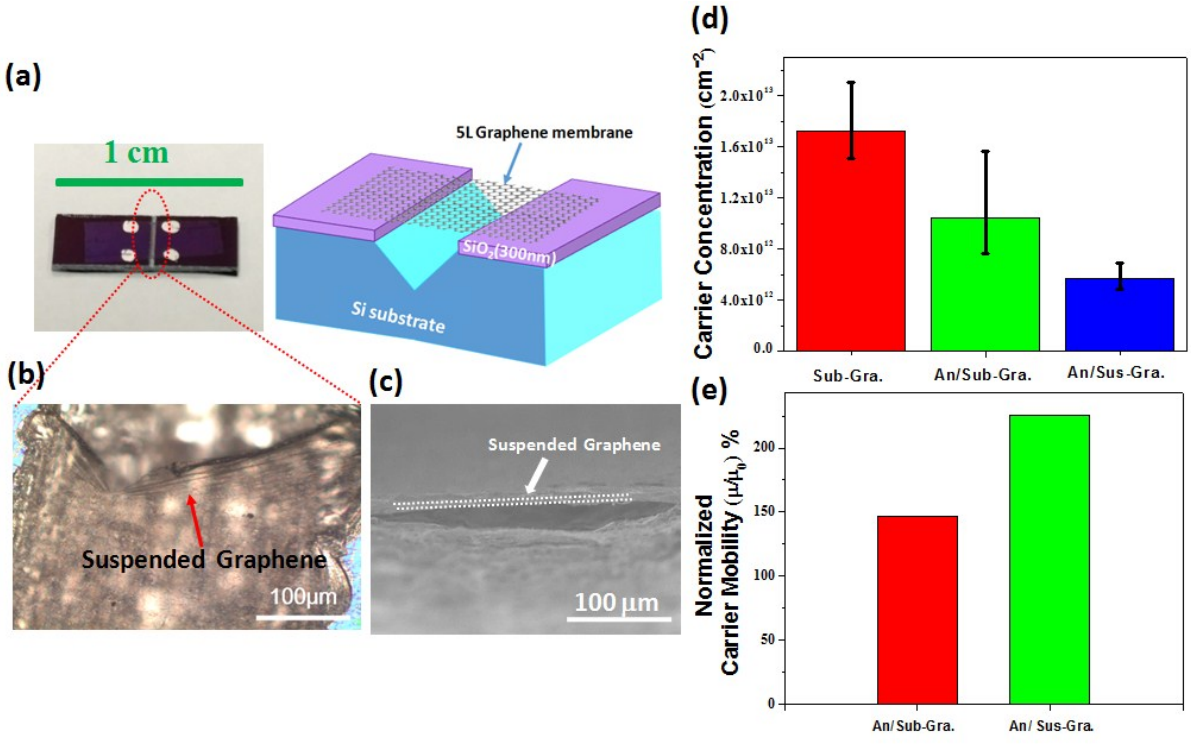


Figure S6. Graphene membrane on a trenched substrate.(a)Photograph of the prepared sample and its structural illustration, where the suspended graphene membrane bridged over a long(~ 0.5 cm) trenched substrate(SiO_2/Si) with a width of approximately 300μm.(b)Optical microscopy image of the top-view of the sample. (c)

SEM cross-sectional image (tilted angle of ~5°) of the graphene membrane. (d) and (e) Comparison of the carrier concentration and the normalized carrier mobility, which was obtained by Hall measurements(with respect to the graphene on the substrate, Sub-Gra.) of substrate-supported graphene after annealing treatment(denoted as An/Sub-Gra.) and suspended graphene after annealing treatment(denoted as An/Sus-Gra.). The result shows that the hole carrier concentration for An/Sus-Gra. is 37-fold lower than that of Sub-Gra., indicating the significantly reduced charge doping from the substrate. In addition, (e) the normalized carrier mobility for the suspended graphene is 224% higher than that of the substrate supported graphene sample. Note that the measured mobility for Sub-Gra. ranged from 800~950 cm^2/vs for the 6 samples and averaged at 920 cm^2/vs(μ_0). And the averaged mobility for annealed suspended graphene(labeled as An/Sus-Gra.) was 2060.8 cm^2/vs(μ), therefore the normalized mobility is 2060.8/920= 224(μ_0/ μ)%.

Table S7 Summary of this work compared to other techniques

Method	**Suspended diameter**	**Reference**
Supercritical drying	~10 μm^2	Jannik C. Meyer et al. [1]
Polymer transfer	~55 μm	Benjamĭn Alemăn et al. [2]
Inverted floating	500 μm	Choong-Kwang Lee et al. [3]
Transfer with supporting frame*	~ 7mm	Qin Zhou et. al. [4]
Thermal decomposition	1.5 mm	this work(5L graphene)
Thermal decomposition	600 μm	This work(1L graphene)

* Note that the membrane is composed of multi-layered graphene with ~60 layers, which was obtained by CVD-graphene on Ni substrate.

[1] Meyer, J.C., et al., *The structure of suspended graphene sheets.* Nature, 2007. **446**(7131): p. 60-63.

[2] Aleman, B., et al., *Transfer-Free Batch Fabrication of Large-Area Suspended Graphene Membranes.* Acs Nano, 2010. **4**(8): p. 4762-4768.
[3] Lee, C.K., et al., *Monatomic Chemical-Vapor-Deposited Graphene Membranes Bridge a Half-Millimeter-Scale Gap.* Acs Nano, 2014. **8**(3): p. 2336-2344.
[4] Q. Zhou, J. Zheng, S. Onishi, M. F. Crommie, A. K. Zettl, Proceedings of the National Academy of Sciences 2015, **112**, 8942.