

# Crack- and contamination-free transfer of graphene films

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## Abstract

Owing to its fascinating properties, graphene has exhibited great potentials in the applications of high-performance electronics, flexible devices and encapsulation. The availability of graphene on a wide range of substrates forms the basis for large-area applications, such as graphene integration with silicon-based technologies, which requires graphene on silicon with outperforming carrier mobilities. In this regard, although currently, large-area graphene films have been successfully produced based on chemical vapor deposition methods, with outperforming quality and uniformity. Graphene films were only produced on limited archetypal substrates, such as metal foils. Reliable after-growth transfer techniques, that do not produce cracks and contamination are critical for layering 2D materials onto arbitrary substrates for further applications. During the transfer of graphene, with atomically thin and highly flexible nature, polymer-based transfer medium is usually introduced for avoiding the formation of cracks and wrinkles, which, however, introduces unavoidable contamination on graphene surface. In addition, traditional transfer routes, including etching and bubble-based delamination, involve the aqueous solution-based reactions or processing, which would induce the water-related p-doping in graphene and improve the complexity in the design of industrial equipments for batch transfer.

Here, in our group, based on the structural design of transfer medium and careful modulation of interfacial forces, we have achieved the crack, contamination and wrinkle-free transfer of graphene films over large area: (1) we achieved the mechanical delamination of graphene both from the Cu surfaces and from the polymer surface, based on the uniform oxidation of Cu substrates and the controllable tuning of the interaction between graphene and polymer, and all the transfer process were conducted without using water or organic solvent; (2) we showed that, by incorporating oxyhydril groups-containing volatile molecules, the supporting films can be deformed under heat to achieve a controllable conformal contact, enabling the large-area transfer of graphene wafers without cracks, contamination, and wrinkles onto silicon wafers, and the ultraclean surfaces provide the carrier mobilities of transferred graphene up to  $14,000 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$  at room temperature; (3) we achieved the preparation of high-performance water barrier films based on layer-by-layer transfer of A3-sized graphene films, and the water vapor transmission rate of double-layer graphene can be as low as  $5 \times 10^{-3} \text{ g}/(\text{m}^2 \text{ d})$ , which is one order of magnitude lower than previously reported values; (4) we designed and built the transfer systems for the batch transfer of graphene films, including the automatic spin-coater, delaminator and laminator

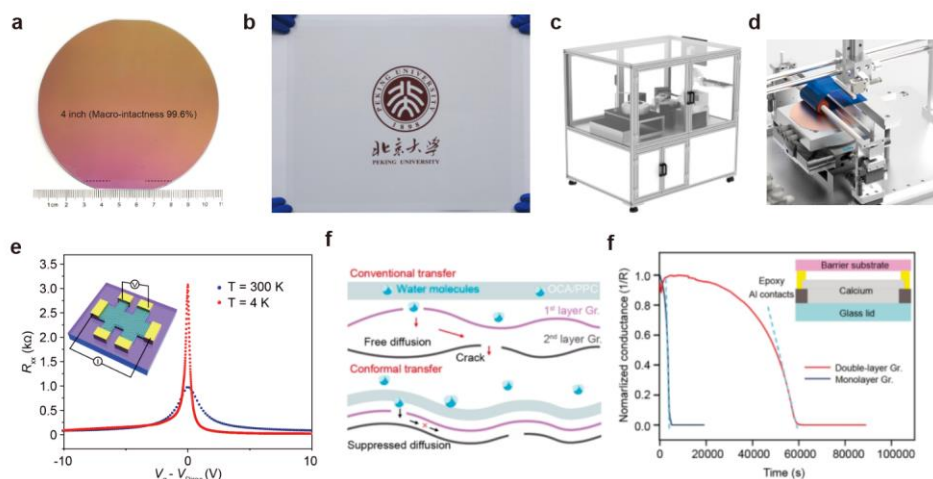
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## References

[1] Yixuan Zhao, Yuqing Song, Zhaoning Hu, Wendong Wang, Zhenghua Chang., et al. Large-area transfer of two-dimensional materials free of cracks, contamination and wrinkles via controllable conformal contact. *Nat. Commun.* , 13(2022), 4409.

[2] Yuqing Song, Wentao Zou, Qi Lu, Li Lin, Zhongfan Liu. Graphene transfer: Paving the

Figures



**Figure 1:** The crack, contamination and wrinkle-free transfer of large-area graphene films and the related applications.