The Roadmap of Graphene: From Fundamental Research to Broad Applications

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Owing to its atomically thin structure and fascinating properties, graphene has emerged as an important material in modern physics and chemistry. Tremendous breakthroughs have been made in the mass production of graphene materials and the development of a wealth of applications in electronics, photonics, batteries, wearables, and sensors. Currently, commercially available graphene products can be divided into graphene films, graphene oxide (GO), and reduced graphene oxide (rGO), and the mainstream preparation techniques include chemical vapor deposition (CVD) and chemical exfoliation. The global graphene market size exceeded USD 4 billion by 2021, and over 20000 companies are currently engaged in graphene-related businesses. Scientists, engineers, and enterprises are working in tandem to find scalable approaches to producing graphene with reliable quality and performance, and to unveiling novel and revolutionary applications. The success of graphene industrialization and commercialization will be promising through joint academic and industrial efforts.

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This special issue organized by Profs. Jin Zhang, Hailin Peng, Hua Zhang, Shigeo Maruyama, and Li Lin, provides a body of high-impact reviews and research articles in the fields of synthesis, engineering, and application of graphene materials. We proposed this special issue in *Advanced Functional Materials* on the occasion of the 60th birthday of Prof. Zhongfan Liu, a world-famous expert in graphene research. This special issue consists of 16 reviews and 14 research articles, focusing on the latest summary and progress from fundamental research to broad applications of graphene. This special issue will include a range of sub-topics, such as the controllable synthesis of graphene films, powders, and fibers, innovative production techniques for specific applications, structural and functional engineering of graphene architectures, novel characterization of graphene materials by synchrotron radiation, and advanced applications of graphene materials and composites. We believe that the topics in this special issue of *Advanced Functional Materials* would provide pivotal viewpoints relevant for promoting the mass production and commercialization of graphene, and are valuable for a broad audience of academia and industry professionals engaged in graphene-related research and business.

The commercial success of graphene materials should stand on the reliable technique for the mass production of graphene products with fine control over the quality, cost, and reproducibility. The CVD approach can produce graphene films and powders with excellent scalability and quality at high temperatures. In the last decade, great progress has been made in the controllable CVD synthesis of graphene products, exemplified by wafer-scale single-crystal graphene, wrinkle-free graphene, and the successful removal of surface contamination. Prof. Feng Ding et al. (article number 2203191) provide a detailed summary of the achievements and challenges of graphene CVD synthesis from a theoretical perspective. Future focus is also detailed on the growth of graphene at relatively low temperatures, insulating substrates, and fine control over the thickness and interlayer stacking order. Prof. Yanwu Zhu (article number 2203894) systematically investigates the structural evolution from C60 fullerene crystal to other nanocarbon materials, such as graphene, by combining neural network potential and stochastic surface walking methods.

Prof. Haofei Shi et al. (article number 2202415) reveal that graphene domains are subject to different compressive strains when growing across the twin boundaries of copper substrates during high-temperature CVD. A new understanding of the effect of twin grain boundaries on graphene epitaxy is provided as a guideline for growing high-quality graphene on twinned metal foil. In another study by Prof. Shi (article number 2202376), a selective-area reconstruction method is proposed for the patterned growth of high-quality graphene, in which selective-area oxidation and high-temperature reduction are capable of regulating the surface characteristics of copper substrates, thereby enabling control over nucleation and epitaxial growth. Profs. Bin Wu and Yunqi Liu et al. (article number 2202584) focus on the growth, characterization, and properties of multilayered graphene. Various state-of-the-art synthetic approaches for multilayered graphene and twisted bilayer graphene are detailed in this review, as well as the underlying growth mechanisms.

Innovation in graphene synthesis techniques that provide new graphene products for specific applications has become the theme of some reviews and research articles. Prof. Liangti Qu et al. (article number 202203164) provide a systematic review of laser-processing technologies for producing graphene with high spatial precision, fast reaction rate, and high controllability. The interaction principle and processing strategies of functional graphene treated by lasers are presented in detail, as well as the regulation of graphene optoelectronic properties for advanced photo/electro-devices. Prof. Qingkai Yu et al. (article number 2202381) propose the synthesis of carbon black and 2D graphene in molten salt at high temperatures by bubbling CVD, and reveal that the formation of transition metal chloride anion complexes in NaCl-KCl can significantly enhance the pyrolysis efficiency of methane. As a new phase, bubbles can be employed as the driving force to cleave the graphite layers to produce graphene nanoplates or separate graphene from the growth substrates. Profs. Qingkai Yu, Guqiao Ding, and Xiaoming Xie (article number 2203124) also introduces bubble-based mass production of graphene in an efficient and controllable manner, covering both “top-down” and “bottom-up” routines.

The functional integration of graphene with traditional materials is expected to uncover new applications with outstanding performance that cannot be achieved using individual components. Optical fibers provide the highest-quality optical waveguides for information communication and photon manipulation, and the integration of optical fibers with graphene has enabled significant advances in all-fiber photonics and optoelectronics. Profs. Kaihui Liu and Zhongfan Liu (article number 2202282) systematically introduce various graphene optical fibers and related fabrication methods, and summarize graphene optical fiber devices, including ultrafast fiber lasers, polarizers, modulators, detectors, and sensors. Profs. Jingyu Sun and Zhongfan Liu (article number 2200428) report the direct preparation of ultra-flat graphene on economical quartz substrates at the wafer scale without the formation of folds, multilayer islands, or metallic impurities. The obtained graphene enables atomically smooth growth of GaN films with (001) single-crystallinity over amorphous silica substrates, facilitating the cost-effective integration of emerging III-nitrides for exciting device applications.

A wealth of engineering or modifying strategies from the perspective of morphological and electronic structures have been applied for multifunctional applications, such as compact energy storage, ion/molecular separation, gas barrier films, and flexible electronics. Prof. Hua Zhang (article number 2202319) summarize strategies for hybridizing 2D nanomaterials with a 3D graphene architecture as well as related applications in electrochemical energy storage and conversion. Prof. Guqiao Ding (article number 2202697) report the strategy to fabricate ultra-large graphene oxide by directly oxidizing the electrochemically derived porous graphene networks, and the graphene networks could enable fast diffusion channels for oxidant. The oxidation process consumes only 40 minutes and ultralow oxidant dosage. Profs. Yanfeng Zhang and Zhongfan Liu (article number 2202026) review the recent advances in direct plasma-enhanced CVD growth of vertically oriented graphene films on functional insulating substrates, with special emphasis on the design of practical synthetic strategies for improving material qualities and developing high-end applications. Prof. Richard Kaner et al. (article number 2203101) report a simple electrochemical approach for the direct deposition of a functional graphene framework using electrostatic interactions between graphene oxide and a cationic surfactant. The reported approach leads to supercapacitors with high specific capacitance and areal capacitance, comparable to those of commercial activated carbon supercapacitors. Prof. Dan Li (article number 2201535) produce a graphene-based membrane by reducing graphene oxide with hydrazine and discover that the resultant densely packed rGO membrane retains the interconnected network nanochannels and shows good capacitive performance. Profs. Andreeva and Novoselov (article number 2201904) propose a novel 2D hydrogel material consisting of the self-assembly of abundant and biodegradable thermo-responsive hydroxypropyl cellulose hydrogel and 2D rGO nanolayers. The composite was developed to produce thermal stimuli and construct an electro-thermo-controlled valve for regulating water transfer. A review by Profs. Li Lin and Zhongfan Liu (article number 2203179) provides a detailed summary of graphene doping strategies for modulating the electronic properties of graphene and a perspective on the applications of doped graphene in optoelectronics.

Some reviews have advanced characterization techniques for understanding the structure–property relationship as their main topic. Prof. Lei Fu and Prof. Mengqi Zeng (article number 2202469) introduce how synchrotron radiation techniques can monitor the structural evolution of graphene at different interfaces, such as layer number, interlayer spacing, and stacking order. Prof. Robert Young and Dr. Zheling Li (article number 2202373) monitor the evolution of stress concentration ahead of crack tips in single-crystal graphene flakes based on Raman spectroscopy and, in particular, Raman mapping. Profs. Wanlin Guo and Zhuhua Zhang (article number 2203866) report colossal mechanocaloric effects in superelastic graphene architectures, and the estimated adiabatic temperature change can reach a record value of 155 K under a compressive stress of 0.7 GPa in a wide temperature range.

Finding new “killer” applications by virtue of irreplaceable functionalities and performance is the driving force of graphene industrialization in the future. The design of graphene materials with high electrical conductivity and high permeability, such as graphene films, fabrics, and composites, for applications in electromagnetic wave shielding and absorption is discussed in the aforementioned review by Chao Gao et al. (article number 2204591). Prof. Quan-Hong Yang et al. (article number 2204272) presents an overview of graphene assemblies, membranes, and powders for advanced batteries, and summarize the applications of graphene in compact energy storage, thermal management, and conductive networks for practical energy devices. Prof. Qiang Zhang et al. (article number 2204755) summarize the recent progress in emerging graphene derivatives and analogs for electrocatalysis, and the material requirements for efficient electrocatalysis and graphene advantages are detailed in this review. Prof. Novoselov, Dr. Zheling Li, and Dr. Boyang Mao (article number 2205934) provide a summary of the progress on the graphene-based smart textiles with special emphasis on thermal management and flame retardancy, covering mechanics, material developments, fabric designs, and on-body applications. Profs. Wencai Ren and Huiming Cheng (article number 2203115) systematically review recent advances and significant developments in CVD-grown graphene films for flexible optoelectronics, including photodetectors, organic solar cells, and light-emitting diodes. The ability to fabricate large-area graphene devices on flexible substrates, work-function tuning, and interfacial control of CVD graphene for high-performance devices are discussed in the aforementioned review. Profs. Il Jeon and Shigeo Maruyama (article number 2204594) provides a summary of the applications of graphene and carbon nanotubes in perovskite solar cells. Versatile roles and the synthetic method of the nanocarbons in perovskite solar cells are provided in the review, covering the material growth methods, and functions as transparent electrodes, charge-transporting layers, interfacial layers, additives, and encapsulants. The application of suspended graphene membranes in multidimensional electron microscopy (EM) imaging is highlighted in a review by Prof. Hailin Peng et al. (article number 2202502). Various approaches to producing suspended graphene membranes and their applications in EM characterization, including high-resolution 2D imaging, cryo-EM 3D reconstruction, and 4D in situ liquid EM, are summarized in this review. Profs. Jin Zhang, Zhong Zhang, and Shuzhou Li (article number 2200937), develop a novel holey rGO/poly(p-phenylene-benzimidazole-terephthalamide) (PBIA) composite fiber with a scaffolded structure. Holey rGO functions as a clamp to effectively band plentiful PBIA chains through in-plane holes, which is capable of enhancing the mechanical performance of graphene-based fibers. Prof. Milo Shaffer (article number 2202596) reveals that polyethylene glycol-grafted graphene, which can be stabilized in water, can provide dispensable nucleant systems for protein crystallization. Prof. Yingying Zhang et al. (article number 2200162) propose a hydrophilic, breathable, biocompatible, and washable graphene-decorated electronic textile that is realized with the assistance of silk sericin; such a graphene-based textile enables the fabrication of comfortable and integrated multi-sensing textiles.

Finally, we would like to express our sincere thanks to the editors of *Advanced Functional Materials*, in particular Dr. Muxian Shen, for initiating this special issue and handling all the papers. We would also like to thank all authors and reviewers for their great efforts in making this special issue possible. Any comment, suggestion, and feedback are always welcome at jin-zhang@pku.edu.cn, hlpeng@pku.edu.cn, Hua.Zhang@cityu.edu.hk, maruyama@photon.t.u-tokyo.ac.jp, and linli-cnc@pku.edu.cn.

Conflict of Interest

The authors declare no conflict of interest.

Jin Zhang obtained his B.S. and M.S. degrees at Lanzhou University in 1992 and 1995, respectively, and completed his Ph.D. degree with Prof. Hulin Li and Prof. Zhongfan Liu at Lanzhou University and Peking University in 1997. After a two-year postdoctoral fellowship at the University of Leeds, UK, he returned to Peking University where he was appointed Associate Professor (2000) and promoted to Full Professor in 2006. In 2013, he was appointed as Changjiang professor. His research focuses on the controlled synthesis and spectroscopic characterization of carbon nanomaterials.

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Shigeo Maruyama, FRSC, received his Ph.D. from the University of Tokyo in 1988. Since 2014, he has been a distinguished professor at the Department of Mechanical Engineering, the University of Tokyo. His research interests include nanoscale thermal engineering, molecular dynamics simulation, nanocarbon materials, and solar cell applications. He developed the alcohol catalytic chemical vapor deposition process for the synthesis of single-walled carbon nanotubes. He served as the president of “The Fullerenes, Nanotubes and Graphene Research Society” during 2011–2020 and is the co-chair of the steering committee of “International Conference on Science and Application of Nanotubes and Low-Dimensional Materials”.

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