2D graphene oxide liquid crystal for real-world applications: Energy, environment, and antimicrobial

Cite as: APL Mater. 8, 070903 (2020); https://doi.org/10.1063/5.0012465
Submitted: 02 May 2020 . Accepted: 26 June 2020 . Published Online: 15 July 2020

Taeyeong Yun, Geong Hwa Jeong, Suchithra Padmajan Sasikala, and Sang Ouk Kim

COLLECTIONS

This paper was selected as an Editor’s Pick

ARTICLES YOU MAY BE INTERESTED IN

Understanding the interplay of stability and efficiency in A-site engineered lead halide perovskites
APL Materials 8, 070901 (2020); https://doi.org/10.1063/5.0011851

Scattering mechanisms and mobility enhancement in epitaxial BaSnO3 thin films probed via electrolyte gating
APL Materials 8, 071113 (2020); https://doi.org/10.1063/5.0017227

Atomic-resolution analytical scanning transmission electron microscopy of topological insulators with a layered tetradymite structure
APL Materials 8, 070902 (2020); https://doi.org/10.1063/5.0014113
2D graphene oxide liquid crystal for real-world applications: Energy, environment, and antimicrobial

I. INTRODUCTION

Discovery of graphene has triggered enormous research interest to its extraordinary properties, including mechanical strength, electric and thermal conductivity, high transparency, and large surface area. Given the advance of human civilization dominated by novel material discovery, including the Paleolithic, Neolithic, Bronze, and Iron Ages, the final destination of emerging material should aim at the practical utilization for real-world impact. Motivated from the sufficiently accumulated scientific knowledge from previous academic efforts, graphene is now at the stage of transformation from academia to industry. As the first step, mass production of high quality graphene is highly demanded to bridge this novel material to industrial product.

Among many different synthetic routes to graphene, three typical methodologies have been frequently exploited for a large-scale production, including (1) chemical vapor deposition (CVD), (2) liquid-phase exfoliation, and (3) chemical modification based on graphene oxide (GO). The CVD process may produce large-area graphene with high electrical conductivity and transparency. Unfortunately, an inherent bottleneck stems from the requirement for an expensive high temperature vacuum process and, more significantly, contamination and damage of the synthesized graphene upon subsequent substrate transfer. In particular, the latter is the decisive issue originating from the intrinsic strong interfacial adhesion between the synthesized graphene layer and the bottom metal catalyst. Liquid-phase exfoliation may yield graphene nano-flakes with intact crystalline nature by means of the readily scalable solution phase mechanical process. However, this approach generally suffers from the insufficient exfoliation (typically yields tens of layers stacked graphite-like structure) and low solvent dispersibility without surfactant additives.
From the early days of graphene research, GO has been a representative chemical derivative of graphene, readily obtainable from inexpensive natural graphite via a low cost solution process. However, significant damage of the graphitic plane and corresponding material properties upon the harsh oxidation process has been the major concern for the high-quality graphene production in this genuine mass producible process. In 2009, our research group discovered graphene oxide liquid crystal (GOLC) and unveiled the potential of monolayer dominant chemically modified graphene, highly dispersible in many common solvents. Significantly, the unprecedented monolayer exfoliation of GO in the highly purified form opened up the cost-effective mass production of few-layer stacked high quality graphene platelets with a clean surface, which is essential for the reliability in material properties. In addition, easy controllability of graphene layer alignment under liquid crystalline (LC) ordering enables the novel material processing, such as 1D fibers, 2D films, and 3D porous structures.

In this perspective article, we propose GOLC as a critical precursor for the large-scale production of high quality graphene, potentially useful for a broad range of technological areas. Taking into account the inexpensive raw material and easily scalable solution process, the distinct features of GO offer a representative framework for the practical balance between material performance and economic cost, especially regarding energy and environmental applications. Facing the recent encounter with the COVID19 virus issue, promising potential of GO for antimicrobial application is also highlighted.

II. CRITICAL IMPURITY EFFECT

GO has been conventionally synthesized from Hummer’s method that requires harsh oxidation circumstance. During the oxidation step, a strong acid, especially sulfuric acid, and an oxidant agent, KMnO\textsubscript{4}, were commonly utilized for a high yield. Accordingly, acid and metal ion impurities are inevitably included in the as-synthesized GO solution. Now, it is well-recognized that these impurities have a tremendous effect on the physical and chemical properties of GO even after reduction.\textsuperscript{7,8} The purity of GO is a pivotal factor in terms of the two principal aspects as follows: (1) intersheet interaction among GO platelets and (2) surface property of the mono-atomic GO nanosheet.

Our group first shed light on the insight for the purification process of the as-synthesized GO solution (Scheme 1). The effective removal of impurities enhances the innate repulsive electrostatic interaction among adjacent GO nanosheets, giving rise to the aqueous GO dispersions with high concentration as well as longterm colloidal stability [Fig. 1(a)]. More importantly, we discovered a nematic type liquid crystallinity of the highly purified GO solution, which is attributed to the high shape anisotropy of the monolayer exfoliated GO sheets and high colloidal stability up to sufficient concentration [Fig. 1(b)]. Since our first discovery of GOLC, many research studies have investigated its fundamental characteristics, opening up the new realm of the 2D-based LC system.\textsuperscript{11–14}

The property of GO is strongly related to its surface characteristics due to the genuine monoatomic nature. In this regard, the properties of GO and its reduced form are significantly influenced by the inevitable impurity.\textsuperscript{15} For instance, the electrochemical activity of GO can be significantly affected by the metal impurity, which can bring about uncontrollable undesired side reactions and doping effects, as shown in Fig. 1(c).\textsuperscript{16,17} Given that, the clean surface of highly purified GO is a stringent requirement to ensure the reliable properties of GO particularly aiming at energy and environmental applications, where molecular absorption/desorption and surface reaction/charge transfer are crucial, in general.

Biosafety and antibacterial characteristics of GO have been still under debate principally due to the deficiency of the reliable GO property. For instance, the flake size effect of GO on the antimicrobial property is totally inconsistent.\textsuperscript{18,19} This challenge for the reliable experimental result associated with GO cytotoxicity should be largely due to the different surface characteristics of GO sheets, according to the source of graphite, synthesis...
FIG. 1. The critical impurity effect on GO. (a) Formation and stability of GOLC. (i and ii) 0.1 wt. %, 0.2 wt. %, and 0.4 wt. % (left to right) GO dispersion (i) without any dialysis treatment and (ii) after dialysis treatment. Due to the remaining acidic or ionic impurities, the dispersion, shown in (i), showed weak birefringence, and the precipitation of graphene oxide occurred. The sample in (ii), which are free of acidic and ionic impurities, shows typical isotropic to nematic biphasic phase separation. (b) (Left to right) Aqueous GOLC dispersion (0.5 wt. %) exhibiting chocolate milky appearance; phase-separated 0.2 wt. % dispersion three weeks after preparation; three phase-separated dispersions (0.05 wt. %, 0.2 wt. %, 0.5 wt. %) located between crossed polarizers. Reproduced with permission from Kim et al., Angew. Chem., Int. Ed. 50, 3043 (2011). Copyright 2011 Wiley-VCH. (c) Oxygen reduction reaction for various GO samples was tested in air-saturated 0.5M KOH solution. Reproduced with permission from Mazánek et al., ACS Nano 13, 1574 (2019). Copyright 2019 American Chemical Society. (d) TEM images of E. coli ATCC25922 (i and iii) and S. aureus ATCC25923 (ii and iv) after incubation in MHB media (i and iii) and MHB media with GO (UM preparation) at 100 μg ml⁻¹ (ii and iv). Arrow in the TEM image indicates GO flakes.

method, and impurity level. Noticeably, Barnolina et al. reported that the purity of GO determines its antibacterial activity, where highly purified GO reveals no antibacterial property [Fig. 1(d)]. Evidently, a high level of GO purification is the prime requirement to expand the scope of GO application into biology relevant areas.

III. SOLUTION PROCESS OF GO

The solution process is generally suitable for scalable mass production. The basic criterion for the solution process lies in the stable dispersion of the desired solute in the proper solvent system. Unfortunately, pristine graphene has an inert surface consisting of sp² conjugated domains, attributing to the genuine low compatibility with common solvents. Moreover, the strong restacking tendency among pristine graphene sheets results in the inherent limitation for the solution process. By contrast, GO has an interesting amphiphilic character, consisting of the hydrophilic oxidized sp³ carbon regions coexisting with hydrophobic graphitic crystal domains. This distinct nature endows good dispersibility of GO in many solvents, including water [Fig. 2(a)]. In addition, the solvent dispersibility can be readily controlled along with further chemical functionalization, hetero-atom doping, the degree of reduction, and so on.

Solution phase mixing of GO and other functional components offer a straightforward route to composite structures. As such, uniformly dispersed molecular level fillers can remarkably enhance the material properties even with an exceptionally low loading level. Note that despite the highly damaged graphitic structure, GO can still be an interesting filler particularly in energy and environmental applications, while exploiting its surface functionalities and reactivities. Moreover, its LC ordering can greatly enhance the material structure and properties optimized for a desired application area.

Besides, GO may synergistically grant additional functionalities to existing products, such as molecule barrier, light response, flame
FIG. 2. The solution process of GO. (a) Photograph of graphite oxide directly exfoliated in water and organic solvents by 1 h of ultrasonication (top row) and the stability of GO dispersions after 3 weeks (bottom row). Reproduced with permission from Paredes et al., Langmuir 24, 10560 (2008). Copyright 2008 American Chemical Society. (b) Vertical burning test (UL94) of a nanocomposite foam including 77% CNF, 10% GO, 10% SEP, and 3% BA (in wt. %). The panel shows the foam before test, after 11 s of application of a methane flame, and the foam after the test, showing high fire retardancy. Reproduced with permission from Wicklein et al., Nat. Nanotechnol. 10, 277 (2015). Copyright 2014 Springer Nature. (c) Salt spray testing after 2,000 h showing the comparison between the zinc-based anti-corrosion coatings with (bottom) and without (top) graphene oxide additives. Reproduced with permission from Ramezanzadeh et al., Chem. Eng. J. 320, 363 (2017). Copyright 2017 Elsevier. (d) The photograph of commercialized HEAD graphene tennis racquet. (e) SEM image of breath figure assembly for reduced GO (rGO) macroporous films. Reproduced with permission from Lee et al., Angew. Chem., Int. Ed. 49, 10084 (2010). Copyright 2010 Wiley-VCH. (f) SEM image of large flake GO fiber with an overhand knot; scale bar is 100 μm. Reproduced with permission from Xiang et al., Adv. Mater. 25, 4592 (2013). Copyright 2013 Wiley-VCH. (g) Photograph for the GO mesh structure from extrusion-based 3D printing. Reproduced with permission from Lacey et al., Adv. Mater. 30, 1705651 (2018). Copyright 2018 Wiley-VCH.

retardant, and anti-corrosion [Figs. 2(b) and 2(c)].31–34 In reality, the graphene-based composite products have been already commercialized in sports and leisure fields [Fig. 2(d)].3,35 Until now, those products have mainly concentrated on the enhancement of mechanical properties. More advanced graphene-based products embedded with novel functionalities would encounter with us in the near future.

Highly dispersed GO solution can be utilized for distinct architectures by means of the facile solution process, such as (1) spin-, spray-, and dip-coating; (2) solvothermal process; and (3)
self-assembly. These solution processes are fundamentally based on the unique molecular level interaction among GO sheets, including hydrophilicity, electrostatic interaction, hydrogen bonding, and so on. The 2D lamellar assembled structure consists of parallel stacked GO layers with tunable interlayer gallery, anisotropic transport, and mechanical flexibility. 3D nanoporous architectures composed of a cross-linked GO framework exhibit high surface area, facile ion transport, and mechanical reversibility. Our research group has pioneered novel solution-based molecular self-assembly principles to precisely build-up multi-dimensional structures with nanoscale precision [Fig. 2(c)].26–28 High concentrated GO dispersion enables straightforward wet-spinning of graphene-based carbon fibers as well as extrusion-based 3D printing for unprecedented applications [Figs. 2(f) and 2(g)].29–31 Notably, the GO liquid crystal fiber is the emerging next-generation carbon-based fiber with diverse functionalities that can complement the commercially available traditional carbon fibers.3 Overall, inherent scalability of the solution process is highly advantageous for the commercialization for GO-based products with precisely tailored material structures and properties.32–34

IV. APPLICATIONS

As mentioned above, the material properties of GO are generally considered inferior to those of pristine graphene along with the formation of oxygen functional groups as well as structural defects in the basal plane. However, the meaning of “quality” should be dependent on the relevant application field. In this regard, GO has unique characteristics, including (1) surface functionalities and reactivity, (2) high specific surface area with hydrophilic adhesive nature, and (3) easy control of molecular alignment and ordering under liquid crystallinity. These distinct features of GO are highly beneficial in a broad range of applications, such as supercapacitors, membranes, molecular adsorption, and antimicrobials.

A. Supercapacitor

A supercapacitor is an attractive energy storage device for the miniaturized electronics and electric vehicles, owing to simple device architecture and high power delivery capability. Unusually high performance of graphene-based supercapacitors triggered enormous research interest for the energy storage from the early days of graphene research.35 Initial research efforts concentrated on the hierarchical porous structure to effectively manipulate the high surface area and facilitate ion transport. However, the random porous structure with low packing density revealed the limitation of low volumetric capacity, which is apparent figure-of-merit for real-world application.36

The mechanism of ion storage in pristine graphene-based supercapacitors is based on electric double layer capacitance, where facile ion transport plays a crucial role for the overall capacity. Yoon et al. suggested a valuable insight for the vertical aligned GO to attain high performance supercapacitors.37 The alignment of GO could be readily controlled to the vertical direction with the simple rolling and cutting process [Fig. 3(a)].38 The vertically aligned graphene electrode represents the high packing density (1.18 g/cm³) and exceptionally high volumetric capacitance (171 F/cm³ at 1 A/g) under the aqueous electrolyte system. Interestingly, the capacitance could be well-retained even at extreme high current density (123 F/cm³ at 20 A/g), owing to the facile ion transport along the vertical aligned electrode structure.

Another promising perspective for the capacitive energy storage is the control of interlayer spacing in the graphene assembled structure. Note that the low energy density of the supercapacitor can be readily addressed with the organic or ionic liquid electrolyte to expand the operating voltage window. The interlayer gallery within the tightly stacked lamellar structure of graphene could act as the optimized nanopore for those electrolyte ions.39,40 Recently, Li et al. reported that the interlayer of freestanding graphene oxide film could be precisely tuned by incorporating reduced graphene oxide into GO. Their interlayer optimization led to an extraordinary high energy density (88.1 W h l⁻¹), which is comparable with the commercialized lead-acid battery (50 W h l⁻¹–90 W h l⁻¹), as well as anomalously high volumetric capacity (203 F/cm³ at 1 A/g) [Fig. 3(b)].37

Recently, there have been several reports for the modification of the interlayer distance and vertical alignment control of GO-based electrodes.41–44 Moreover, this perspective has been also applied into other 2D materials, such as MXene and transition metal dichalcogenide (TMD), for the similar purpose of high performance energy storage.45

B. Separation membrane

Exciting new opportunities have been evolved at the intersection of the separation membrane and 2D materials research in the last decade. One-atom thick graphene-based membranes are considered as an ideal candidate for molecular sieving, but the practical challenges are involved with the generation of uniform nanoscale holes on the graphene single layer.46 The discovery of GOLC initiated a rapid progress in the tunable 2D material-based membranes for the selective separation of a wide variety of molecular and species. The prospective applications are not only limited to seawater desalination but also include wastewater treatment by the effective removal of toxic heavy metals, radioactive elements, and organic contaminants.47–49 Additionally, GO membranes have potential in the industrial gas separation, buffer exchange, dialysis, and non-aqueous filtration intend for catalyst recovery, solvent recycling, and redox flow batteries.50–52

The multilayer membrane assembled from GOLC may offer interlayer 2D capillary channels for the size selective sieving of chemical species. Interestingly, ions with smaller size than the gallery channels can rapidly flow through the GO membrane compared to simple bulk diffusion.53,54 In contrast to the avoidable wide distribution of the pore size in the typical polymeric membranes, the selectivity and size exclusion of GO membranes propose broad implications in many applications.55 GO membranes with a controlled size and shape of capillary channels could be achieved by sandwiching GO layers with suitable spacer components and functionalities [Fig. 3(c)].56,57 Nevertheless, the swelling of GO in water is a major challenge for water filtration. Notably, several novel methods to retard the swelling of GO membrane have been reported, including (1) physical confinement with introducing spacer components such as epoxy, (2) partial reduction of GO membranes to decrease the hydrated functional groups, and (3) covalent bonding among the stacked GO nanosheets. Besides, the GO membrane with specific
FIG. 3. Energy and environmental application of GO. (a-i) Schematic illustration of facile ion diffusion transport with vertically aligned reduced GO film (VArGO), (ii) SEM image of VArGO film, and (iii) cyclic voltammetry curve of VArGO at a scan rate of 20 V/s. Reproduced with permission from Yoon et al., ACS Nano 8, 4580 (2014). Copyright 2014 American Chemical Society. (b-i) Schematic illustration of GO and EGM-GO film with different interlayer structure, (ii) SEM image of rGO and EGM-rGO film, and (iii) Ragone plots of the EGM-rGO film-based supercapacitor with state-of-the-art energy storage devices. Reproduced with permission from Li et al., Nat. Energy 5, 160 (2020). Copyright 2020 Springer Nature. (c-i) Schematic and (ii) SEM image of the GO membrane with epoxy encapsulate for selective sieving of ions while allowing water flux along graphene planes (scale bar 1 μm) and (iii) rate of ion permeation through GO membranes with increasing interlayer distances. Reproduced with permission from Abraham et al., Nat. Nanotechnol. 12, 546 (2017). Copyright 2017 Springer Nature. (d-i) Schematic illustration of heavy metal adsorption of GO by electrostatic interaction, and (ii) Effect of initial Pb(ii) ions concentration on adsorption of the LS-GO-PANI ternary nanocomposite at 30 °C. Reproduced with permission from Peng et al., J. Mol. Liq. 230, 496 (2017). Copyright 2017 Elsevier. Reproduced with permission from Yang et al., ACS Sustainable Chem. Eng. 2, 1203 (2014). Copyright 2014 American Chemical Society. (e-i) Schematic diagram of the GO and virus interaction exposed at different temperature. The physiochemical interaction between viruses and reactive oxygenated groups were indicated by dotted lines. The release of viral RNA is described in red, and (ii) TEM image of negative stained GO captured H9N2 complexes with thermal heating treatment. Reproduced with permission from Song et al., Small 11, 1171 (2015). Copyright 2015 Wiley-VCH.
chemical functionalities can be utilized for the precise sieving of biomolecules.\textsuperscript{77,78} Indeed, GO membranes represent the next-generation membrane with high-flux, cost-effective separation of ions and molecules.\textsuperscript{20} However, their exact mechanism for the high flux and exclusive selectivity should be further understood for the formidable reliable long-term usability in practical separation applications.

C. Molecular adsorption

GO can effectively absorb heavy metals, oils, and organic compounds, such as dye. The adsorption capacity of GO is superior to graphene or other low-dimensional materials, owing to its hydrophilic surface functionalities, high surface area, and controllability of gallery distance among GO layers.\textsuperscript{21} Notably, for the heavy metal adsorption, oxygen surface groups of GO endow negative charge to electrostatically attract the metal cations. This capacity is mainly attributed to carboxyl (−COOH) groups, among several oxygen functional groups, such as epoxy (C−O−C), hydroxyl (C−OH), and carboxyl (COOH) groups. Unfortunately, the amounts of epoxide and carbonyl (C=O) groups are known to increase with the oxidizing agent, while GO is prepared from graphite.\textsuperscript{22,23} Therefore, how to increase the density of carboxyl groups at the GO surface is the major concern for the heavy metal adsorption.\textsuperscript{24} Another advantage of GO for molecular adsorption is the high dispersibility in hydrophilic solvents and easy surface modification with other functional groups. Yang et al. prepared GO functionalized with lignosulfonate (LS) and polyaniline (PANI) for the adsorption of Pb ions [Fig. 3(d)].\textsuperscript{25} The amino groups in PANI may improve the coordinate capability of sulfonic groups on LS chains and carboxyl groups on GO nanosheets for Pb ions.

More recently, the principal research trend focuses on the effects of environmental factors, such as temperature, pH, and heavy metal concentration on the adsorption capacity of GO. Regrettfully, studies on the structural properties of GO and competitive/selective adsorption in the presence of multiple heavy metals are insufficient.\textsuperscript{26} Most heavy metals such as Pb, Hg, Cd, Cr, and As are rare metals. Although global uses of the rare metals are gradually increasing, the recycle of them has been insignificantly considered. Structural assembly of GO can facilitate the hierarchical structures with unique pore size, interlayer distance, and desirable functional groups on GO. This is highly demanded to recycle those rare metals through competitive and selective adsorption/desorption of heavy metals from the distinct GO-based capture system.

D. Antimicrobial property

In contrast to the generic hydrophobic characteristics of carbon materials, including carbon nanotubes, fullerene, and activated carbon, GO has inborn hydrophilic nature stemming from the surface oxygen functional groups. In this regard, GO is intrinsically biocompatible nanomaterial and even more along with further surface modification with bio-specific functionalities. Taken together with its hydrophilic nature stemming from the surface oxygen functional group and physicochemical interaction. Indeed, GO membranes represent the next-generation membrane with high-flux, cost-effective separation of ions and molecules.\textsuperscript{20} However, their exact mechanism for the high flux and exclusive selectivity should be further understood for the formidable reliable long-term usability in practical separation applications.

V. CONCLUSION AND OUTLOOK

We have highlighted the current progress of GO research for the real-world graphene application, particularly focusing on the energy, environmental, and antimicrobial area. We emphasized the significance of GO purification via “GOLC route” for the desirable high quality graphene commercialization. Purified GO enables a well-dispersible colloidal state, endowing a unique characteristic of 2D LC. In fact, such an ionic impurity effect, including pH and ionic strength, on the colloidal stability and liquid crystallinity is a well-established academic subject in the traditional colloid science. Note that this original concept of 2D LC has been growing and influencing other 2D materials, such as 2D TMDs and MXene. Regrettting the old yet relatively ignored history for the first discovery of 2D LC based on the colloidal clay platelet, recent emergence of 2D LC is particularly noticeable in terms of the practical material design for 3D fibers, 2D films, and 3D porous architectures. This is the typical example of an emerging scientific area led by practical requirements.

The wonder material, graphene, has passed through tremendous technological hype and now is transforming into the realistic form for practical utilization. Unfortunately, the best quality large-area CVD graphene is still suffering from the difficulty in the damage-free separation from the metal catalyst. This can be overcome in the typical lab-scale scientific research but hardly addressable in the large-scale mass production. Nowadays, metal-free direct synthesis at a target substrate structure is the major direction in human enzyme, which diminishes the health risk associated with the exposure of GO-based biocompatible nanomaterial.\textsuperscript{30,34} However, further research on the biocompatibility of GO in vitro and in vivo should be pursued to ensure the controversial safety issue of GO.

Very recently, in 2020, the COVID 19 issue is threatening mankind over worldwide. One of the effective solutions to address this issue should be to intrinsically screen the virus from the human skin. The biocompatible GO with a high surface area could be an ideal framework for the adsorption of those harmful species. Notably, GO has an innate antimicrobial property, attributed to the oxygen functional group and physicochemical interaction.\textsuperscript{31} Song et al. reported the development of the GO-based label-free methodology to rapidly detect and disinfect environmental viruses, such as enteric EV71 and H9N2 [Fig. 3(e)].\textsuperscript{32} The hydroxyl, epoxy, and carboxyl groups of GO facilitate the effective interaction with the virus surface mediated with hydrogen bonding, electrostatic interaction, and even redox reaction. More importantly, the papers about dealing with the COVID 19 virus by exploiting graphene have already been reported with reusable mask and rapid detection for COVID 19.\textsuperscript{33,34}

Easy solution processibility of GO is also another critical benefit for the practical antimicrobial applications. GO can be directly employed along with the versatile structure formation into fibers, films, fabrics, and so on, particularly taking advantage of GOLC-based facile solution assembly. Besides, GO can be readily functionalized with antimicrobial metals, such as Cu and Ag.\textsuperscript{35} Nonetheless, the use of GO in the field of biological research including antimicrobial activity is still in its infancy, and many relevant research efforts are anticipated for the sustainability of human life.
the relevant industry. Liquid-phase exfoliation is highly active in the current industry over worldwide owing to the lowest cost production of high crystalline structures. However, the large stacking number (typically >10) is the critical parameter for any desired properties and applications along with the inborn mineral impurity effect even from source graphite. Moreover, without proper chemical modification, pristine graphene platelets reveal the intrinsic strong tendency for restacking in solvent dispersion and even dried states.

GOLC-based mass production of graphene platelets is emerging as a promising pivotal route to the commercialization of the high quality, few-layer-stack 2D structure. Insufficient recovery of the graphitic structure via the oxidation/reduction route could be further improved for better mechanical and electrical properties. Nonetheless, highly aligned graphene layers in 1D fibers or 2D films, enabled by GOLC, can largely compensate the unavoidable damage of 2D building blocks. In addition, the least impurity effect from the purified GO route endows a reliable electrochemical activity while avoiding undesirable side reaction, which is highly demanded for high performance batteries and supercapacitors. Molecular adsorption, the critical step for catalysis and separation, can also be strengthened by the minimal contamination effect. Taking advantage of the well-balanced material performance with respect to practical economic burden, we anticipate that high quality graphene via the GOLC route should contribute to the recent global issues in energy, environment, and pandemic, severely threatening human life.

ACKNOWLEDGMENTS

This work was financially supported by the Nanomaterial Technology Development Program Planning (Grant No. NRF-2016M3A7B4905613) through the National Research Foundation of Korea (NRF) funded by the Ministry of Science, ICT and Future and KIST-KAIST Joint Research Lab (Grant No. 2V05750).

DATA AVAILABILITY

The data that support the findings of this study are available within the article.

REFERENCES

1. K. S. Novoselov, A. K. Geim, S. V. Morozov, D. Jiang, Y. Zhang, S. V. Dubonos, I. V. Grigorieva, and A. A. Firsov, Science 306, 666 (2004).
2. A. K. Geim and K. S. Novoselov, Nat. Mater. 6, 183 (2007).
3. W. Hong, K. Hum, S.-H. Bae, J. Shim, H. Kim, L. Kong, Y. Meng, K. Wang, C. Kim, and J. Kim, Nat. Nanotechnol. 14, 927 (2019).
4. S. Bae, H. Kim, Y. Lee, X. Xu, J.-S. Park, Y. Zheng, J. Balakrishnan, T. Lei, H. Ri Kim, Y. I. Song, Y.-J. Kim, K. S. Kim, R. Osyilmaz, J.-H. Ahn, B. H. Hong, and S. Iijima, Nat. Nanotechnol. 5, 574 (2010).
5. K. R. Paton, E. Varrla, C. Backes, R. J. Smith, U. Khan, A. O'Neill, C. Boland, M. Lotya, O. M. Istrate, P. King, T. Higgs, S. Barwick, P. May, P. Puzczarski, I. Ahmed, M. Moebius, H. Pettersson, E. Long, J. Coelho, S. E. O'Brien, E. K. McGuire, R. M. Sanchez, G. S. Duesberg, N. McEvoy, T. J. Pennycook, C. Downing, A. Crossley, V. Nicolosi, and J. N. Coleman, Nat. Mater. 13, 624 (2014).
6. S. Stankovich, D. A. Dikin, R. D. Piner, K. A. Kohlhaas, A. Kleinhammes, Y. Jia, Y. Wu, S. T. Nguyen, and R. S. Ruoff, Carbon 45, 1558 (2007).
7. C. H. A. Wong, Z. Sofer, M. Kubelskova, J. Kučera, S. Matějková, and M. Pumera, Proc. Natl. Acad. Sci. U. S. A. 111, 13774 (2014).
8. F. M. Hu, T. Ma, H.-Q. Lin, and J. E. Gubernatis, Phys. Rev. B 84, 075414 (2011).
9. A. V. Krasheninnikov and R. M. Nieminen, Theor. Chem. Acc. 129, 625 (2011).
10. E. Kim, T. H. Han, S. H. Lee, J. Y. Kim, C. W. Ahn, J. M. Yun, and S. O. Kim, Angew. Chem., Int. Ed. 50, 3043 (2011).
11. K. E. Lee, J. E. Kim, U. N. Matti, J. Lim, J. O. Hwang, J. Shim, J. J. Oh, T. Yun, and S. O. Kim, ACS Nano 8, 9073 (2014).
12. R. Narayan, J. E. Kim, J. Y. Kim, K. E. Lee, and S. O. Kim, Adv. Mater. 28, 3045 (2016).
13. Z. Xu and C. Gao, Nat. Commun. 2, 571 (2011).
14. B. Senyuk, N. Behabtu, A. Martinez, T. Lee, D. E. Tsentalovich, G. Ceriotti, J. M. Tour, M. Pasquali, and I. I. Smalyukh, Nat. Commun. 6, 7157 (2015).
15. A. Ambrosi, S. Y. Chee, B. Khezri, R. D. Webster, Z. Sofer, and M. Pumera, Angew. Chem., Int. Ed. 51, 500 (2012).
16. M. Lotya, O. M. Istrate, P. King, T. Higgins, S. Barwich, P. May, P. Puczkarski, S. Iijima, Nat. Nanotechnol.
17. C. Kim, and J. Kim, Nat. Nanotechnol.
18. E. K. McGuire, B. M. Sanchez, G. S. Duesberg, N. McEvoy, T. J. Pennycook, I. Ahmed, M. Moebius, H. Pettersson, E. Long, J. Coelho, S. E. O'Brien, M. Lotya, O. M. Istrate, P. King, T. Higgins, S. Barwich, P. May, P. Puczkarski, S. Iijima, Nat. Nanotechnol.
19. A. K. Geim and K. S. Novoselov, Nat. Mater. 6, 183 (2007).
20. S. Stankovich, D. A. Dikin, G. H. B. Dommett, K. M. Kohlhaas, E. J. Zimney, E. A. Stacy, R. D. Piner, S. T. Nguyen, and R. S. Ruoff, Nano Lett. 9, 1593 (2009).
21. U. N. Matti, W. J. Lee, J. M. Lee, Y. Oh, J. Y. Kim, E. Kim, J. Shim, T. H. Han, and S. O. Kim, Adv. Mater. 26, 40 (2014).
22. W. Du, H. Wu, H. Chen, G. Xu, and C. Li, Carbon 158, 568 (2020).
23. D. G. Papageorgiou, I. A. Kinloch, and R. J. Young, Prog. Mater. Sci. 90, 75 (2017).
24. S. Stankovich, D. A. Dikin, G. H. B. Dommett, K. M. Kohlhaas, E. J. Zimney, E. A. Stacy, R. D. Piner, S. T. Nguyen, and R. S. Ruoff, Nature 442, 282 (2006).
25. R. J. Young and M. Liu, J. Mater. Sci. 51, 3861 (2016).
26. S. H. Lee, H. W. Kim, J. O. Hwang, W. J. Lee, J. Kwon, C. W. Bielawski, R. S. Ruoff, and S. O. Kim, Angew. Chem., Int. Ed. 49, 10084 (2010).
27. S. H. Lee, D. H. Lee, W. J. Lee, and S. O. Kim, Adv. Funct. Mater. 21, 1338 (2011).
28. J. Shim, J. M. Yun, T. Yun, P. Kim, K. E. Lee, W. J. Lee, R. Ryoo, D. J. Pine, G.-R. Yi, and S. O. Kim, Nano Lett. 14, 1388 (2014).
29. O. Hwang, J. S. Park, D. S. Choi, J. Y. Kim, S. H. Lee, K. E. Lee, Y.-H. Kim, M. H. Song, S. Yoo, and S. O. Kim, ACS Nano 6, 159 (2012).
30. U. N. Matti, J. Lim, K. E. Lee, W. J. Lee, and S. O. Kim, Adv. Mater. 26, 615 (2014).
31. S. Padmajan Sasikala, K. E. Lee, J. Lim, H. J. Lee, S. H. Koo, I. H. Kim, H. J. Jung, and S. O. Kim, ACS Nano 11, 9424 (2017).
42 G. H. Jeong, S. P. Sasikala, T. Yun, G. Y. Lee, W. J. Lee, and S. O. Kim, Adv. Mater. “Nanoscale assembly of 2D materials for energy and environmental applications,” (published online, 2020).

43 C. Xiang, C. C. Young, X. Wang, Z. Yan, C.-C. Hwang, G. Cerioni, J. Lin, J. Kono, M. Pasquali, and J. M. Tour, Adv. Mater. 25, 4592 (2013).

44 C. Xin, T. Yao, H. Sun, S. M. Scott, D. Shao, G. Wang, and J. Lian, Science 349, 1083 (2015).

45 S. D. Lacey, D. J. Kirsch, Y. Li, J. T. Morgenstern, B. C. Zarket, Y. Yao, J. Dai, L. Q. Garcia, B. Liu, T. Gao, S. Xu, S. R. Raghavan, J. W. Connell, Y. Lin, and L. Hu, Adv. Mater. 30, 1705651 (2018).

46 Y. Jiang, Z. Xu, T. Huang, Y. Liu, F. Guo, X. Ji, W. Gao, and C. Gao, Adv. Funct. Mater. 28, 1707202 (2018).

47 B. Fang, D. Chang, Z. Xu, and C. Gao, Adv. Mater. 32, 1902664 (2020).

48 S. Pei, Q. Wei, K. Huang, H.-M. Cheng, and W. Ren, Nat. Commun. 9, 145 (2018).

49 Z. Xu, Y. Liu, X. Zhao, L. Peng, H. Sun, Y. Xu, X. Ren, C. Jin, P. Xu, M. Wang, and C. Gao, Adv. Mater. 28, 6449 (2016).

50 Z. Zhong, W. Sun, Q. Wei, X. Qian, H.-M. Cheng, and W. Ren, Nat. Commun. 9, 3348 (2018).

51 Y. Zhu, S. Murali, M. D. Stoller, K. J. Ganesh, W. Cai, P. J. Ferreira, A. Pirkle, R. M. Wallace, K. A. Cychoz, M. Thommes, D. Su, E. A. Stach, and R. S. Ruoff, Science 332, 1537 (2011).

52 Y. Gogotsi and P. Simon, Science 334, 917 (2011).

53 Y. Yoon, K. Lee, S. Kwon, S. Seo, H. Yoo, S. Kim, Y. Shin, Y. Park, D. Kim, J.-Y. Choi, and H. Lee, ACS Nano 8, 4580 (2014).

54 Y. Qian, X. Zhang, C. Liu, C. Zhou, and A. Huang, Desalination 460, 56 (2019).

55 K. H. Thebo, X. Qian, Q. Zhang, L. Chen, H.-M. Cheng, and W. Ren, Nat. Commun. 9, 1486 (2018).

56 Z. Li, S. Gaddipelli, H. Li, C. A. Howard, D. J. L. Brett, P. R. Shearing, Z. Guo, I. P. Parpin, and F. Li, Nat. Energy 5, 160 (2020).

57 W. Dai, T. Ma, Q. Yan, J. Gao, X. Tan, L. Lv, H. Hou, Q. Wei, J. Yu, J. Wu, Y. Yao, S. Du, R. Sun, N. Jiang, Y. Wang, J. Kong, C. Wong, S. Maruyama, and C.-T. Lin, ACS Nano 13, 11561 (2019).

58 Y. Xia, T. S. Mathis, M.-Q. Zhao, B. Anasori, A. Dang, Z. Zhou, H. Cho, Y. Gogotsi, and S. Yang, Nature 557, 409 (2018).

59 M. Aerce, D. Voiry, and M. Chhowalla, Nat. Nanotechnol. 10, 313 (2015).

60 S. P. Surwade, S. N. Smirnov, I. V. Vlassiouk, R. R. Unocic, G. M. Veith, S. Dai, and S. M. Mahurin, Nat. Nanotechnol. 10, 459 (2015).

61 P. Perreault, A. Fonseca de Faria, and M. Elimelech, Chem. Soc. Rev. 44, 5861 (2015).

62 H. K Mahalingam, S. Wang, and S. P. Nunes, ACS Appl. Nano Mater. 1, 4661 (2018).

63 S. Dervin, D. D. Dionysiou, and S. C. Pillai, Nanoscale 8, 15115 (2016).

64 I. Prozorovska and P. R. Kidambi, Adv. Mater. 30, 1801179 (2018).

65 B. Qi, X. He, G. Zeng, Y. Pan, G. Li, G. Liu, Y. Zhang, W. Chen, and Y. Sun, Nat. Commun. 8, 825 (2017).

66 P. Sun, K. Wang, and H. Zhu, Adv. Mater. 28, 2287 (2016).

67 R. R. Nair, H. A. Wu, P. N. Jayaram, I. V. Grigorieva, and A. K. Geim, Science 335, 442 (2012).

68 R. K. Joshi, P. Carbone, F. C. Wang, V. G. Kravets, Y. Su, I. V. Grigorieva, H. A. Wu, A. K. Geim, and R. R. Nair, Science 343, 752 (2014).

69 B. Mi, Science 343, 740 (2014).

70 J. Abraham, K. S. Vasu, C. D. Williams, K. Gopinadhan, Y. Su, C. T. Cherian, J. Dix, E. Prestat, S. J. Haigh, I. V. Grigorieva, P. Carbone, A. K. Geim, and R. R. Nair, Nat. Nanotechnol. 12, 546 (2017).

71 M. Hu and B. Mi, Environ. Sci. Technol. 47, 3715 (2013).

72 Q. Zhang, X. Qian, K. H. Thebo, H.-M. Cheng, and W. Ren, Sci. Bull. 63, 788 (2018).

73 C. Sathe, X. Zou, J.-P. Leburton, and K. Schulten, ACS Nano 5, 8842 (2011).

74 D. Li, W. Zhang, X. Yu, Z. Wang, Z. Su, and G. Wei, Nanoscale 8, 19491 (2016).

75 W. Peng, H. Li, Y. Liu, and S. Song, J. Mol. Liq. 230, 496 (2017).

76 Y. Liu, R. Ma, X. Wang, Y. Ma, Y. Yang, L. Zhuang, S. Zhang, R. Jehan, J. Chen, and X. Wang, Environ. Pollut. 252, 62 (2019).

77 K. Krishnamoorthy, M. Veerapandian, K. Yun, and S. J. Kim, Carbon 53, 38 (2013).

78 Y. Tang, Y. Liu, W. Guo, T. Chen, H. Wang, S. Yu, and F. Gao, J. Phys. Chem. C 118, 24866 (2014).

79 J. Chen, Y. Zhang, M. Zhang, B. Yao, Y. Li, L. Huang, C. Li, and G. Shi, Chem. Sci. 7, 1874 (2016).

80 J. Yang, J.-X. Wu, Q.-F. Liu, and T.-T. Lin, ACS Sustainable Chem. Eng. 2, 1203 (2014).

81 Y. Ge and Z. Li, ACS Sustainable Chem. Eng. 6, 7181 (2018).

82 S. Syama and P. V. Mohanan, Nano-Micro Lett. 11, 6 (2019).

83 R. Kurapati, J. Russier, M. A. Squillaci, E. Treossi, C. Ménard-Moyon, A. E. Del Rio-Castillo, E. Vazquez, P. Samori, V. Palermo, and A. Bianco, Nat. Nano 11, 3985 (2015).

84 S. P. Mukherjee, A. R. Gilga, B. Lazzaretto, B. Brandner, M. Fielden, C. Vogt, L. Newman, A. F. Rodrigues, W. Shao, P. M. Fournier, M. S. Toprak, A. Star, K. Kostarelos, K. Bhattacharya, and B. Fadeel, Nanoscale 10, 11880 (2018).

85 S. Ye, K. Shao, Z. Li, N. Guo, Y. Zuo, Q. Li, Z. Lu, L. Chen, Q. He, and H. Han, ACS Appl. Mater. Interfaces 7, 21571 (2015).

86 Y.-N. Chen, Y.-H. Hsieh, C.-T. Hsieh, D.-Y. Tsou, and P.-L. Chang, Int. J. Environ. Res. Public Health 13, 430 (2016).

87 X. X. Yang, C. M. Li, Y. F. Li, J. Wang, and C. Z. Huang, Nanoscale 9, 16086 (2017).

88 P. Kumar, P. Huo, R. Zhang, and B. Liu, Nanomaterials 9, 737 (2019).

89 I. Sengupta, P. Bhattacharya, M. C. D. Castile, E. Vazquez, P. Samori, V. Palermo, and A. Bianco, Small 11, 3985 (2015).

90 Y. Gu, Z. Guo, X. Liu, Y. Zhang, Y. Sun, Y. Duan, J. Zhan, and J. Lu, Environ. Pollut. 168, 460 (2013).

91 S. Nair, H. A. Wu, P. N. Jayaram, I. V. Grigorieva, and A. K. Geim, Science 335, 442 (2012).