Control of water for high-yield and low-cost sustainable electrochemical synthesis of uniform monolayer graphene oxide

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With the rapid development of graphene industry, low-cost sustainable synthesis of monolayer graphene oxide (GO) has become more and more important for many applications such as water desalination, thermal management, energy storage and functional composites. Currently, chemical oxidation, represented by the Hummers' method, is still the main method for the synthesis of GO, but suffers from severe environmental pollutions, explosive risks, and high cost. With the increasingly stringent environmental and safety laws, the production of GO by chemical oxidation is increasingly restricted[1]. Compared to the conventional chemical oxidation methods, water electrolytic oxidation of sulfuric acid graphite-intercalation-compound (SA-GIC) shows significant advantages in environmental-friendliness, safety and efficiency[2], but suffers from non-uniform oxidation, which has become the main obstacle that hinders the industrial electrochemical (EC) synthesis of GO. Here, we found that the deintercalation of SA-GIC caused by water absorption from environment and aqueous electrolyte is responsible for the un-uniform oxidation of graphite. Moreover, both the EC oxidation and deintercalation are significantly affected by the diffusion of water from the electrolyte to the interior of SA-GIC. The competition between these two processes determines whether SA-GIC could be uniformly oxidized. Based on these understandings, we developed a liquid membrane electrolysis method to precisely control the water diffusion to achieve a dynamic equilibrium between EC oxidation and deintercalation[3], which enables industrial sustainable synthesis of uniform GO. The yield of GO can reach ~180 wt.%, with >99% being monolayers, and the cost is only ~1/7 of that of the Hummers' method. Additionally, this technique offers exceptional tunability in GO properties. These findings have uncovered the key puzzle that has long plagued the development of EC synthesis of GO. The liquid membrane electrolysis technique paves the way for the sustainable mass production of GO at a low cost, which will greatly advance its industrial applications in various fields.

References

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