# A Defect-free Principle for Advanced Graphene Cathode of Al-ion battery

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**Abstract:** Aluminum-ion battery (AlB) is emerging as a promising energy storage system due to its high power density, high anode capacity, low flammability and stable cycling. However, the main challenge associated with AlB is the low cathodic capacity, which limits its energy density. Here, for the first time, we report a defect-free few-layer graphene aerogel cathode with excellent performance and reproducibility. This defect-free graphene aerogel cathode affords a high capacity of 100 mAh $g^{-1}$ with average discharge voltage of 1.95 V at 50 C, capacity retention of 97% after 25000 cycles and high-rate capability of 97 mAh $g^{-1}$ at 500 C (7.2 seconds charging time), making the best-performing carbon cathode for AlB so far. This prominent cathode material is designed by a general defect-free principle, that is the fewer defects, the better performance, to get rid of the inactive defect components and maximize the graphene active material. This most basic defect-free principle opens an avenue for AlB to achieve high power density and high energy density with flexibility and high safety, and also guides us to fabricate better batteries based on graphene electrodes.

## Introduction

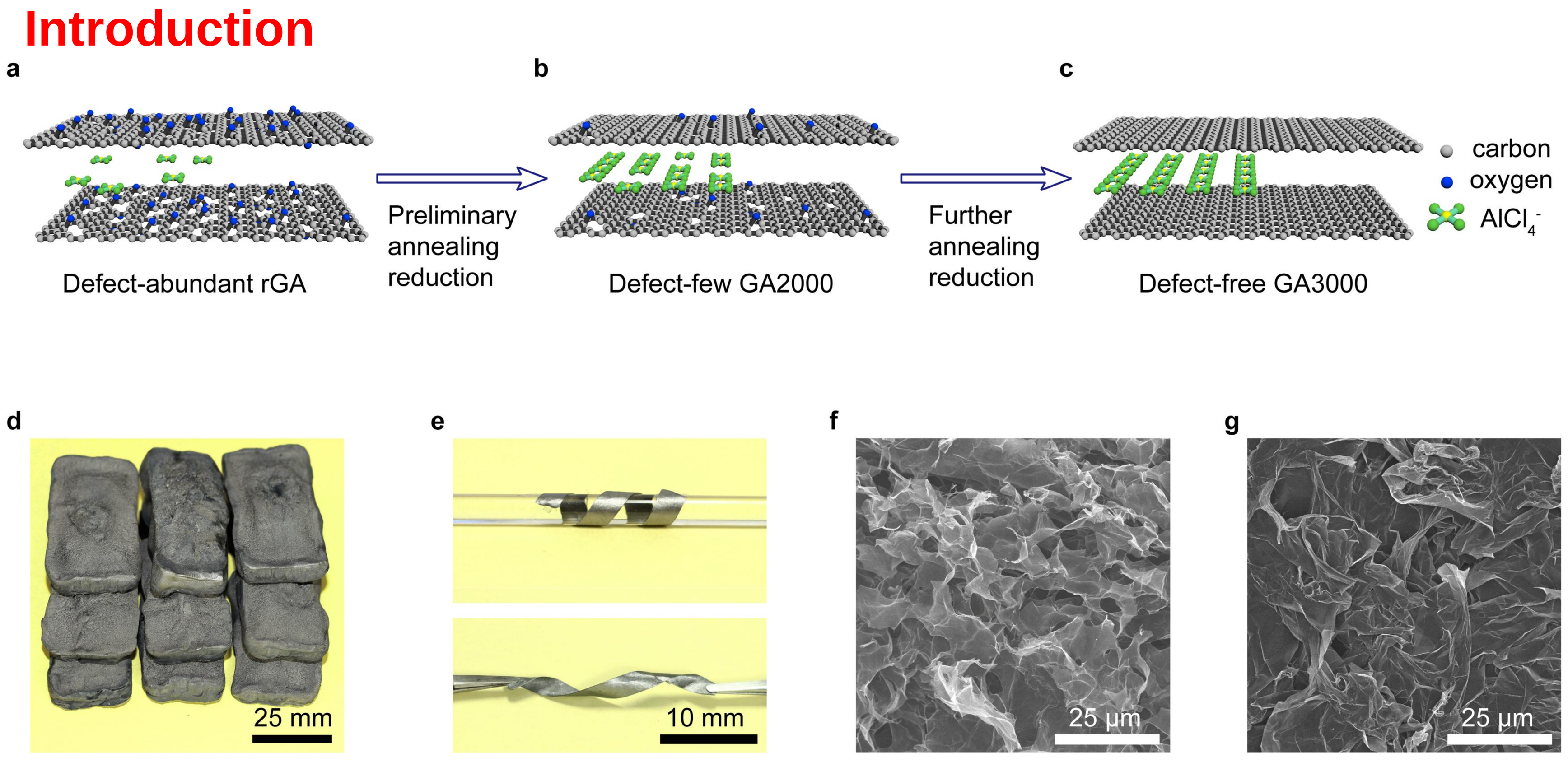


**Fig. 1.** Design and production of GA cathode. a-c) Schematic of the defect-free design. Defects ($sp^3$ carbons and oxygen-containing groups) in rGA (a) and GA-2000 (b) reduce the electrical conductivity, impede the transportation of $AlCl_4^-$ and cannot act as active sites; the defect-free $sp^2$ carbon networks in GA-3000 (c) act as active sites and facilitate the transportation of $AlCl_4^-$. d) Digital camera image of GA-3000. e) A compressed GA-3000 strip is coiled around a glass rod to illustrate the good flexibility and twisted by two tweezers to show the twistability. f, g) SEM images of GA-3000 (f) and compressed GA-3000 paper (g).

## Key advantage of defect-free design:

(1) Elimination of inactive defects facilitating the fast intercalation of large-sized anions and simultaneusly providing more active sites for energy storage.

(2) Greatly enhanced electrical conductivity recovering defective graphene into electrical highway.

(3) High reproducibility of material quality and cell performance favoring large-scale manufacture.

**From a wetland with abundant traps (defect-abundant rGA, Fig.1a) to a bumpy road with many traps (defect-few GA-2000, Fig. 1b, 2a) , and eventually to a trap-free smooth highway (defect-free GA-3000, Fig. 1c, 2d).**

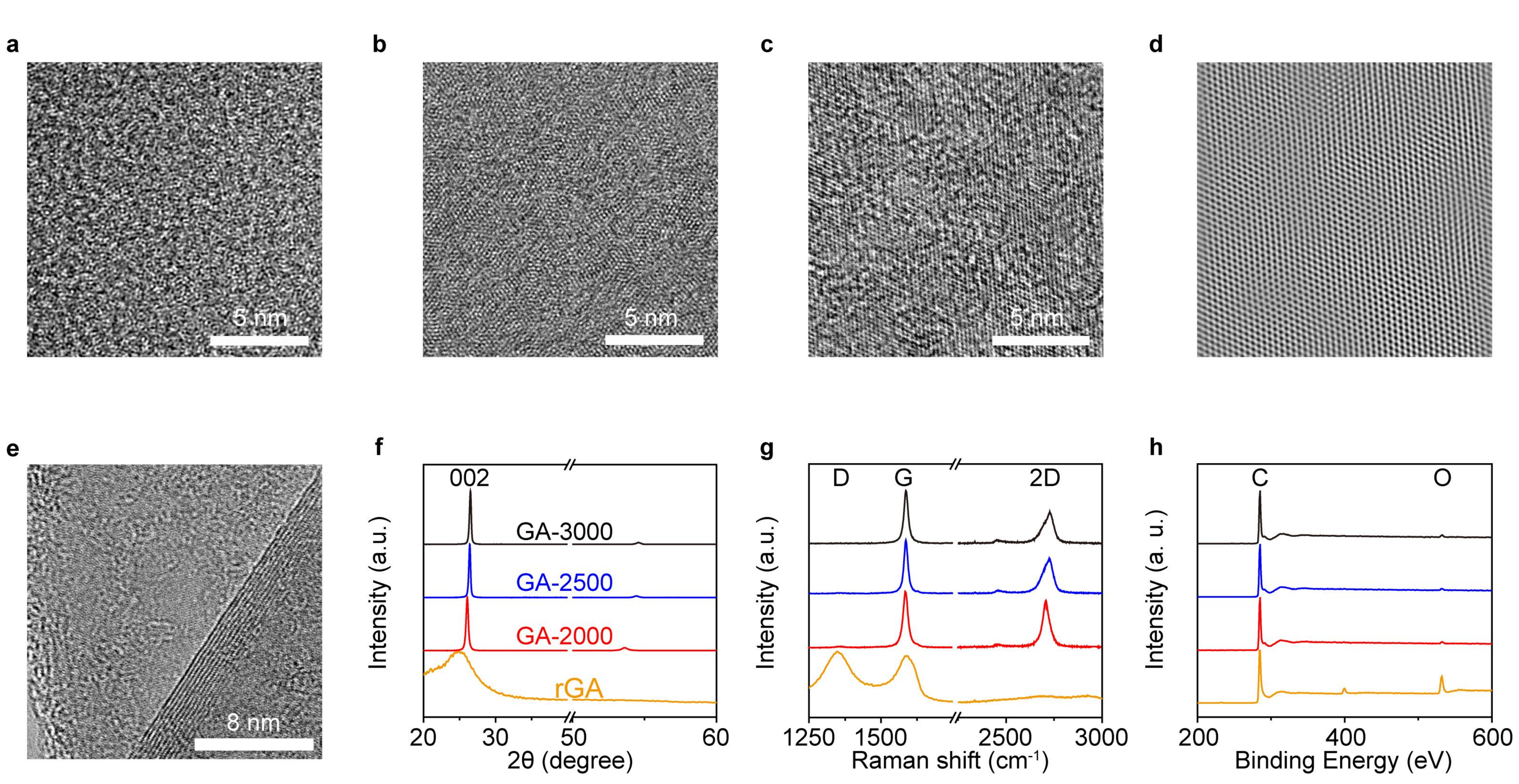


**Fig.2** Structural characterizations of GAs. a-e) HRTEM images of GA-2000 (a), GA-2500 (b), GA-3000 (c) and IFFT of GA-3000 (d) showing the enhanced crystallinity of graphene with higher temperature annealing and stacked 4-10 graphene layers (e). f) X-ray diffraction (XRD) pattern of different Gas. g-h) Raman (g) and XPS (h) spectra illustrating the evolution of different GAs.

## Significant electrochemical performance:

**(1) 97% capacity retention after 25,000 cycles.**

**(2) Fast charge in 7.2 seconds without capacity loss.**

**(3) Excellent reproducibility within 100 samples.**

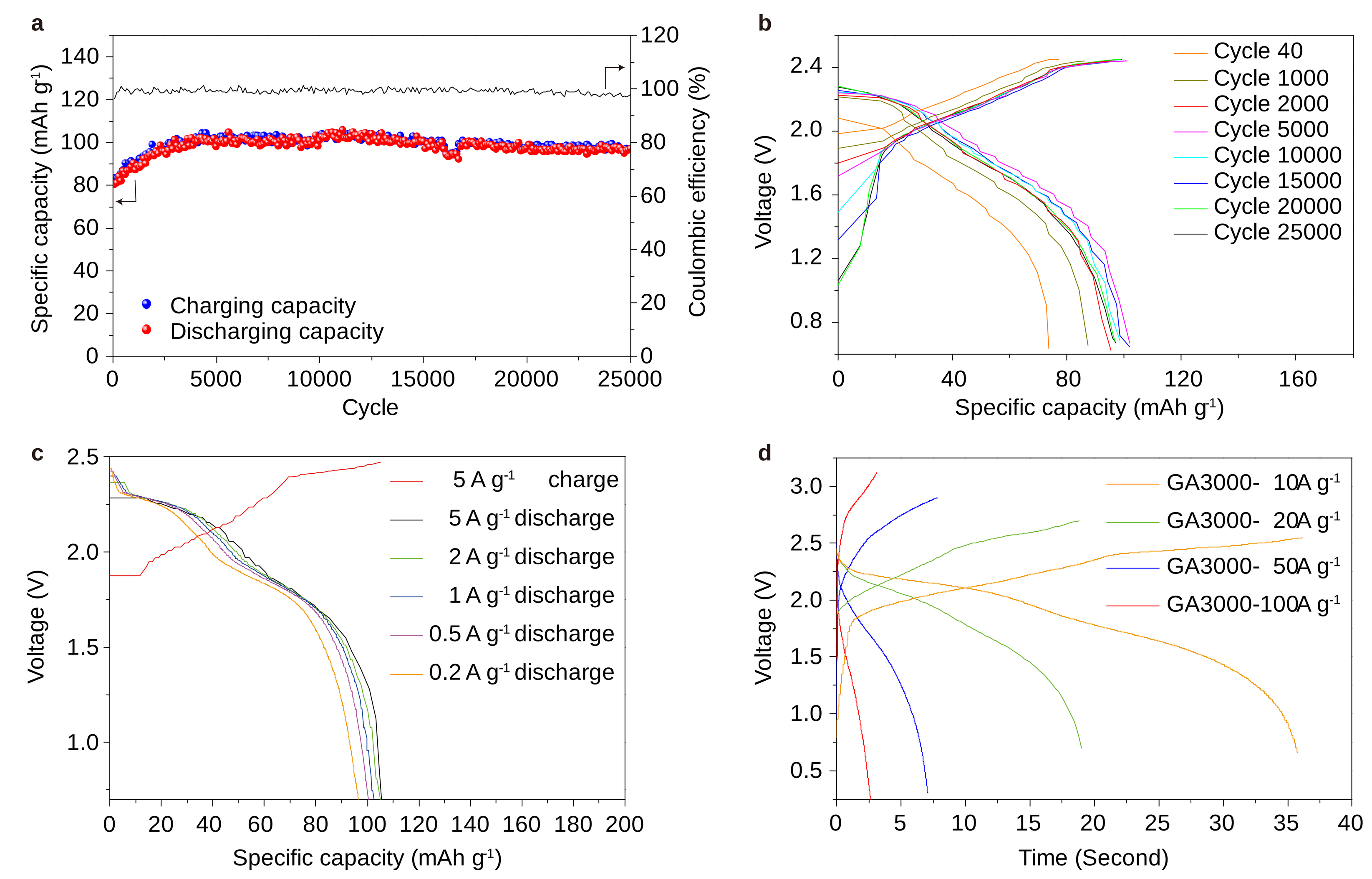


**Fig. 3.** Electrochemical performances of GA cathodes. a) Galvanostatic cycling of GA-3000 (5 A $g^{-1}$) over 25000 cycles. b) Detailed charge and discharge curves of GA-3000 cathode at different cycles (50 C). c) Charge and discharge profiles of GA-3000 cathode charged at 50 C and discharged at lower current densities (from 2 C to 50 C). d) Charge and discharge curves of GA-3000 at higher current densities (from 100 C to 1000 C)

## Useful Aluminum-graphene battery:

**(1) Low-cost aluminum foil as anode and graphene as cathode.**

**(2) High discharge voltage for power supply.**

**(3) Fast charge in 36 seconds and light LED for 15 minutes.**

**(4) Incomparable non-flammability even when placed in flame.**

**(5) Meaningful flexibility battery for future wearable electronic devices.**


**Fig. 4.** Exhibition of Al-GBs. a-c) Aluminum anode (a) and graphene cathode (b) of Al-GB while powering a LED and its sustainability (c). d) Al-GB placed in the flame of alcohol lamp while powering a LED. e, f) Coiled (e) and folded (f) Al-GB pouch cell while powering a LED light.

## Conclusions

We have proposed and demonstrated a general defect-free principle for designing advanced graphene cathode of AlB with a highly controllable and scalable methodology. The defect-free GA cathode affords a high specific capacity of 100 mAh $g^{-1}$ from 2 C to 50 C (average discharge voltage around 1.95 V) with unprecedent high-rate performance (97 mAh $g^{-1}$, 1.55 V at 500 C and 74 mAh $g^{-1}$ at 1000 C) and excellent cycle stability (97% capacity retention after 25000 cycles), making it the best performing cathode for AlB so far. Benefiting from the defect-free principle (i.e., the fewer defects, the better performances), the Al-GB can afford an energy density of ~60 Wh $kg^{-1}$ comparable to commercial lead-acid batteries, a power density of ~30 kW $kg^{-1}$ comparable to the best graphene-based supercapacitor with ultralong cycling life and high safety. Such a system can be called as aluminium-graphene superbattery, and we believe that there is still plenty of room for improving the performance. The firstly established defect-free principle would offer new sights into graphene-based electrodes of rechargable batteries.

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## References

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