Plasma-etched functionalized graphene as a metal-free electrode catalyst in solid acid fuel cells

Xubin Lu, [‡ab]
Xin Yang,[‡c] Muhammad Tariq, [d] Fan Li,[e] Matthias Steimecke,[b] Jia Li, [*c] Aron Varga,[af] Michael Bron[*b] and Bernd Abel [*a]
[a]Leibniz Institute of Surface Modification, Permoserstraße 15, 04318 Leipzig, Germany.
[b]Martin-Luther-Universität Halle-Wittenberg, Technische Chemie I, Von Danckelmann-Platz 4, D-06120 Halle (Saale), Germany.
[c]Laboratory for Computational Materials Engineering, Division of Energy and Environment, Graduate School at Shenzhen, Tsinghua University, Shenzhen 518055, P. R. China.
[d]Martin-Luther-Universität Halle-Wittenberg, Institut für Physik, FG Experimentelle Polymerphysik, Von-Danckelmann-Platz 3, D-06120 Halle (Saale), Germany.
[e]Max-Planck-Institut für Microstrukturphysik, Weinberg 2, D-06120 Halle (Saale), Germany.
[f]BMW Group, Taunusstraße 41, 80807 Munich.
E-mail: xubin.lu@chemie.uni-halle.de

Abstract

Graphene has demonstrated considerable potential to replace costly precious metal catalysts in a broad range of technological applications[1, 2]. As such, extensive research is being conducted on investigating the substitution capability of graphene, particularly with respect to the fuel cell industry[3, 4]. In this work, graphene was subjected to O_2 or N_2 plasma treatment to create catalytically active sites through the introduction of functional surface groups and defect site generation. Density functional theory (DFT) calculations were used to evaluate the effect of plasma treatment on the activity of the graphene sheets in the oxygen reduction reaction (ORR) coupled with experimental results. ORR activities were adequately enhanced by plasma treatment. DFT calculations further identified that zigzag carbon atoms could be the most active site for the ORR in O_2 and N_2 plasma-etched graphene. In addition, the armchair carbon atom adjacent to the heteroatom is another primary active site for N_2 plasma-etched graphene. The C=O functional groups in conjunction with the armchair carbon also showed high ORR activity. The O_2 and N_2 plasma-treated graphene was successfully used as an electrode catalyst in a solid acid fuel cell (SAFC). These results provide important basic information about the value of using plasma-etched graphene in SAFCs and will aid future efforts in the development of catalytically active and stable non-precious metal materials for use in fuel cell cathodes.

REFERENCES

[1] Z. Liu, Z. Zhao, Y. Wang, S. Dou, D. Yan, D. Liu, Z. Xia, S. Wang, In Situ Exfoliated, Edge-Rich, Oxygen-Functionalized Graphene from Carbon Fibers for Oxygen Electrocatalysis, Adv. Mater., 29 (2017) 1606207.
[2] L. Xue, Y. Li, X. Liu, Q. Liu, J. Shang, H. Duan, L. Dai, J. Shui, Zigzag carbon as efficient and stable oxygen reduction electrocatalyst for proton exchange membrane fuel cells, Nat. Commun., 9 (2018) 3819.
[3] J. Deng, P.J. Ren, D.H. Deng, L. Yu, F. Yang, X.H. Bao, Highly active and durable non-precious-metal catalysts encapsulated in carbon nanotubes for hydrogen evolution reaction, Energy Environ. Sci., 7 (2014) 1919-1923.
[4] X. Lu, X. Yang, M. Tariq, F. Li, M. Steimecke, J. Li, A. Varga, M. Bron, B. Abel, Plasma-etched functionalized graphene as a metal-free electrode catalyst in solid acid fuel cells, J. Mater. Chem. A, 8 (2020) 2445-2452.

FIGURES

Figure 1: Simple schematic drawing of electrochemical cell (Graphene|CsH_2PO_4|Pt/C/CsH_2PO_4)