Supporting Information

Tailoring active sites *via* synergy between graphitic and pyridinic N for the enhanced catalytic efficiency of carbocatalyst

Jieyuan Li[†], Shi Yin[†], Fan Dong[‡], Wanglai Cen[§*], Yinghao Chu[†*]

†. College of Architecture and Environment, Sichuan University, P. R. China

‡. Chongqing Key Laboratory of Catalysis and New Environmental Materials, College of Environment and Resources, Chongqing Technology and Business University, P. R. China

§. Institute of New Energy and Low Carbon Technology, Sichuan University, P. R. China

*Corresponding authors

Prof. Dr. Wanglai Cen (cenwanglai@163.com)

Prof. Dr. Yinghao Chu (chuyinghao@scu.edu.cn)

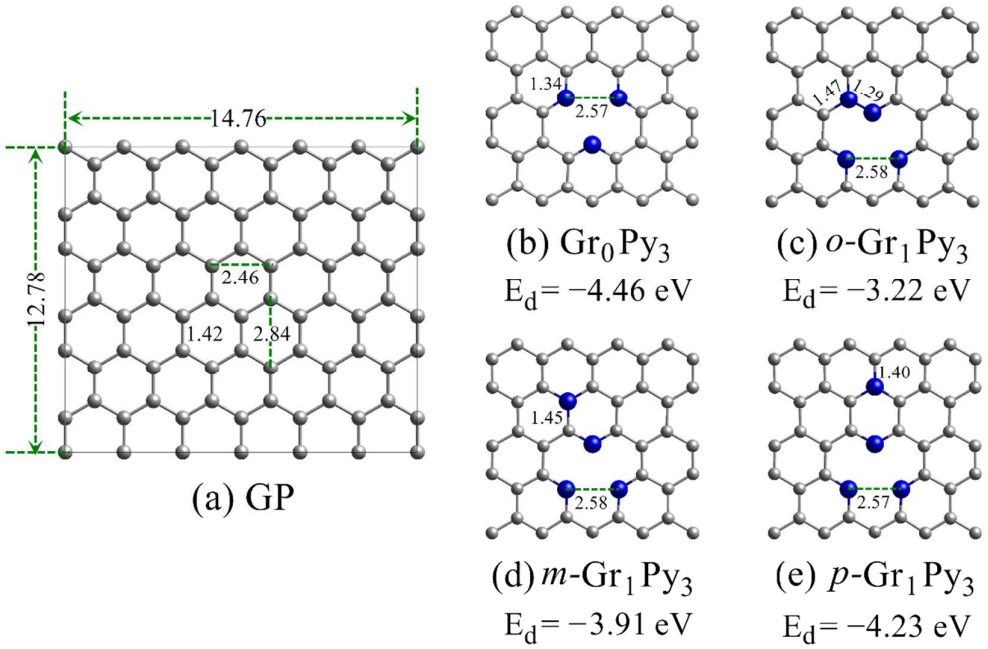

Figure S1. Possible local structure and doping energy (E_d) of different N doped GP. Blue (grey) spheres depict N (C) atoms. (a) Pristine GP; (b) Gr_0Py_3; (c) *o*-Gr_1Py_3; (d) *m*-Gr_1Py_3; (e) *p*-Gr_1Py_3. Negative means heat release.

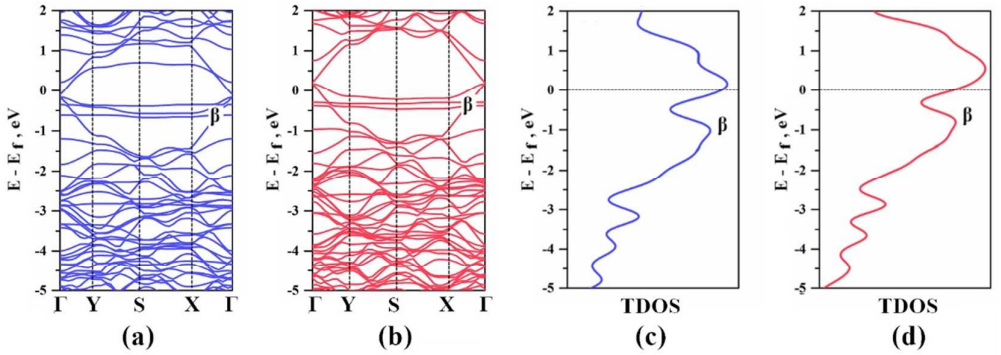

Figure S2. Calculated band structures (a, b) and Total density of states (c, d) of Gr_0Py_3 and Gr_1Py_3, respectively. The Fermi level is set to 0 eV.

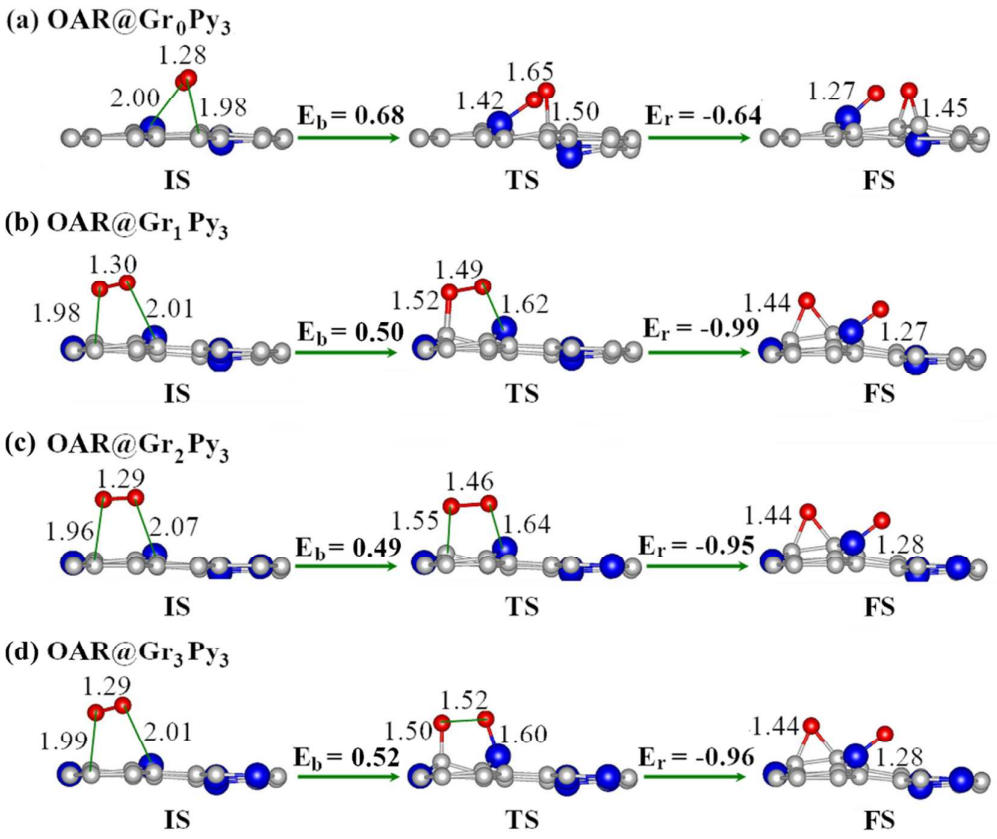

Figure S3. Minimum energy pathway (MEP) of O_2 activation reactions (OAR) by Gr_0Py_3 (a), Gr_1Py_3 (b), Gr_2Py_3 (c) and Gr_3Py_3 (d). E_b and E_r stand for the energy barrier and reaction energy respectively. Red, blue and grey spheres depict O, N and C atoms. All lengths are given in Å.

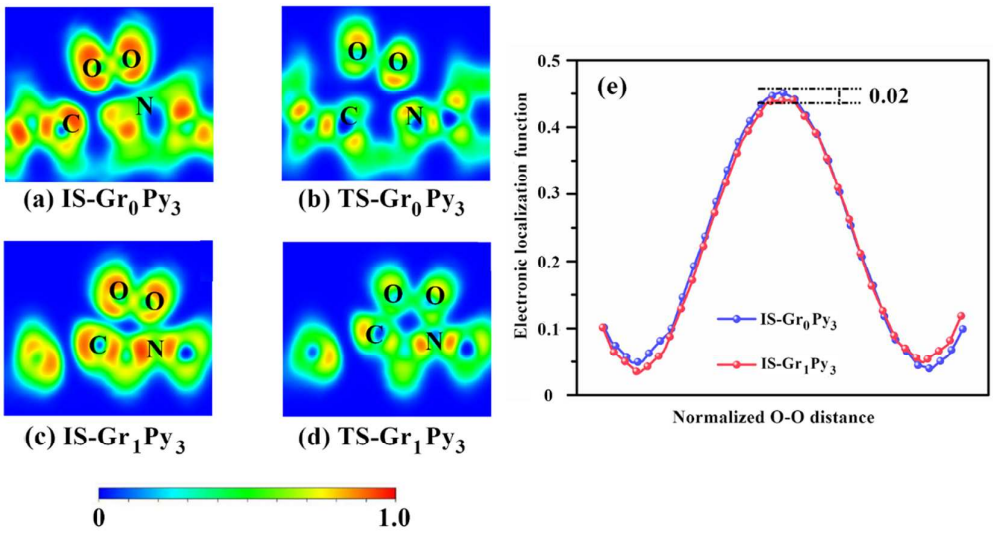

Figure S4. The electronic localization function (ELF) for O_2 molecules in the initial state (IS) and transition (TS) of the OAR in Gr_0Py_3 (a, b) and Gr_1Py_3 (c, d). The line profile of ELF for IS of the OAR in Gr_0Py_3 and Gr_1Py_3 (e).

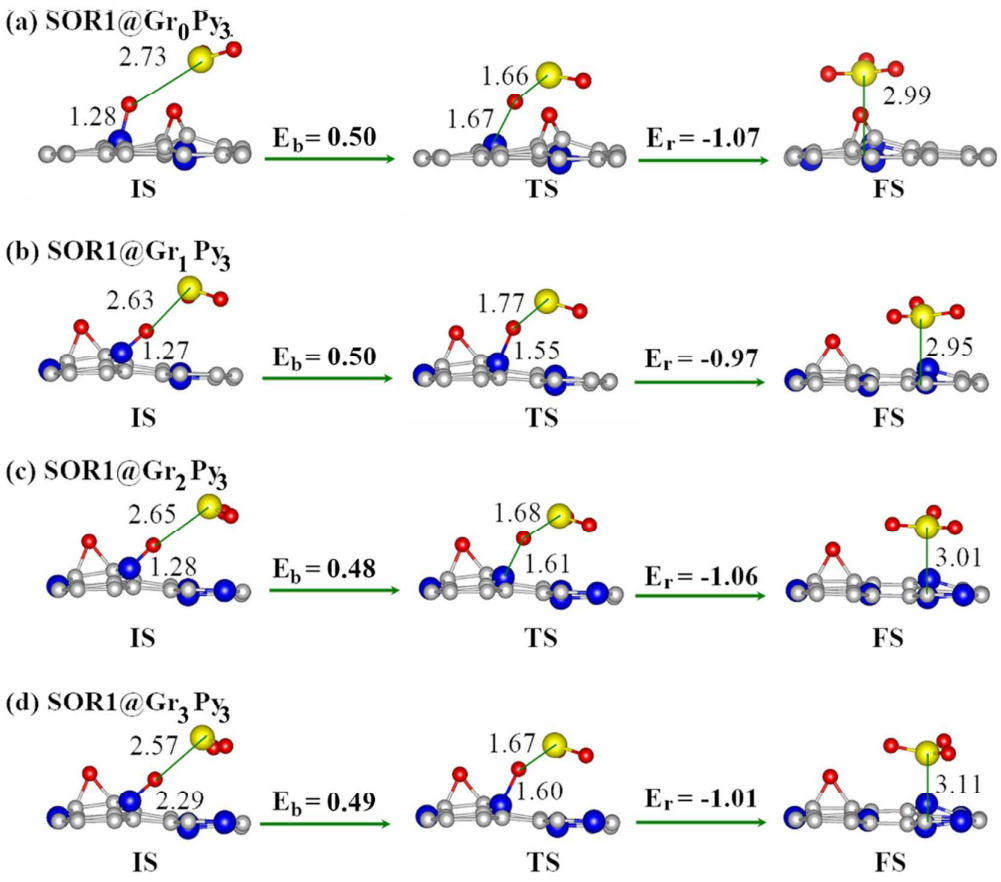

Figure S5. Minimum energy pathway (MEP) of SO_2 oxidation reactions 1 (SOR1) by Gr_0Py_3 (a), Gr_1Py_3 (b), Gr_2Py_3 (c) and Gr_3Py_3 (d). E_b and E_r stand for the energy barrier and reaction energy respectively. Red, blue and grey spheres depict O, N and C atoms. All lengths are given in Å.

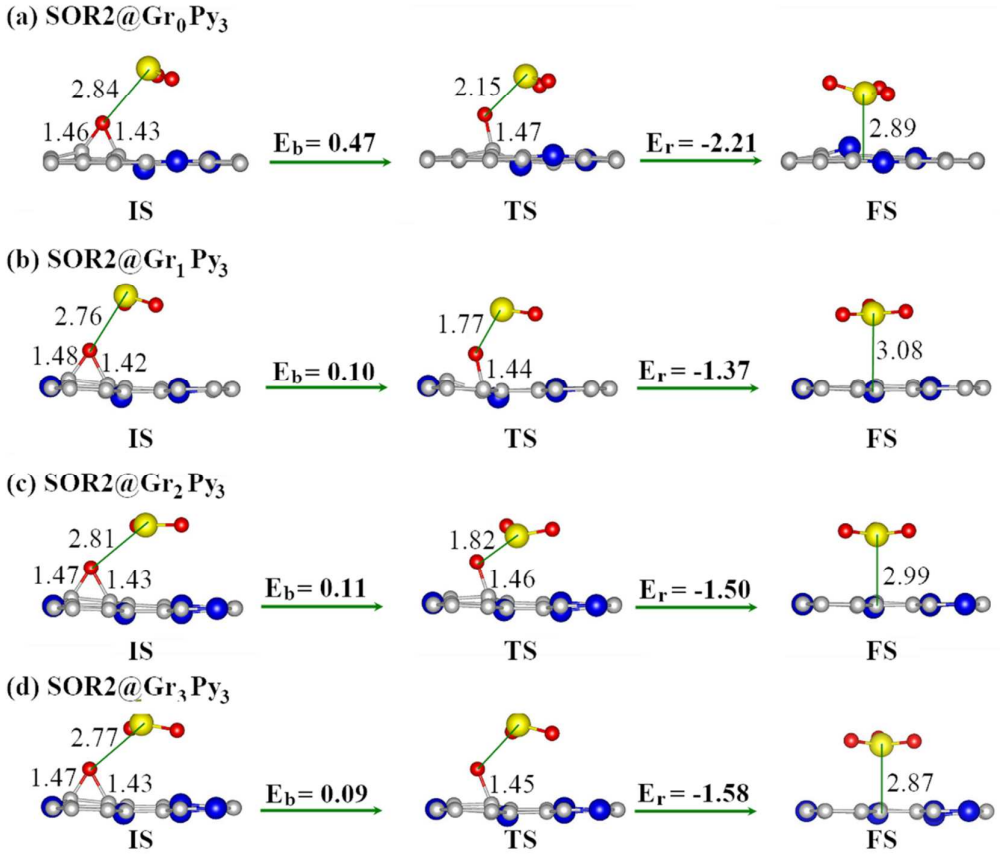

Figure S6. Minimum energy pathway (MEP) of SO_2 oxidation reactions 2 (SOR2) by Gr_0Py_3 (a), Gr_1Py_3 (b), Gr_2Py_3 (c) and Gr_3Py_3 (d). E_b and E_r stand for the energy barrier and reaction energy respectively. Red, blue and grey spheres depict O, N and C atoms. All lengths are given in Å.