

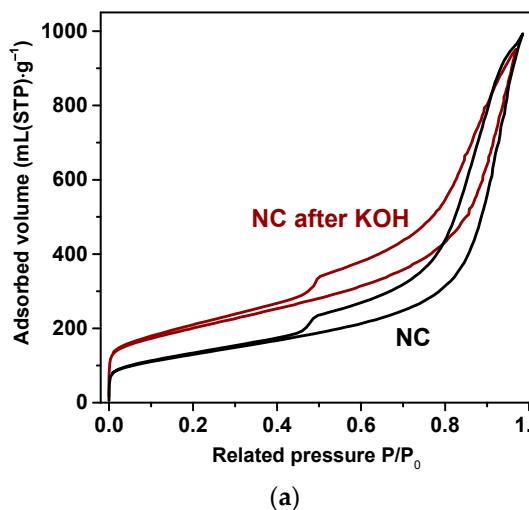
Electrochemical performance of potassium hydroxide and ammonia activated porous nitrogen-doped carbon in sodium-ion batteries and supercapacitors

Yuliya V. Fedoseeva ^{1,*}, Elena V. Shlyakhova ¹, Svetlana G. Stolyarova ¹, Anna A. Vorfolomeeva ¹, Alina D. Nishchakova ¹, Mariya A. Grebenkina ¹, Anna A. Makarova ², Konstantin A. Kovalenko¹, Alexander V. Okotrub ¹, Lyubov G. Bulusheva ^{1,*}

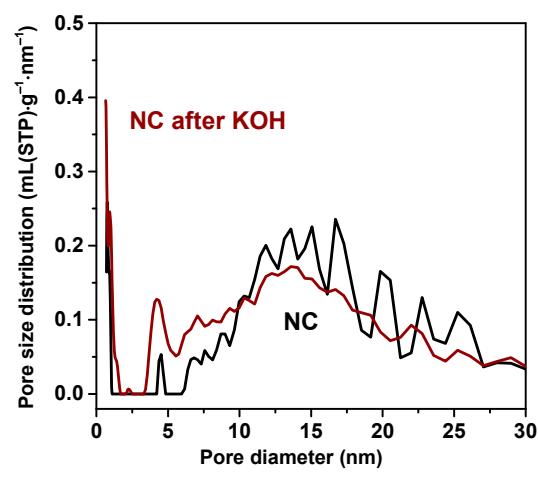
¹ Nikolaev Institute of Inorganic Chemistry SB RAS, 3 Acad. Lavrentiev Ave., 630090 Novosibirsk, Russia

² Physical Chemistry, Institute of Chemistry and Biochemistry, Free University of Berlin, 14195 Berlin, Germany

* Correspondence: fedoseeva@niic.nsc.ru (Y.V.F), bul@niic.nsc.ru (L.G.B.); Tel.: +73833305352 (Y.V.F.&L.G.B.)



(a)



(b)

Figure S1. (a) Nitrogen adsorption-desorption isotherms at 77 K and (b) DFT pore size distributions of NC and NC after the treatment with potassium hydroxide.

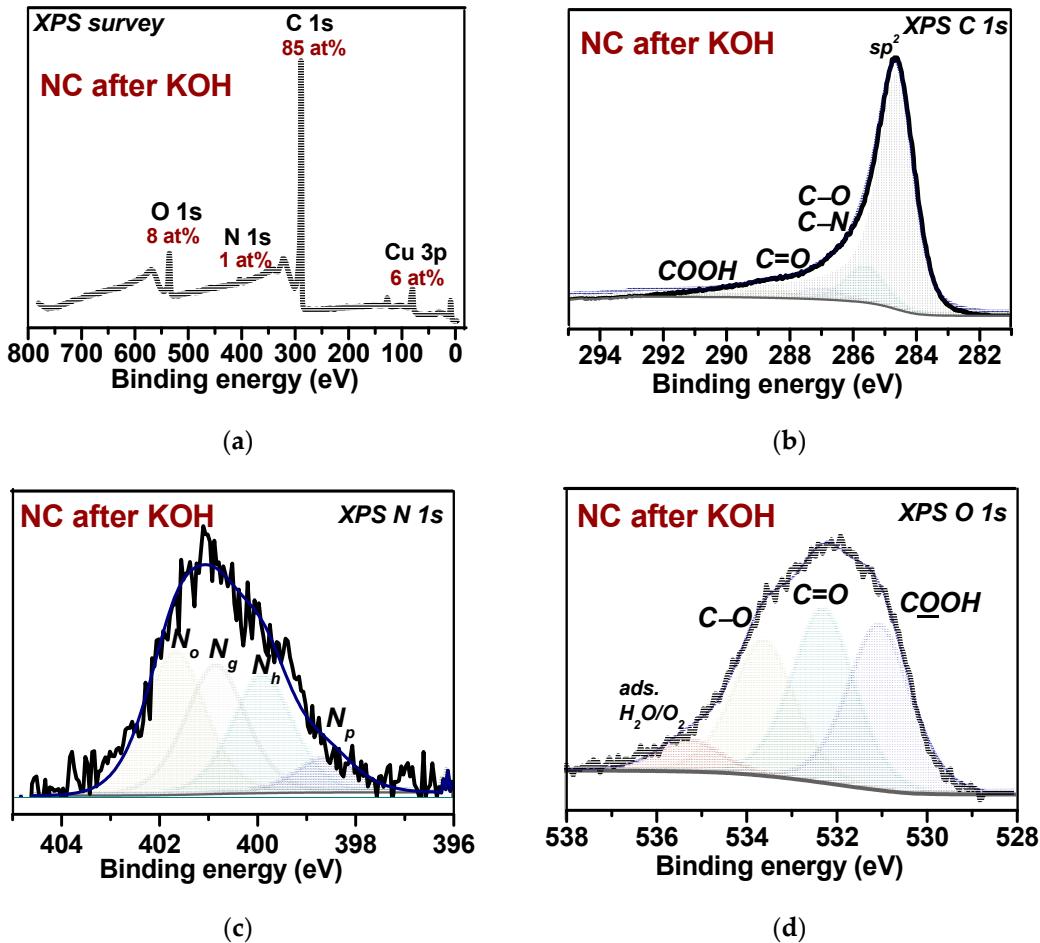


Figure S2. XPS (a) survey, (b) C 1s, (c) N 1s and (d) O 1s spectra of NC after the treatment with potassium hydroxide.

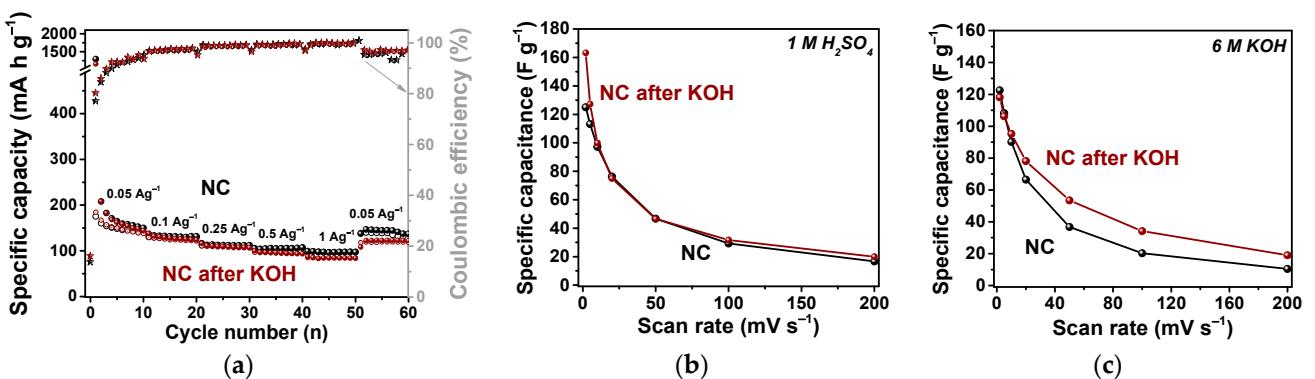


Figure S3. (a) Rate capability of NC and NC after KOH-treatment electrodes at current densities of 0.05–1.00 A·g⁻¹ in SIB cell; gravimetric capacitance of NC and NC after KOH-treatment at scan rates of 2–200 mV·s⁻¹ in (b) 1 M H₂SO₄ and (c) 1 M KOH electrolytes in supercapacitors.

Table S1. Capacitance in aqueous supercapacitors (SCs) in 1 M H₂SO₄ and 6 M KOH electrolytes and capacity in sodium-ion batteries (SIB) for nitrogen-doped carbon materials are reported in the literature

Material	Nitrogen concentration	Specific surface area	Capacitance in aqueous SCs or SIB capacity
SCs in 1 M H ₂ SO ₄ electrolyte			
activated carbon¹	2 wt.%	2062 m ² g ⁻¹	358 Fg ⁻¹ at 0.5 mV s ⁻¹
nitrogen-doped porous carbon²	3.3 wt.%	1463 m ² g ⁻¹	282 Fg ⁻¹ at 2 mV s ⁻¹ 148 Fg ⁻¹ at 100 mV s ⁻¹
mica templated carbon³	10 wt.%	87.5 m ² g ⁻¹	105 Fg ⁻¹ at 1 mV s ⁻¹
MOF-templated porous carbon⁴	5.3 wt.%	822 m ² g ⁻¹	72 Fg ⁻¹ at 5 mV s ⁻¹
	5.2 wt.%	586 m ² g ⁻¹	37 Fg ⁻¹ at 5 mV s ⁻¹
N-doped reduced graphene oxide⁵	3 at.%	355 m ² g ⁻¹	220 Fg ⁻¹ at 25 mV s ⁻¹
porous carbon nanosheets⁶	12.7 at.%	410 m ² g ⁻¹	305 Fg ⁻¹ at 2 mV s ⁻¹
activated carbon⁷	5.5 at.%	657 m ² g ⁻¹	200 Fg ⁻¹ at 10 mV s ⁻¹
SCs in 6 M KOH electrolyte			
nitrogen-containing carbon nanotubes⁸	6.1 wt.%	-	160 Fg ⁻¹ at 50 mV s ⁻¹
nitrogen-doped carbon nanoparticles⁹	10.5 wt.%	365 m ² g ⁻¹	156 Fg ⁻¹ at 2 mV s ⁻¹ 80 Fg ⁻¹ at 100 mV s ⁻¹
nitrogen-doped carbon microfibers¹⁰	4.5 wt.%	230 m ² g ⁻¹	189 Fg ⁻¹ at 5 mV s ⁻¹

¹ A. Elmouwahidi, Z. Zapata-Benabithe, F. Carrasco-Marín, C. Moreno-Castilla, Activated carbons from KOH-activation of argan (*Argania spinosa*) seed shells as supercapacitor electrodes, *Bioresource Technology*. 111 (2012) 185–190. <https://doi.org/10.1016/j.biortech.2012.02.010>.

² M. Zhou, F. Pu, Z. Wang, S. Guan, Nitrogen-doped porous carbons through KOH activation with superior performance in supercapacitors, *Carbon*. 68 (2014) 185–194. <https://doi.org/10.1016/j.carbon.2013.10.079>.

³ M. Kodama, J. Yamashita, Y. Soneda, H. Hatori, S. Nishimura, K. Kamegawa, Structural characterization and electric double layer capacitance of template carbons, *Materials Science and Engineering: B*. 108 (2004) 156–161. <https://doi.org/10.1016/j.mseb.2003.10.097>.

⁴ Q. Wang, W. Xia, W. Guo, L. An, D. Xia, R. Zou, Functional Zeolithic-Imidazolate-Framework-Templated Porous Carbon Materials for CO₂ Capture and Enhanced Capacitors, *Chem. Asian J.* 8 (2013) 1879–1885. <https://doi.org/10.1002/asia.201300147>.

⁵ Y.-H. Lee, K.-H. Chang, C.-C. Hu, Differentiate the pseudocapacitance and double-layer capacitance contributions for nitrogen-doped reduced graphene oxide in acidic and alkaline electrolytes, *Journal of Power Sources*. 227 (2013) 300–308. <https://doi.org/10.1016/j.jpowsour.2012.11.026>.

⁶ Q. Wang, J. Yan, Z. Fan, Nitrogen-doped sandwich-like porous carbon nanosheets for high volumetric performance supercapacitors, *Electrochimica Acta*. 146 (2014) 548–555. <https://doi.org/10.1016/j.electacta.2014.09.036>.

⁷ W. Zhang, Z. Ren, Z. Ying, X. Liu, H. Wan, Activated nitrogen-doped porous carbon ensemble on montmorillonite for high-performance supercapacitors, *Journal of Alloys and Compounds*. 743 (2018) 44–51. <https://doi.org/10.1016/j.jallcom.2017.12.327>.

⁸ O. Ornelas, J.M. Sieben, R. Ruiz-Rosas, E. Morallón, D. Cazorla-Amorós, J. Geng, N. Soin, E. Siores, B.F.G. Johnson, On the origin of the high capacitance of nitrogen-containing carbon nanotubes in acidic and alkaline electrolytes, *Chem. Commun.* 50 (2014) 11343–11346. <https://doi.org/10.1039/C4CC04876H>.

⁹ L.-P. Lv, Z.-S. Wu, L. Chen, H. Lu, Y.-R. Zheng, T. Weidner, X. Feng, K. Landfester, D. Crespy, Precursor-controlled and template-free synthesis of nitrogen-doped carbon nanoparticles for supercapacitors, *RSC Adv.* 5 (2015) 50063–50069. <https://doi.org/10.1039/C5RA06697B>.

¹⁰ R. Liu, L. Pan, L. Wan, D. Wu, An evaporation-induced tri-consistent assembly route towards nitrogen-doped carbon microfibers with ordered mesopores for high performance supercapacitors, *Phys. Chem. Chem. Phys.* 17 (2015) 4724–4729. <https://doi.org/10.1039/C4CP05211K>.

nitrogen-containing mesoporous carbon¹¹	8.4 wt.%	930 m ² g ⁻¹	230 Fg ⁻¹ at 5 mV s ⁻¹ 215 Fg ⁻¹ at 100 mV s ⁻¹
nitrogen-doped hierarchically porous carbon material¹²	6.3 at.%	1091 m ² g ⁻¹	187 Fg ⁻¹ at 5 mV s ⁻¹
SIBs			
N-rich carbon nanosheets¹³	12 at.%	352 m ² g ⁻¹	185 mAhg ⁻¹ at 0.1 Ag ⁻¹
N-doped nano-sized carbon spheres¹⁴	9.12 at.%	604 m ² g ⁻¹	245 mAhg ⁻¹ at 0.03 Ag ⁻¹
N-doped amorphous carbon nanofibers¹⁵	7.29 at.%	370 m ² g ⁻¹	321 mA h g ⁻¹ at 0.05 Ag ⁻¹
N-doped carbon/graphene¹⁶	7.54 at.%	94 m ² g ⁻¹	303 mAhg ⁻¹ at 0.05 Ag ⁻¹ 274 mAhg ⁻¹ at 0.1 Ag ⁻¹ 94 mAhg ⁻¹ at 5 Ag ⁻¹
N-rich hollow carbon-onion-constructed nanosheets¹⁷	16.54 at.%	108 m ² g ⁻¹	262 mAhg ⁻¹ at 0.1 Ag ⁻¹ 151 mAhg ⁻¹ at 5 Ag ⁻¹
N-doped porous carbon nanofibers¹⁸	-	138 m ² g ⁻¹	174 mAhg ⁻¹ at 0.05 Ag ⁻¹ 156 mAhg ⁻¹ at 0.1 Ag ⁻¹
N-doped carbon fiber aerogels¹⁹	12.2 at.%	65 m ² g ⁻¹	~170 mAhg ⁻¹ at 0.1 Ag ⁻¹ ~140 mAhg ⁻¹ at 1 Ag ⁻¹
N-doped carbon nanobubbles²⁰	11.6 at.%	672 m ² g ⁻¹	248 mAhg ⁻¹ at 0.3 Ag ⁻¹
N-doped carbon nanotubes²¹	5 at.%	737 m ² g ⁻¹	185 mAhg ⁻¹ at 0.1 Ag ⁻¹ 153 mAhg ⁻¹ at 1 Ag ⁻¹

¹¹ A. Olejniczak, M. Leżańska, A. Pacuła, P. Nowak, J. Włoch, J.P. Łukaszewicz, Nitrogen-containing mesoporous carbons with high capacitive properties derived from a gelatin biomolecule, *Carbon*. 91 (2015) 200–214. <https://doi.org/10.1016/j.carbon.2015.04.025>.

¹² R. Zeng, X. Tang, B. Huang, K. Yuan, Y. Chen, Nitrogen-Doped Hierarchically Porous Carbon Materials with Enhanced Performance for Supercapacitor, *ChemElectroChem.* 5 (2018) 515–522. <https://doi.org/10.1002/celc.201701021>.

¹³ B. Yang, S. Liu, Y. V. Fedoseeva, A. V. Okotrub, A.A. Makarova, X. Jia, J. Zhou, Engineering selenium-doped nitrogen-rich carbon nanosheets as anode materials for enhanced Na-Ion storage, *J. Power Sources*. 493 (2021) 229700. <https://doi.org/10.1016/j.jpowsour.2021.229700>.

¹⁴ A. Agrawal, S. Janakiraman, K. Biswas, A. Venimadhav, S.K. Srivastava, S. Ghosh, Understanding the improved electrochemical performance of nitrogen-doped hard carbons as an anode for sodium ion battery, *Electrochim. Acta*. 317 (2019) 164–172. <https://doi.org/10.1016/j.electacta.2019.05.158>.

¹⁵ R. Hao, Y. Yang, H. Wang, B. Jia, G. Ma, D. Yu, L. Guo, S. Yang, Direct chitin conversion to N-doped amorphous carbon nanofibers for high-performing full sodium-ion batteries, *Nano Energy*. 45 (2018) 220–228. <https://doi.org/10.1016/j.nanoen.2017.12.042>.

¹⁶ H. Liu, M. Jia, B. Cao, R. Chen, X. Lv, R. Tang, F. Wu, B. Xu, Nitrogen-doped carbon/graphene hybrid anode material for sodium-ion batteries with excellent rate capability, *J. Power Sources*. 319 (2016) 195–201. <https://doi.org/10.1016/j.jpowsour.2016.04.040>.

¹⁷ S. Liu, B. Yang, J. Zhou, H. Song, Nitrogen-rich carbon-onion-constructed nanosheets: An ultrafast and ultrastable dual anode material for sodium and potassium storage, *J. Mater. Chem. A*. 7 (2019) 18499–18509. <https://doi.org/10.1039/c9ta04699b>.

¹⁸ Y. Qu, M. Guo, F. Zeng, C. Zou, C. Yuan, X. Zhang, Q. Li, H. Lu, Synthesis of nitrogen-doped porous carbon nanofibers as an anode material for high performance sodium-ion batteries, *Solid State Ionics*. 337 (2019) 170–177. <https://doi.org/10.1016/j.ssi.2019.04.025>.

¹⁹ Y. Lu, D. Li, C. Lyu, H. Liu, B. Liu, S. Lyu, T. Rosenau, D. Yang, High nitrogen doped carbon nanofiber aerogels for sodium ion batteries: synergy of vacancy defects to boost sodium ion storage, *Appl. Surf. Sci.* 496 (2019) 143717. <https://doi.org/10.1016/j.apsusc.2019.143717>.

²⁰ L. Sun, J. Xie, X. Zhang, L. Zhang, J. Wu, R. Shao, R. Jiang, Z. Jin, Controllable synthesis of nitrogen-doped carbon nanobubbles to realize high-performance lithium and sodium storage, *Dalt. Trans.* 49 (2020) 15712–15717. <https://doi.org/10.1039/d0dt03258a>.

²¹ Kang Ding, Biao Gao, Jijiang Fu, Weili An, Hao Song, Xingxing Li, Qiuyun Yuan, Xuming Zhang, Kaifu Huo, and Paul K. Chu. Intertwined Nitrogen-Doped Carbon Nanotubes for High-Rate and Long-Life Sodium-Ion Battery.pdf, *ChemElectroChem*. 4 (2017) 2542–2546. <https://doi.org/https://doi.org/10.1002/celc.201700590>.

N-doped double-shell hollow carbon sphere²²	-	674 m ² g ⁻¹	250 mAhg ⁻¹ at 0.06 Ag ⁻¹ 137 mAhg ⁻¹ at 1.2 Ag ⁻¹
N-doped carbon spheres²³	-	-	152 mAhg ⁻¹ at 0.05 Ag ⁻¹ 83 mAhg ⁻¹ at 1 Ag ⁻¹
hierarchically porous N-doped carbon²⁴	2.1 wt.%	800 m ² g ⁻¹	205 mAhg ⁻¹ at 0.1 Ag ⁻¹ 132 mAhg ⁻¹ at 1 Ag ⁻¹

²² L. Bu, X. Kuai, W. Zhu, X. Huang, K. Tian, H. Lu, J. Zhao, L. Gao, Nitrogen-doped double-shell hollow carbon spheres for fast and stable sodium ion storage, *Electrochim. Acta.* 356 (2020) 136804. <https://doi.org/10.1016/j.electacta.2020.136804>

²³ M. Khan, N. Ahmad, K. Lu, Z. Sun, C. Wei, X. Zheng, R. Yang, Nitrogen-doped carbon derived from onion waste as anode material for high performance sodium-ion battery, *Solid State Ionics.* 346 (2020) 115223. <https://doi.org/10.1016/j.ssi.2020.115223>.

²⁴ J. Ou, L. Yang, Z. Zhang, Chrysanthemum derived hierarchically porous nitrogen-doped carbon as high performance anode material for Lithium/Sodium ion batteries, *Powder Technol.* 344 (2019) 89–95. <https://doi.org/10.1016/j.powtec.2018.11.100>.