View Article Online View Journal

ChemComm

Accepted Manuscript

This article can be cited before page numbers have been issued, to do this please use: G. Lan, Y. Wang, Y. Qiu, X. Wang, J. Liang, W. Han, H. Tang, H. Liu, J. Liu and Y. Li, *Chem. Commun.*, 2017, DOI: 10.1039/C7CC09370E.



This is an Accepted Manuscript, which has been through the Royal Society of Chemistry peer review process and has been accepted for publication.

Accepted Manuscripts are published online shortly after acceptance, before technical editing, formatting and proof reading. Using this free service, authors can make their results available to the community, in citable form, before we publish the edited article. We will replace this Accepted Manuscript with the edited and formatted Advance Article as soon as it is available.

You can find more information about Accepted Manuscripts in the **author guidelines**.

Please note that technical editing may introduce minor changes to the text and/or graphics, which may alter content. The journal's standard <u>Terms & Conditions</u> and the ethical guidelines, outlined in our <u>author and reviewer resource centre</u>, still apply. In no event shall the Royal Society of Chemistry be held responsible for any errors or omissions in this Accepted Manuscript or any consequences arising from the use of any information it contains.



rsc.li/chemcomm

Published on 19 December 2017. Downloaded by University of Florida Libraries on 20/12/2017 02:32:29

Journal Name



Wheat flour derived N-doped mesoporous carbons extrudate as superior metal-free catalysts for acetylene hydrochlorination

Received 00th January 20xx, Accepted 00th January 20xx

Guojun Lan^a, Yan Wang^a, Yiyang Qiu^a, Xiaolong Wang^a, Ji Liang^b, Wenfeng Han^a, Haodong Tang^a, Huazhang Liu^a, Jian Liu^{*c,d} and Ying Li^{*a}

DOI: 10.1039/x0xx00000x

www.rsc.org/

N-doped mesoporous carbons extrudate with major quaternary N species are successfully prepared through a direct carbonization of wheat flour/gluten with silica, which is a cheap and convenient for scale-up production approach. The obtained carbon extrudate metal-free catalyst enables the high efficient production of VCM through acetylene hydrochlorination, with a superior catalytic performance and excellent stability (>85% conversion and vinyl chloride selectivity over 99% at 220 °C).

The vinyl chloride monomer (VCM) has been widely used to manufacture polyvinyl chloride (PVC), which is the raw chemical for a wide range of engineering plastics. The production of VCM is mainly through the acetylene hydrochlorination reaction, utilizing activated carbon supported mercury (II) chloride (HgCl₂/AC) as the catalyst, which is highly toxic and harmful to both human health and environment. ^[1] The pioneering work and systematic studies carried out by Hutchings and co-workers showed that AuCl₃ might be the best replacement of HgCl₂ for this process because of its high activity. Johnson Matthey technology claimed the successful commercialization gold catalysts with low loading and thiosulfate as ligand in 2016^[2]. Despite the recent commercialization of gold catalysts for acetylene hydrochlorination, it is still a demand to exploration of new low-cost catalysts system with high activity and stability.

Doping of carbons by nitrogen modifies not only the carbon surface chemistry but also the electronic structure by donating a lone pair of electrons on the undoped carbon lattice, which may enhance the adsorption of HCl and activate acetylene.^[4] To this end, various N-doped carbon materials

have been explored as novel catalysts for acetylene hydrochlorination.^[4b, 4c, 5, 6] For example, N-doped carbon nanotubes (N-CNTs) can enhance the formation of the covalent bond between C_2H_2 and N-CNTs to promote the catalytic activity for acetylene hydrochlorination.^[5] Bao and coworkers demonstrated that a nanocomposite of N-doped carbon derived from silicon carbide has an excellent performance for acetylene hydrochlorination with a conversion of 80% at 200 °C, 30 h⁻¹.^[4c] Although it has been demonstrated that the N-doped carbon materials are active and promising metal-free catalysts for acetylene hydrochlorination, it still remains challenging to develop and scale up the fabrication of N-doped carbon materials through facile, green, and low-cost routes.^[7]

In the past decade, the utilization of biomass to obtain carbonaceous materials has attracted much research and industrial interest.^[8] Different biomass such as glucose, sucrose, fructose, starch, furfural, dopamine, and grass have been tried as renewable and cheap precursors for nanocarbons. Among them, the carbon materials prepared via starch (i.e. Starbon®) have gained much attention due to their high surface area, tuneable surface chemistry, and controllable electrical conductivity. In addition, starch is a sustainable, reusable, and environmentally benign material, that is easy to form composites in particulate/monolithic forms.^[9] However, the Starbon® prepared from pure starch does not contain N dopant, and the active sites have to be introduced by additional nitrogen precursors, either in-situly or ex-situly.^[10] Compared with starch, natural wheat flour contains abundant gluten (ca. 10-12%) and certain amount of fat, vitamins, minerals, etc (ca. 13-20%), which are all rich in nitrogen.^[11] Every year, about 700 million tons of wheat is produced worldwide, and gluten is one of the main by-products in the wheat industry,^[12] which needs to be removed in some cases because of the gluten-related health issues. These factors thus make it very convenient to directly utilize wheat flour and/or gluten as naturally abundant and low-cost precursors for Ndoped carbons to catalyze the acetylene hydrochlorination.

^{a.} Institute of industry catalysis, Zhejiang University of technology Chaowang road 18, Hangzhou, PR China E-mail: liying@zjut.edu.cn.

^{b.} Institute for Superconducting & Electronic Materials, Australian Institute of

Innovative Materials, University of Wollongong, Innovation Campus, Squires Way, North Wollongong, NSW 2500, Australia. ^{c.} State Key Laboratory of Catalysis, iChEM, Dalian Institute of Chemical Physics,

Chinese Academy of Sciences, 457 Zhongshan Road, Dalian 116023, China

^d Department of Chemical and Process Engineering, University of Surrey, Guildford, Surrey, GU2 7XH, UK E-mail: jian.liu@surrey.ac.uk.

Electronic Supplementary Information (ESI) available: Experimental details, Figure S1-9 and Table S1-2 see DOI: 10.1039/x0xx00000x

Published on 19 December 2017. Downloaded by University of Florida Libraries on 20/12/2017 02:32:29

Page 2 of 5

DOI: 10.1039/C7CC09370E Journal Name

Based on these considerations and inspired by the flourbased food cooking method, herein, we for the first time demonstrate a facile and easily scaled-up method to synthesize N-doped mesoporous carbon, using wheat flour or gluten as both C and N precursors without any other additional nitrogen source. Scheme 1 describes the preparation process for N-doped mesoporous carbon (N-MCs) from wheat flour and silica spheres (i.e. the hard template for mesopores). The mixture of these components was made into dough by adding a suitable amount of deionized water. This dough can be easily and controllably made into any desirable shapes. In this study, it was made into cylinder-shaped extrudate ($\phi 2.5 \times 30$ mm) using a home-made noodle machine (Figure S1-2). These extrudate were then subjected to carbonization at 850°C in an inert atmosphere and subsequent HF etching to remove the silica templates. After these treatments, the cylinder shape of the material can be well-maintained, which is hard to achieve in other methods and is extremely important for acetylene hydrochlorination in the fix-bed reactors. For comparison, pure starch (derived from corn flour) and gluten, are used as precursors to prepare the mesoporous carbons, which are denoted as MC-St, N-MC-G, respectively. MC-Su sample was prepared by using sucrose as carbon source and a MCN sample was prepared by using ethylenediamine and carbon tetrachloride as carbon and nitrogen precursors according to references^[13-14] for comparison.



Scheme 1. The preparation process of the nitrogen doped mesoporous carbon N-MC-W using wheat flour as C&N sources.

The porosity of the materials was investigated by nitrogen adsorption-desorption technique (Figure 1a, Figure S3). Typically, the materials possess type IV isotherms with an H1 hysteresis loop extending from middle to high pressure region, indicating the existence of both mesopores and macropores on the material. The specific surface area of N-MC-W is 672 $m^2 g^{-1}$ and the pore volume is 0.86 cm³ g⁻¹, which are on the similar level as the other analogues (Table 1). The pore size distribution of the materials is then calculated by the Barrett-Joyner-Halenda method, which is centred at ca. 9.7 nm in the mesopore region for all the materials (inset of Figure 1a). The micro-structure of the materials was then studied by transmission electron microscopy (TEM, Figure 1b, c). Large amount of mesopores can be clearly observed on the material, which is consistent with the results of the nitrogen adsorptiondesorption results.

The chemical composition of the materials was then studied by element analysis (Table 1). The N contents are 1.6%

for N-MC-W and 4.2% for N-MC-G, respectively. The higher N content in the N-MC-G can be attributed to the higher N content in gluten than in flour. In contrast, the materials derived from sucrose (MC-Su) or starch (MC-St) contain much lower N contents (0.5 and 0.3%, respectively), possibly due to experimental error or comes from the impurities of the raw materials. The distribution of the elements was subsequently traced by elemental mapping under energy-filtered TEM (Figure 1d-f and S4), which shows a homogeneous distribution of both C, N, and O in the materials and indicates the large amount of N active sites throughout the materials.



Figure 1. N_2 sorption and pore size distribution (a), HRTEM (b) and STEM (c) and C (d), N (e), and O (f) element mapping for N-MC-W.

Table 1 texture properties and elemental composition of various carbons

	S. A. (m²	P. V. (cm ³ g ⁻¹)	P.D ^a (nm)	Deac	Elements Composition (%)			
Samples				Tivati				
				on	С	н	Ν	0
	g ⁻¹)			(%				
				h ⁻¹) ^b				
AC	1038	0.52	1.8	0.40	91.1	0.8	<0.3	7.8
MC-Su	862	1.25	9.7	0.61	87.1	2.6	<0.3	9.5
MC-St	715	0.60	9.7	0.22	85.8	1.4	<0.3	12.5
N-MC-W	672	0.86	9.7	0.16	80.9	1.7	1.6	15.8
N-MC-G	582	0.75	9.7	0.04	81.2	1.8	4.2	12.6
N-MC-G-	450	0.60	0.6		012	1 5	47	0.5
used	450	0.09	9.0	-	04.5	1.5	4.7	9.5
MCN	422	0.55	4.5	1.20	70.6	2.3	16.5	10.8

[a]calculated based on BJH method; [b] calculated based on the conversion of acetylene ($X_{10h}\text{-}X_{30h})/20h.$

The catalytic activity of these N-doped mesoporous carbons was evaluated for acetylene hydrochlorination at 220 $^{\circ}$ C and space velocity of 30 h⁻¹ (Figure 2a). Both N-MC-W from wheat flour and N-MC-G from gluten show a high acetylene conversion of 73.3% and 85.5%, respectively. In comparison, MC-Su and MC-St, prepared from sucrose and starch as carbon precursors, have a C₂H₂ conversion lower than 30%. The relatively higher performance of N-MC-G than N-MC-W can be the results of its higher N content. Figure S5 gives the TPD-MS profiles corresponding to the MC-Su, MC-St, N-MC-W and N-MC-G. It can be seen that the amounts of CO₂ and CO desorbed for these catalysts are similar, which are in accordance with the element analysis. These results indicate

Published on 19 December 2017. Downloaded by University of Florida Libraries on 20/12/2017 02:32:29

that the oxygen-containing group is not the main reason for the big difference of activity. Consequently, N-MC-G and N-MC-W are readily suitable for acetylene hydrochlorination; while MC-Su and MC-St are less favorable unless further treatments.



Figure 2 (a): The performance of various mesoporous carbon N-MC-G, N-MC-W, MC-Su and MC-St in acetylene hydrochlorination. Reaction conditions: 220 $^{\circ}$ C, 0.1 MPa, GHSV = 30 h⁻¹; (b): N1s XPS spectra of N-MC-W; (c): The stability test of N-MC-G, AC, and MCN in acetylene hydrochlorination. Reaction conditions: 220 $^{\circ}$ C, 0.1 MPa, GHSV = 30 h⁻¹; (d): N1s XPS spectra of N-MC-G and MCN.

The stability of the carbon materials in gas phase reaction is critical. The N-MC-G was then tested for acetylene hydrochlorination at 220 $^{\circ}$ C, 30 h^{-1} for 100 hours, in comparison with the commercial activated carbon and MCN (Figure 2b), and the corresponding deactivation rates of various carbon materials are summarized in Table 1. It is noticeable that, apart from their much higher catalytic activity, the N-MC-G and N-MC-W also have significantly lower deactivation rates (0.04 and 0.16%/h) compared with other materials (in the range of 0.22-1.20%/h). It is remarkable that, among the materials, the N-MC-G possesses the lowest deactivation rate, which is almost one-fold lower than that of AC (the detailed characterization of activated carbon was given in our previous work ^[15]). It should be notes that although MCN samples has very high N loadings (15.4 wt%), its conversion of acetylene and stability is lower than that of N-MC-G and N-MC-W samples. The MCN have lower surface area and a smaller pore size compared with N-MC-G. But these differences will not make the big difference in catalytic performance. From Table 1, it can be seen that the surface area of N-MC-G and N-MC-W are lower than MC-St and MC-Su. But the activity is much higher. This result indicates that the surface area has smaller effect than that of N contents. The graphitization degree of N-MC-G and MCN are characterized by XRD and Raman spectroscopy, given in Figure S6. It can be seen that XRD pattern and Raman spectra of the N-MC-G and MCN and calculated I_D/I_G ratios are similar. These results indicate that the graphitic degree is not the main reason for the big difference of activity.

Great efforts has been made from various research groups to identify the active sites of N doped carbon materials in acetylene hydrochlorination by using experimental and theory calculations methods, however, the role of different N species in active sites is still under debate^[1,3-5]. To better understand the roles of the nitrogen species during the catalysis and to identify the deactivation mechanism of the materials, we then conducted X-ray photo electron spectroscopy on the fresh and used catalysts (Figure 2b and d). The XPS spectra of N-MC-W and N-MC-G can be deconvoluted into four peaks, which are assigned to pyridinic (398.3 \pm 0.2 eV), pyrrolic (400.0 \pm 0.2 eV), quaternary (401.1 \pm 0.2 eV) and oxidized (403.0-405.6 eV) N species, respectively.^[11a, 16] The fitting analysis data shows that the main N specie in N-MC-W and N-MC-G are the quaternary nitrogen, which is around 60% in the samples (Figure S7a). While the major N species in MCN are pyrolic N and pyridinic N, which contributed above 75% to all N species.

DOI: 10.1039/C7CC09370E COMMUNICATION

In present work, on the apparent, the activity of N-MC-G with quaternary nitrogen as main N specie is more active than MCN with pyrolic N and pyridinic N as main N specie. However the current catalytic activity comparison in N-doped carbon materials is based on the N species contents. Bao et al. indicate that acetylene hardly adsorbs on the quaternary and pyridinic N sites because of the endothermic nature of the adsorption process and they concluded that carbon atoms bonded with pyrrolic N atoms are the active sites.^[4c] Kang et al. summarized that the defects in graphene could efficiently improve the catalytic performance to DFT calculated results.^[17] Therefore the origin of catalytic activity in carbon materials is affiliated to the status of carbon atom. The surface oxygen functional groups, edge dangling bonds, and defective sites in nanocarbon materials may also contribute to their catalytic activity. The contribution of different N species in active sites in different carbon materials may different. As reported in references, Wei et al. reported that the guaternary N is the biggest contributing N-containing form in N-doped carbon nanotubes.^[5] While Dai et al. demonstrated that pyrrolic N is the most important nitrogen species graphitic carbon nitride and N-doped active carbon catalyst.^[4b] Li et al. indicated the pyridinic N are the active sites in N-doped porous carbon derived from ZIF-8.[6a]

After the long-term stability test, the sample (denoted as N-MC-G-used) was subsequently analysed to further identify the stability of various N species during the catalysis. From the elemental analysis data given in Table 1, it can be found that the N content for the fresh and used catalysts are 4.2 and 4.7%, respectively. The C content increases from and 81.2 to 84.3% and the oxygen content decreases from 12.6% to 9.5%. In comparison, the XPS survey scan shows that the surface N contents of N-MC-G subjected to a larger amount of decrease after the 100 h reaction (5.7% vs. 4.5%, Table S1). The actual content of quaternary N for the spent N-MC-G catalyst is decreased compared with the fresh N-MC-G catalyst. (For the fresh N-MC-G catalyst, the loading of quaternary N= 5.67%×59.97% = 3.40%; for the spent N-MC-G catalyst the loading of quaternary N= 4.48% × 62.89% = 2.85%). Considering the low detection depth of XPS technique, the inconsistent results of elemental analysis and XPS results consequently reveal that it is mainly the surface N species that significantly decreased during the test, while the bulk N species remains. This may be caused by the carbon deposition on the surface of the catalyst and which is further been

COMMUNICATION

approved by the loss of the surface area (Table 1). By comparing the nitrogen adsorption-desorption isotherms and pore size distributions of the fresh and used N-MC-G (Figure S8), it can be seen that the isotherm curves are similar, indicates that the carbon deposition is not heavy. But most of micropore and parts of mesopore are blocked from pore size distributions. This may seriously affect the mass transfer of reactants. The mass loss of fresh and used N-MC-G catalysts under an oxygen atmosphere is compared in Figure S9. The coke deposition is only 0.66% calculated based on the mass loss differences of N-MC-G and N-MC-G-used in the 100-450 oC range. That means the coke deposition occurs but not very heavy, this is consistent with the low deactivation rate of this catalyst during 100 hours. As reported in references ^[18], the N doped carbon materials also show excellent performance been used as the support in acetylene hydrochlorination for Au based mercury free catalysts, therefore, Au/N-MC-W catalyst was prepared and tested. It can be seen that the activity of its support gold catalyst can be greatly improved (Table S2). Further studied on the catalytic performance of N-doped mesoporous carbons extrude are under progress in our lab.

Conclusions

Published on 19 December 2017. Downloaded by University of Florida Libraries on 20/12/2017 02:32:29

In conclusion, the N-doped mesoporous carbons were successfully prepared by using wheat flour or gluten as both carbon and nitrogen source, which shows high activity and stability for acetylene hydrochlorination as a metal free catalyst. The wheat flour, an excellent natural-born mixture of starch with gluten, is an excellent C and N precursor to prepare N-MCs with homogenously distributed N atom doped as graphitic framework active site, and the sticky properties of gluten make the shape of the final carbon materials easier and more convenience. The wheat flour derived granular N-doped mesoporous carbon have many remarkable advantages which make them ideal catalysts for a number of key industrial processes: (i) highly homogeneous distribution of the heteroatom dopants (N) within the composite; (ii) easy, costeffective, and environmentally safe raw material for upscale syntheses. (iii) 3D shaped open-cell structures with low mass transfer limitation and relatively high surface area. The kilogram scale preparation of the gluten derived N dope mesoporous carbon is under investigations in our lab.

Conflicts of interest

There are no conflicts to declare.

Acknowledgements

The authors would like to thank for Professor Xianping Yao in Hangzhou research institute of chemical technology Co Ltd for providing the starch sample and fruitful disscusion for understanding the properties of the starch and related materials. The financial support from Natural Science Foundation of China (NSFC Grant No. 21776257) and Natural Science Foundation of Zhejiang Province (LY17B030010) is gratefully acknowledged

Notes and references

- a) M. Y. Zhu, Q. Q. Wang, K. Chen, Y. Wang, C. F. Huang, H. Dai, F. Yu, L. H. Kang, B. Dai, *ACS Catal.* **2015**, *5*, 5306-5316; b) J. Oliver-Meseguer, A. Doménech-Carbó, M. Boronat, A. Leyva-Pérez, A. Corma, *Angew. Chem. Int. Ed.* **2017**, *56*, 6435–6439.
- 2 a) G. Malta, S. A. Kondrat, S. J. Freakley, C. J. Davies, L. Lu, S. Dawson, A. Thetford, E. K. Gibson, D. J. Morgan, W. Jones, P. P. Wells, P. Johnston, C. R. A. Catlow, C. J. Kiely, G. J. Hutchings, *Science* 2017, *355*, 1399-1402; b) P. Johnston, N. Carthey, G. J. Hutchings, *J. Am. Chem. Soc.* 2015, *137*, 14548-14557; c) G. Malta, a S. J. Freakley, S. A. Kondrata, G. J. Hutchings, *Chem. Commun.*, 2017, 53, 11733-11746.
- 3 K. Zhou, J. Jia, C. Li, H. Xu, J. Zhou, G. Luo, F. Wei, *Green Chem.* **2015**, *17*, 356-364.
- 4 a) Y. J. Gao, G. Hu, J. Zhong, Z. J. Shi, Y. S. Zhu, D. S. Su, J. G. Wang, X. H. Bao, D. Ma, *Angew. Chem. Int. Ed.* 2013, *52*, 2109-2113; b) X. Y. Li, Y. Wang, L. H. Kang, M. Y. Zhu, B. Dai, *J. Catal.* 2014, *311*, 288-294; c) X. Y. Li, X. L. Pan, L. Yu, P. J. Ren, X. Wu, L. T. Sun, F. Jiao, X. H. Bao, *Nat. Commun.* 2014, *5*, 4688.
- 5 K. Zhou, B. Li, Q. Zhang, J. Q. Huang, G. L. Tian, J. C. Jia, M. Q. Zhao, G. H. Luo, D. S. Su, F. Wei, *ChemSusChem* **2014**, *7*, 723-728.
- 6 a) S. L. Chao, F. Zou, F. F. Wan, X. B. Dong, Y. L. Wang, Y. X. Wang, Q. X. Guan, G. C. Wang, W. Li, *Sci. Rep.* 2017, *7*, 39789;
 b) X. Y. Li, J. L. Zhang, W. Li, *J. Ind. Eng. Chem.* 2016, 44, 146-154.
- 7 T. C. Wang, N. A. Vermeulen, I. S. Kim, A. B. F. Martinson, J. F. Stoddart, J. T. Hupp, O. K. Farha, *Nat. Protoc.* **2016**, *11*, 149-162.
- a) R. J. White, N. Brun, V. L. Budarin, J. H. Clark, M. M. Titirici, *ChemSusChem* 2014, 7, 670-689; b) R. J. White, V. L. Budarin, J. H. Clark, *Chem. Eur. J.* 2010, 16, 1326-1335; c) S. P. Wang, C. L. Han, J. Wang, J. Deng, M. L. Zhu, J. Yao, H. R. Li, Y. Wang, *Chem. Mater.* 2014, 26, 6872-6877.
- 9 a) V. Budarin, J. H. Clark, J. J. E. Hardy, R. Luque, K. Milkowski, S. J. Tavener, A. J. Wilson, *Angew. Chem. Int. Ed.* **2006**, *45*, 3782-3786; b) R J. White, V Budarin, R Luque, J H. Clark D J. Macquarriea, *Chem. Soc. Rev.* **2009**, 38, 3401-3418
- 10 a) C. Tang, H. F. Wang, X. Chen, B. Q. Li, T. Z. Hou, B. S. Zhang, Q. Zhang, M. M. Titirici, F. Wei, *Adv. Mater.* **2016**, 28, 6845-6851; b) P.P. Yu, Z.M. Zhang, L.X. Zheng, F. Teng, L.F. Hu, X.S. Fang, *Adv. Energy Mater.* **2016**, 6, 1601111
- 11 X. Y. Wang, X. N. Guo, K. X. Zhu, Food Chem. 2016, 201, 275-283.
- 12 M. V. Ostermann-Porcel, A. N. Rinaldoni, L. T. Rodriguez-Furlán, M. E Campderrós, J. Sci. Food Agric. 2017, 97: 2934-2941.
- 13 Z. L. Jiang, G. J. Lan, X. Y. Liu, H. D. Tang, Y. Li, *Catal. Sci. Technol.* **2016**, *6*, 7259-7266.
- 14 a) A. Vinu, Adv. Funct. Mater. 2008, 18, 816-827; b) Dai, B.; Li, X. Y.; Zhang, J. L.; Yu, F.; Zhu, M. Y., Chem. Eng. Sci. 2015, 135, 472-478.
- 15 Y. Wang, Y. Y. Qiu, X. L. Wang, Z. H. Li, G. J. Lan, Y Li, Chem. React. Eng. Technol. 2017, 33, 4, 298-304.
- 16 a) S. G. Zhang, S. Tsuzuki, K. Ueno, K. Dokko, M. Watanabe, Angew. Chem. Int. Ed. **2015**, 54, 1302-1306; b) X. G. Wang, B. Dai, Y. Wang, F. Yu, ChemCatChem **2014**, 6, 2339-2344.
- 17 F. Zhao, L. H. Kang, Chemistryselect 2017, 2, 6016-6022.
- a) B. Dai, X. Y. Li, J. L. Zhang, F. Yu, M. Y. Zhu, *Chem. Eng. Sci.* **2015**, 135, 472-478; b) J. Zhao, J. T. Xu, J. H. Xu, T. T. Zhang, X.
 X. Di, J. Ni, X. N. Li, *Chem. Eng. J.* **2015**, 262, 1152-1160.

This journal is © The Royal Society of Chemistry 20xx

ChemComm Accepted Manuscript

Published on 19 December 2017. Downloaded by University of Florida Libraries on 20/12/2017 02:32:29.

Table of Contents



Granular N-doped mesoporous carbons extrudate are successfully prepared through a food cooking inspired easy shaped method by using wheat flour/gluten as both carbon and nitrogen sources. This biomass derived metal-free N-doped mesoporous carbons featured superior catalytic performance and excellent stability for acetylene hydrochlorination.