Wide potential window (1.3V) supercapacitive study of high entropy alloy and green carbon composite electrode with its application towards aqueous

symmetric device

Gobinda Chandra Mohanty¹*, Anu Verma²

¹School of Nanoscience and Technology Indian Institute of Technology, Kharagpur, India

²School of Environmental Science and Engineering, Indian Institute of Technology, Kharagpur, India

*Corresponding authors.

E-mail address:

gobinda2020@kgpian.iitkgp.ac.in

Synthesis of green carbon:

The green carbon is prepared by pyrolysis technique of Rice straw agricultural waste. At first rice straw collected and cleaned by tap water followed by DI-water and dried. Then the RS-straw was cut into small pieces 1 to 2 mm size. Then the rice straw used in the for pyrolysis in absence of oxygen at 600 °C for 1 h to get the pyrolyzed material. This obtained material is further processed through ball milling as well as probe sonication to obtained green carbon.

Synthesis Process of green graphene



Figure S1: Schematic of synthesis of RS-biochar (green carbon) by pyrolysis technique.

3. Results and discussion

3.1. XRD

The XRD pattern of green carbon is shown the **Fig. S2(a)**. The angle of 2θ ranges from 20 to 30 with few intense peaks refers to graphene carbon structure with few Silica rich entity¹. This XRD behaviour matched with graphene-silica rich green carbon. The previous is also backing same pattern^{2, 3}.

3.2. FTIR

Additionally, using the FT-IR spectrum, it has been possible to determine whether surface functional groups are present in green carbon. According to **Fig. S2 (b)**, the peak values are observed at 3443 cm⁻¹, 1639 cm⁻¹, 1049 cm⁻¹, 784 cm⁻¹, and 463 cm⁻¹. According to⁴ the peaks at 3443 cm⁻¹ and 1634 cm⁻¹ are ascribed to the aromatic C=C bonds and the -OH stretching vibration, respectively. According to ⁵a prominent peak seen at 1050 cm⁻¹ is associated with asymmetric stretching (Si-O-Si), whereas 780 cm⁻¹ is caused by symmetric vibration (Si-O-Si).

In addition, a band detected at 463 cm⁻¹ could be attributed to the bending vibration of the Si-O-Si bond Older studies provide strong backing for these observations.

3.3. Raman studies

In order to investigate how graphene-based structures formed in the prepared sample, the Raman spectrum of the sample was also examined. The Raman spectrum displays two bands, as illustrated in **Fig. S2(c)**. According to⁶, 1344 cm⁻¹ and 1582 cm⁻¹ bands, respectively), these are the D and G bands. Accordingly the sp² carbon structure is responsible for the G-band, which is a feature of graphitic substances (first order scattering of E_{2g} phonon). The presence of defects, such as A_{1g} symmetry and sp³ disordered carbon, is what is responsible for the D-band, on the other hand. Indicating the presence of a multi-layer graphene structure, the relative intensity ratio of the Raman bands (I_D/I_G) is determined to be 1.



3.4. BET

Through N_2 physical adsorption measurements carried out at 77 K, an analysis of the porous structure of GGs has been investigated, and the results are displayed in **Fig. S2 (d)**. According to^{7,8} this graph can be categorised as a type-II curve. It is seen that the graphs exhibit increased adsorption up to a relative pressure (p/p°) of 0.8 with a relatively low N_2 sorption profile. This might be attributable to the capillary condensation that occurs within the GGs mesoporous channels. However, between relative pressures of 0.8 and 1.0, a relatively higher N_2 sorption characteristic could be observed, which suggests that GGs may have a microporous structure⁹. BET specific surface area and mean pore diameter were reported to be 7.3 m²/g and 4.6 nm, respectively. Furthermore, 7.4 cm³/g of total pore volume was discovered.



Figure S2: BET adsorption, desorption, and pore size distribution.



Figure S3: (*a*, *b*) *CV* and *GCD* curves of *FeCoNiCrMn HEA* electrode, (*c*, *d*) *CV* and gcd curves of green carbon electrode.

References:

 Autthawong, T.; Namsar, O.; Yu, A.; Sarakonsri, T. Cost-Effective Production of SiO2/C and Si/C Composites Derived from Rice Husk for Advanced Lithium-Ion Battery Anodes. *J. Mater. Sci. Mater. Electron.* 2020, *31* (12), 9126–9132. https://doi.org/10.1007/s10854-020-03442-3.

- (2) Sharma, S. K.; Sharma, G.; Sharma, A.; Bhardwaj, K.; Preeti, K.; Singh, K.; Kumar, A.; Pal, V. K.; Choi, E. H.; Singh, S. P.; Kaushik, N. K. Synthesis of Silica and Carbon-Based Nanomaterials from Rice Husk Ash by Ambient Fiery and Furnace Sweltering Using a Chemical Method. *Appl. Surf. Sci. Adv.* 2022, *8*, 100225. https://doi.org/10.1016/j.apsadv.2022.100225.
- (3) Tian, H.; Zhang, C.; Wang, Q.; Miao, J.; Zhang, Y.; Li, X.; Guo, Y.; Wang, M.; Chen, Y. SiO2 Aerogel @ Carbon Nanotube Anchored on Graphene Sheet as Anode Material for Lithium Ion Battery. *J. Mater. Sci. Mater. Electron.* 2021, *32* (9), 11478–11488. https://doi.org/10.1007/s10854-021-05661-8.
- Haeri, S. Z.; Ramezanzadeh, B.; Asghari, M. A Novel Fabrication of a High Performance SiO2-Graphene Oxide (GO) Nanohybrids: Characterization of Thermal Properties of Epoxy Nanocomposites Filled with SiO2-GO Nanohybrids. *J. Colloid Interface Sci.* 2017, 493, 111– 122. https://doi.org/10.1016/j.jcis.2017.01.016.
- (5) Narayanan, D. P.; Sankaran, S.; Narayanan, B. N. Novel Rice Husk Ash Reduced Graphene Oxide Nanocomposite Catalysts for Solvent Free Biginelli Reaction with a Statistical Approach for the Optimization of Reaction Parameters. *Mater. Chem. Phys.* 2019, 222, 63–74. https://doi.org/10.1016/j.matchemphys.2018.09.078.
- (6) Choudhary, R.; Koppala, S.; Swamiappan, S. Bioactivity Studies of Calcium Magnesium Silicate Prepared from Eggshell Waste by Sol–Gel Combustion Synthesis. J. Asian Ceram. Soc. 2015, 3 (2), 173–177. https://doi.org/10.1016/j.jascer.2015.01.002.
- Oh, W.-C.; Nguyen, D. C. T.; Areerob, Y. Enhanced Photocatalytic H2-Production and Photocatalytic Degradation Activity of Cadmium Oxide–Graphene Nanocomposite Grown on Mesoporous Silica under Visible Light Irradiation. *J. Porous Mater.* 2020, *27* (1), 151–163. https://doi.org/10.1007/s10934-019-00796-w.
- (8) Zhu, L.; Nguyen, D. C. T.; Woo, J.-H.; Zhang, Q.; Cho, K. Y.; Oh, W.-C. An Eco-Friendly Synthesized Mesoporous-Silica Particle Combined with WSe2-Graphene-TiO2 by Self-

Assembled Method for Photocatalytic Dye Decomposition and Hydrogen Production. *Sci. Rep.* **2018**, *8* (1), 12759. https://doi.org/10.1038/s41598-018-31188-w.

(9) Wang, J.; Chen, T.; Xu, B.; Chen, Y. Fabrication and Characterization of Porous Core–Shell Graphene/SiO2 Nanocomposites for the Removal of Cationic Neutral Red Dye. *Appl. Sci.* **2020**, *10* (23), 8529. https://doi.org/10.3390/app10238529.