Upcycling Waste Cotton Cloth into Carbon Textile: A Durable and Scalable Layer for Vanadium Redox Flow Batteries Applications

(Executive Summary)

Mohamed Adel Allam[1], **Mohammad Ali Abdelkareem**[1,2,3] , **Hussain Alawadhi**[1], **Abdul Ghani Olabi**[2,4], **Abdulmonem Fetyan**[5,6*]

1 Center for Advanced Materials Research, Research Institute of Sciences and Engineering, University of Sharjah, Sharjah PO Box 27272, United Arab Emirates.

2 Sustainable Energy & Power Systems Research Centre, RISE, University of Sharjah, Sharjah PO Box 27272, United Arab Emirates.

3 Chemical Engineering Department, Faculty of Engineering, Minia University, Egypt.

4 Mechanical Engineering and Design, School of Engineering and Applied Science, Aston University, Aston Triangle, Birmingham, B4 7ET, UK.

5 College of Interdisciplinary Studies, Zayed University, Academic City, Dubai, UAE.

6 Research & Development Centre, Dubai Electricity and Water Authority (DEWA), PO Box 564, Dubai, United Arab Emirates.

*Corresponding author: abdulmonem.fetyan@zu.ac.ae.

1. Introduction

The decrease in traditional fuel sources and the increasing worry regarding ecological contamination have accelerated the investigation of eco-friendly and enduring energy alternatives, like renewable power[1]. Nevertheless, these sustainable sources face obstacles, including sporadic availability and variable output[2]. To address these challenges, storage systems such as redox flow batteries (RFBs) have emerged to bridge the gap between electricity production and consumption[3]. Vanadium redox flow batteries (VRFBs), which Skyllas-Kazacos developed, are the RFB variation that has garnered the most interest because of their attractive features, which include their adjustable design, extended lifespan, and unique power and energy storage capabilities[4]. VRFBs utilize positive and negative vanadium ions in both electrolytes, reducing the risk of cross-contamination and ensuring prolonged cycle life and increased energy efficiency[5].

The electrodes employed in VRFB systems mainly incorporate carbon-derived substances owing to their outstanding attributes, such as superb electrical conductivity, stability, resistance to corrosion, and a broad operational potential range[6]. Graphite felts and carbon papers are commonly treated at ultra-high temperatures (>2000ºC) and extensively employed as electrodes in commercial VRFB setups[7]. Besides the complex fabrication process associated with these felts, their intrinsic low electrochemical activity and poor wettability require an additional activation, increasing their total cost significantly. Additionally, the relatively high cost poses a concern for large-scale commercial applications[8]. Numerous approaches have been undertaken to alleviate these constraints, such as thermal processing, plasma processing, electrochemical activation, microwaving, and hydrothermal treatment, among others, applied to conventional carbon electrodes[9-12]. These activation techniques enhance electrode wettability and augment vanadium ion reactions' catalytic efficacy.

Carbon electrocatalysts examples include carbon nanotubes (CNTs), graphene, and carbon nanofibers, alongside metal oxides such as PbO_2, CeO_2, Nd_2O_3, Mn_3O_4, Nb_2O_5, Co_3O_4, WO_3, TiO_2, TiN, and NiO, have been proposed to bolster the performance of the cell owing to their provision of significant surface area and favorable electrical conductivity[13-21]. Nonetheless, practical applications are impeded by challenges such as demanding synthesis conditions, elevated metal precursor costs, or restricted battery performance improvements[22]. Electrospun carbon nanofibers integrated with nanoparticle catalysts have emerged as innovative electrodes, boasting expansive surface areas, heightened electron conductivity, and robust electrocatalytic activity, demonstrating potential in VRFBs[23]. Nevertheless, scaling up these applications necessitates the creation of cost-effective and readily manufactured electrodes via a streamlined approach.

In recent decades, biomass-derived materials, known for their flexibility and low cost, have gained attention in energy devices like supercapacitors and lithium batteries[24-26]. In the realm of RFBs, the majority of documented research involving pure biomass electrodes utilizes materials inherently present in fibrous forms, which retain their fibrous structure even after undergoing carbonization[27]. Typical examples of these are coconut shells[28], corn protein[29], cotton pads[30], wood[31], silk fiber[32] and silk protein[33]. In our commitment to sustainability and waste recycling, we aim to develop a versatile and integrated electrode using biomass waste material for direct utilization in VRFBs. What sets us apart is not just the utilization of discarded cotton but its direct comparison with a more commercially established counterpart, namely pristine graphite felt post-thermal treatment. We opt for a cost-effective and high-yield plant material sourced from discarded clothes, making it an ideal precursor due to its simplicity, affordability, and widespread availability. This study marks the inaugural presentation of a biomass-derived carbonized wasted cloth (CWC) electrode fabricated via a simplified carbonization procedure, circumventing the post-activation steps typically associated with commercial graphite felts. The electrochemical effectiveness of the CWC electrode was evaluated through comprehensive full-cell testing and compared with a thermally treated graphite felt alternative. Analysis of the material's structure and surface area was conducted using various characterization techniques.

2. Research Question(s)

The main idea behind the study is to find an alternative sustainable and cost-effective material that can be used as electrode material in redox flow batteries. The research questions that we tried to address are whether wasted cotton cloths can be cost-effectively transformed and used as carbon material and how the performance of the lab-scale battery will be when equipping the cell with state-of-the-art electrodes.

3. Research Methods

The study involves several steps, starting with the electrode preparation step, followed by material characterization and electrochemical testing:

1. Preparation of the electrode material: Multiple layers of wasted cotton cloth were carbonized at 950 °C for two hours under N2 gas atmosphere.
2. Materials characterization: The prepared electrode material was characterized using a scanning electron microscope, X-ray diffraction, and Raman to better understand the nature of the prepared material.
3. Electrochemical testing: The state-of-the-art material was compared to heat-treated carbon felts for their electrochemical performance utilizing cyclic voltammetry, electrochemical impedance, and lab-scale battery tester to assess the overall system energy efficiency and capacity.

4. Key Findings

Integrating carbonized waste cloth (CWC) in batteries yields lower polarization, 67% higher energy efficiency, and enhanced rate capability.
Cost-effective CWC electrodes for VRFBs enhance sustainability in energy storage.
CWC electrodes reduce overpotentials with improved surface area and hydrophilicity.

5. Implications

Utilizing recycled materials aligns with sustainable practices, reducing waste and promoting a circular economy. Repurposing waste carbon cloth to be used as electrodes in VRFBs can significantly reduce the production costs of VRFBs, making them more affordable for long-duration large-scale energy storage projects. Such electrodes can improve the electrochemical performance of VRFBs, potentially increasing their efficiency and lifespan. However, practitioners need to ensure the quality and consistency of the recycled material and further investigate the potential degradation over time.

Policymakers can create incentives for companies that adopt sustainable practices, such as grants for using recycled materials and establishing standards for using recycled materials in energy storage systems, which can foster greater adoption of these technologies. Such adaptation will encourage the use of recycled materials in energy storage solutions, leading to achieving sustainability goals that reduce the environmental impact of energy production and result in high support for green policies.

Companies in the recycling and waste management sectors can find new business opportunities by supplying recycled carbon cloth to battery manufacturers, opening new market opportunities for such sectors.

6. Conclusion

High-performance carbonized wasted cloth electrodes were successfully created through a safe, cost-effective, and environmentally friendly preparation method. Compared to thermally treated graphite felt electrodes, these electrodes demonstrated substantial enhancements in conductivity and reduced polarization. The prepared electrodes exhibited remarkable electrocatalytic activity for V^{2+}/V^{3+} and VO^{2+}/VO_2^+ redox pairs in VRFBs. The CWC electrode possessed a significant surface area, a high level of carbonization, abundant mesopores distributions, amorphous structures, and high mechanical stability. The intricate interconnections observed between the fibers and the tubular character of the fiber contribute to enhanced electrolyte diffusion rates and the provision of ample active sites, rendering the CWC electrodes highly catalytic towards vanadium redox reactions when employed in VRFB applications. Notably, batteries outfitted with CWC-11 electrodes, possessing equivalent thickness to TT-GF, demonstrated remarkable energy efficiency of 67% and a capacity of discharge around 943 mAh at a current density of 100 mA cm^{-2}, surpassing those equipped with treated graphite electrodes, which recorded an energy efficiency of 58% and a capacity of discharge around 714 mAh. These favorable outcomes stem from accelerated electron transfer kinetics on the surface of the CWC electrodes, facilitating rapid redox reactions. This innovative, cost-effective, and high-performance material holds significant promise as a contender for advanced electrochemical storage systems.

Reference

[1] J. L. Holechek, H. M. E. Geli, M. N. Sawalhah, and R. Valdez, "A Global Assessment: Can Renewable Energy Replace Fossil Fuels by 2050?," *Sustainability*, vol. 14, no. 8. doi: 10.3390/su14084792

[2] W. Strielkowski, L. Civín, E. Tarkhanova, M. Tvaronavičienė, and Y. Petrenko, "Renewable Energy in the Sustainable Development of Electrical Power Sector: A Review," *Energies*, vol. 14, no. 24. doi: 10.3390/en14248240

[3] A. G. Olabi *et al.*, "Redox Flow Batteries: Recent Development in Main Components, Emerging Technologies, Diagnostic Techniques, Large-Scale Applications, and Challenges and Barriers," *Batteries*, vol. 9, no. 8. doi: 10.3390/batteries9080409

[4] M. Skyllas-Kazacos, M. H. Chakrabarti, S. A. Hajimolana, F. S. Mjalli, and M. Saleem, "Progress in Flow Battery Research and Development," *Journal of The Electrochemical Society,* vol. 158, no. 8, p. R55, 2011/06/27 2011.

[5] A. Fetyan *et al.*, "Electrospun Carbon Nanofibers as Alternative Electrode Materials for Vanadium Redox Flow Batteries," *ChemElectroChem,* vol. 2, no. 12, pp. 2055-2060, 2015/12/01 2015.

[6] Z. He *et al.*, "Electrode materials for vanadium redox flow batteries: Intrinsic treatment and introducing catalyst," *Chemical Engineering Journal,* vol. 427, p. 131680, 2022/01/01/ 2022.

[7] K. J. Kim, M.-S. Park, Y.-J. Kim, J. H. Kim, S. X. Dou, and M. Skyllas-Kazacos, "A technology review of electrodes and reaction mechanisms in vanadium redox flow batteries," *Journal of Materials Chemistry A,* 10.1039/C5TA02613J vol. 3, no. 33, pp. 16913-16933, 2015.

[8] L. F. Castañeda, F. C. Walsh, J. L. Nava, and C. Ponce de León, "Graphite felt as a versatile electrode material: Properties, reaction environment, performance and applications," *Electrochimica Acta,* vol. 258, pp. 1115-1139, 2017/12/20/ 2017.

[9] B. Sun and M. J. E. a. Skyllas-Kazacos, "Modification of graphite electrode materials for vanadium redox flow battery application—I. Thermal treatment," *Electrochimica acta* vol. 37, no. 7, pp. 1253-1260, 1992.

[10] B. Sun and M. J. E. A. Skyllas-Kazacos, "Chemical modification of graphite electrode materials for vanadium redox flow battery application—part II. Acid treatments," *Electrochimica Acta,* vol. 37, no. 13, pp. 2459-2465, 1992.

[11] W. Zhang *et al.*, "Electrochemical activation of graphite felt electrode for VO2+/VO2+ redox couple application," *Electrochimica Acta* vol. 89, pp. 429-435, 2013.

[12] Y. I. Cho *et al.*, "Effects of microwave treatment on carbon electrode for vanadium redox flow battery," vol. 2, no. 6, pp. 872-876, 2015.

[13] C. Yao, H. Zhang, T. Liu, X. Li, and Z. J. J. o. P. S. Liu, "Carbon paper coated with supported tungsten trioxide as novel electrode for all-vanadium flow battery," *Journal of Power Sources,* vol. 218, pp. 455-461, 2012.

[14] K. J. Kim *et al.*, "Novel catalytic effects of Mn 3 O 4 for all vanadium redox flow batteries," *Chemical Communications,* vol. 48, no. 44, pp. 5455-5457, 2012.

[15] H. Zhou *et al.*, "CeO2 decorated graphite felt as a high-performance electrode for vanadium redox flow batteries," *RSC Advances,* 10.1039/C4RA12339E vol. 4, no. 106, pp. 61912-61918, 2014.

[16] A. Fetyan, G. A. El-Nagar, I. Derr, P. Kubella, H. Dau, and C. Roth, "A neodymium oxide nanoparticle-doped carbon felt as promising electrode for vanadium redox flow batteries," *Electrochimica Acta,* vol. 268, pp. 59-65, 2018/04/01/ 2018.

[17] B. Li *et al.*, "Nanorod niobium oxide as powerful catalysts for an all vanadium redox flow battery," *Nano letters,,* vol. 14, no. 1, pp. 158-165, 2014.

[18] V. Mahanta, M. Raja, H. Khan, and R. J. J. o. T. E. S. Kothandaraman, "Drastic improvement in capacity-retention and polarization of vanadium redox flow battery with hydrophilic CO3O4

nanostructure modified activated graphite felt electrodes," *Journal of The Electrochemical Society* vol. 167, no. 16, p. 160504, 2020.
[19] N. Yun, J. J. Park, O. O. Park, K. B. Lee, and J. H. J. E. A. Yang, "Electrocatalytic effect of NiO nanoparticles evenly distributed on a graphite felt electrode for vanadium redox flow batteries," *Electrochim. Acta* vol. 278, pp. 226-235, 2018.
[20] J. Vázquez-Galván, C. Flox, J. Jervis, A. Jorge, P. Shearing, and J. J. C. Morante, "High-power nitrided TiO2 carbon felt as the negative electrode for all-vanadium redox flow batteries," *Carbon* vol. 148, pp. 91-104, 2019.
[21] L. Wei, T. Zhao, L. Zeng, Y. Zeng, and H. J. J. o. P. S. Jiang, "Highly catalytic and stabilized titanium nitride nanowire array-decorated graphite felt electrodes for all vanadium redox flow batteries," *Journal of Power Sources,* vol. 341, pp. 318-326, 2017.
[22] G. Wei, C. Jia, J. Liu, and C. Yan, "Carbon felt supported carbon nanotubes catalysts composite electrode for vanadium redox flow battery application," *Journal of Power Sources,* vol. 220, pp. 185-192, 2012/12/15/ 2012.
[23] A. Fetyan *et al.*, "Comparison of Electrospun Carbon−Carbon Composite and Commercial Felt for Their Activity and Electrolyte Utilization in Vanadium Redox Flow Batteries," *ChemElectroChem,* vol. 6, no. 1, pp. 130-135, 2019/01/02 2019.
[24] J. Xue *et al.*, "An all-cotton-derived, arbitrarily foldable, high-rate, electrochemical supercapacitor," *Physical Chemistry Chemical Physics,* 10.1039/C3CP51571K vol. 15, no. 21, pp. 8042-8045, 2013.
[25] A. Jain, R. Balasubramanian, and M. P. Srinivasan, "Hydrothermal conversion of biomass waste to activated carbon with high porosity: A review," *Chemical Engineering Journal,* vol. 283, pp. 789-805, 2016/01/01/ 2016.
[26] B. Zheng, N. Li, J. Yang, and J. Xi, "Waste cotton cloth derived carbon microtube textile: a robust and scalable interlayer for lithium-sulfur batteries," *Chem Commun (Camb),* vol. 55, no. 16, pp. 2289-2292, Feb 19 2019.
[27] M. W. Thielke, G. Tian, and A. J. J. J. o. P. M. Sobrido, "Sustainable electrodes for the next generation of redox flow batteries," *Journal of Physics: Materials,* vol. 5, no. 2, p. 024004, 2022.
[28] M. Ulaganathan *et al.*, "Bio-mass derived mesoporous carbon as superior electrode in all vanadium redox flow battery with multicouple reactions," *Journal of Power Sources,* vol. 274, pp. 846-850, 2015/01/15/ 2015.
[29] M. Park, J. Ryu, Y. Kim, J. J. E. Cho, and E. Science, "Corn protein-derived nitrogen-doped carbon materials with oxygen-rich functional groups: a highly efficient electrocatalyst for all-vanadium redox flow batteries," *Energy & Environmental Science,* vol. 7, no. 11, pp. 3727-3735, 2014.
[30] Z. H. Zhang, T. S. Zhao, B. F. Bai, L. Zeng, and L. Wei, "A highly active biomass-derived electrode for all vanadium redox flow batteries," *Electrochimica Acta,* vol. 248, pp. 197-205, 2017/09/10/ 2017.
[31] M. Jiao *et al.*, "Holey three-dimensional wood-based electrode for vanadium flow batteries," *Energy Storage Materials,* vol. 27, pp. 327-332, 2020/05/01/ 2020.
[32] R. Wang and Y. Li, "Twin-cocoon-derived self-standing nitrogen-oxygen-rich monolithic carbon material as the cost-effective electrode for redox flow batteries," *Journal of Power Sources,* vol. 421, pp. 139-146, 2019/05/01/ 2019.
[33] M. E. Lee *et al.*, "Silk Protein-Derived carbon fabric as an electrode with high Electro-Catalytic activity for All-Vanadium redox flow batteries," *Applied Surface Science,* vol. 567, p. 150810, 2021/11/30/ 2021.