

## COMMUNICATION

[View Article Online](#)  
[View Journal](#) | [View Issue](#)Cite this: *RSC Sustainability*, 2025, 3, 4451Received 23rd July 2025  
Accepted 5th August 2025

DOI: 10.1039/d5su00618j

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## Farm to cable: life cycle assessment of carbon electrocatalysts derived from coffee waste

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To realize the full potential of biomass waste-derived electrodes, their synthesis must be fully reproducible and their environmental life cycle impact must be assessed quantitatively. We now show how a systematic comparison of coffee waste as a source for direct hydrazine fuel cell anodes can inform the first full life cycle assessment (LCA) of this application. The result is a practical process yielding active and replicable materials, whose quantified environmental footprint points to actionable avenues for further development.

Although biomass contains enough carbon to replace fossil fuel as a chemical feedstock,<sup>1</sup> not all biomass-based processes are sustainable or able to yield reproducible products, given that the biomass feedstocks they treat vary in composition. Spent coffee waste, of which 6.5 million tons are produced globally each year,<sup>2</sup> is a case in point: although used as a source for solid carbon powders for purification,<sup>3</sup> supercapacitors,<sup>4</sup> batteries and fuel cells,<sup>5</sup> it remains a poorly defined substance. Coffee is sourced from many geographies and processed by many methods, resulting in coffee-derived carbons with broad distributions of structures, chemical compositions, and ecological footprints. After consumption, spent coffee grounds can be treated as a waste or valorized to economic products like fuel, building materials, or used agricultural amendments.<sup>6–8</sup> The treatment of spent coffee waste should be evaluated using a systematic environmental life cycle assessment (LCA) to understand preferred options for end-of-life (EoL) management. The reproducibility of biomass-derived carbons is especially important when designing fuel cell anodes, *e.g.* for

## Sustainability spotlight

Coffee ground waste and other biomass resources could be promising sources of carbon and nitrogen for manufacturing anode catalysts for future fuel cells. Coupled with earth-abundant iron, such FeNC catalysts could reduce the use of critical raw materials like platinum, which are currently the primary materials for electrocatalysts. However, biowaste is an unpredictable precursor for precise electrodes, with varied composition and structure. Here we show how careful biowaste selection, together with prospective life cycle assessment focused on climate change impacts, can guide the design of advanced FeNC electrodes for direct hydrazine fuel cells. This material sources the carbon from coffee waste, thus recycling the biogenic carbon in the waste compared with composting or land-filling. We conclude that manufacturing FeNC catalysts from coffee and more generally, other bio-organic waste, can make progress towards meeting UN Sustainable Development Goals of climate action (SDG13) and affordable and clean energy (SDG7).

oxidizing hydrazine hydrate ( $\text{N}_2\text{H}_4 \cdot \text{H}_2\text{O}$ ),<sup>9</sup> an energy dense ( $3.45 \text{ kWh L}^{-1}$ ),<sup>10</sup> safe and transportable alternative to anhydrous hydrazine and compressed  $\text{H}_2$  gas.<sup>11</sup> The hydrazine oxidation reaction ( $\text{HzOR}, \text{N}_2\text{H}_4 + 4\text{OH}^- \rightarrow \text{N}_2 + 4\text{H}_2\text{O} + 4\text{e}^-$ ) has the highest known theoretical cell potential in  $\text{O}_2$ -based fuel cells ( $1.56 \text{ V}$ , *viz.*  $1.23 \text{ V}$  for  $\text{H}_2$ ). However, the reaction overpotential strongly depends on the catalytic site, restricting the use of poorly reproducible biomass as a precursor.

Herein, we show how spent coffee waste obtained from a variety of sources can yield reproducible, high-value Fe- and N-doped carbon electrodes (FeNCs) for direct hydrazine fuel cell (DHFC) anodes. By performing a full cradle-to-gate LCA spanning from spent coffee waste collection to fuel cell utilization (Scheme 1), we quantify key environmental impacts of the process, compare them to commercial alternatives, conventional EoL treatments and outline actionable directions for further research and optimization to reduce global warming, toxicity and other environmental impacts. The case of coffee waste is therefore a model for all biowaste-to-electrode development processes. It highlights the importance of waste variety choice and the resulting environmental life cycle impact of the FeNC production process.

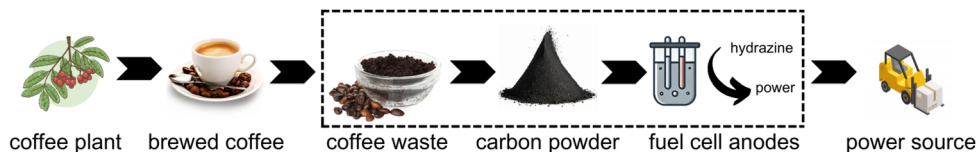
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Scheme 1 Production process and system boundaries (dotted line) of waste coffee-derived Fe–N–C electrocatalysts.

The FeNC powders were synthesized from six coffee waste samples collected from various sources in Israel (Table S1). The coffee waste samples were dried, ground and chemically activated by pyrolysis with  $\text{K}_2\text{CO}_3$  at  $800^\circ\text{C}$ .<sup>12</sup> These carbons ( $\text{NC}_{\text{source}}$ ) were loaded with iron salts and phenanthroline ligands, which provide most N to the catalyst, and heated to  $600^\circ\text{C}$ , yielding atomically dispersed Fe– $\text{N}_4$  moieties ( $\text{FeNC}_{\text{source}}$ ).<sup>13–15</sup> The carbons have multi-modal ('hierarchical') pore sizes, ranging from macropores ( $>50\text{ nm}$ ) to micropores ( $<2\text{ nm}$ ), as determined by scanning electron microscopy and  $\text{N}_2$  sorption (Fig. 1). Their BET-derived specific surface areas (SSAs) are high at  $1200\text{--}2200\text{ m}^2\text{ g}^{-1}$ , as are their electrochemical surface areas ( $80\text{--}230\text{ m}^2\text{ g}^{-1}$ , Fig. 1c). Both high surface area and multi-sized porosity enable fast flow of reactants to active sites during catalysis. The carbons are sufficiently graphitized, providing electronic conductivity, as testified by the Raman intensity ratio of disordered (D) and graphitic (G) bands, with  $I_{\text{D}}/I_{\text{G}}$  values typical for successful electrocatalysts (Fig. S1 and Table S2).<sup>16–18</sup>

The FeNC catalysts were electrocatalytically active towards hydrazine oxidation in  $1\text{ M KOH}$  (Fig. 2a), with onset potentials ranging between  $0.21$  and  $0.38\text{ V vs. RHE}$ .  $\text{FeNC}_{\text{Nescafe}}$  and  $\text{FeNC}_{\text{Nespresso}}$  showed earlier onsets than most known FeNC electrocatalysts for the HzOR ( $0.21\text{--}0.23\text{ V vs. RHE}$ ).<sup>19,20</sup> The synthesis of  $\text{FeNC}_{\text{Nescafe}}$  was repeated in five batches to yield  $0.2\text{ g}$  of  $\text{FeNC}_{\text{Nescafe-mixed}}$ . Its HzOR activity was compared to that of commercial FeNC from Pajarito Powder, USA ( $\text{FeNC}_{\text{Pajarito}}$ ),<sup>21</sup> showing similar performance in a lab-scale experiment (Fig. 2b). Moreover, when compared in a full platinum group

metal PGM-free direct hydrazine fuel cell,  $\text{FeNC}_{\text{Nescafe-mixed}}$  anodes yielded higher open circuit voltages (OCV), higher peak power densities, and higher limiting current densities relative to  $\text{FeNC}_{\text{Pajarito}}$  (Table S3).

To build on the promising electrocatalytic activity of the coffee-derived FeNC, as verified by fuel cell testing, LCA (ISO 14040/44)<sup>22,23</sup> was applied to evaluate the production of the coffee-derived  $\text{FeNC}_{\text{Nescafe-mixed}}$  electrocatalyst and to compare and benchmark its environmental performance with a functionally equivalent commercial electrocatalyst,  $\text{FeNC}_{\text{Pajarito}}$ , which relies on chemical sources of carbon and nitrogen (nicarbazin) rather than biomass. Both electrocatalysts were tested in a direct hydrazine fuel cell producing  $1000\text{ mW}$  power. A previous LCA study was reported for  $\text{FeNC}_{\text{Pajarito}}$  when used in an oxygen reducing PEM fuel cell cathode.<sup>24</sup> Another LCA exists for a biomass-derived FeNC used in Fischer–Tropsch thermal catalysis.<sup>25</sup> Using a cradle-to-gate system boundary, we evaluated three production scenarios: (i) lab-scale, as described above; (ii) optimized lab-scale, with improved coffee batch yield, oven energy efficiency, heating rates and atmosphere, and FeNC product yield relative to the lab-scale scenario; and (iii) pilot scale with high product yield. Life cycle inventories were built following laboratory scale-up methods<sup>26</sup> commonly used with laboratory data;<sup>27,28</sup> the pilot scenario assumed the usage of renewable ethanol solvent with solvent recovery by distillation.<sup>29,30</sup> To assess the environmental trade-offs of valorizing coffee waste into high-performance products, we also compared the treatment of  $1\text{ kg}$  of coffee waste *via* three conventional EoL options: landfilling with energy recovery; composting; and

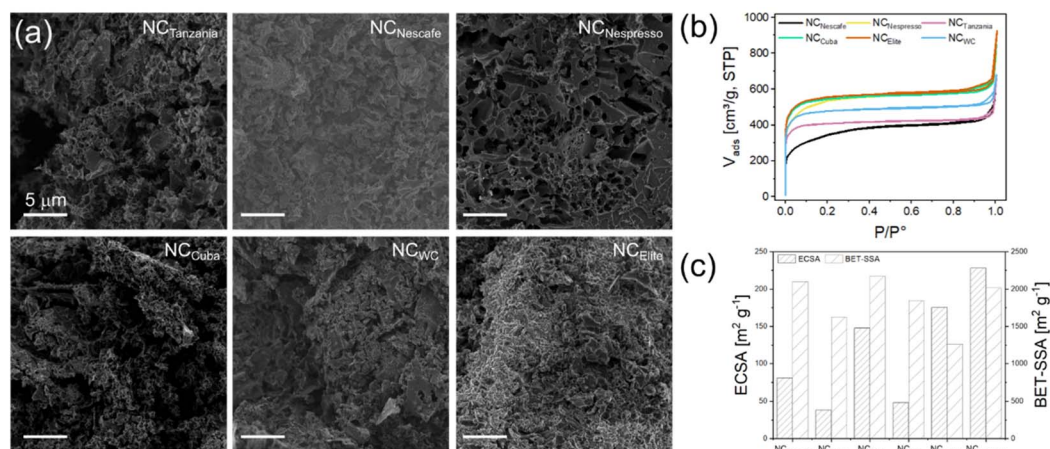


Fig. 1 (a) Characteristic scanning electron micrographs of the coffee-derived carbons. (b)  $\text{N}_2$  sorption isotherms at  $77\text{ K}$ . (c) Specific surface areas (SSAs-BET) and electrochemical surface areas (ECSAs) of the carbons.



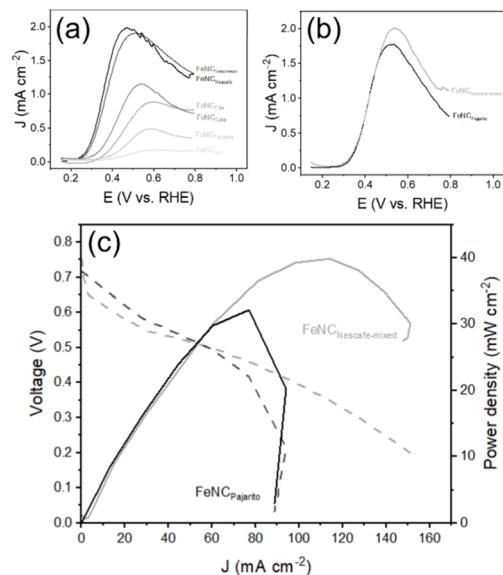


Fig. 2 (a) Linear sweep voltammograms of hydrazine oxidation on six coffee-waste-derived FeNC catalysts (20 mM N<sub>2</sub>H<sub>4</sub>, 1 M KOH, 10 mV s<sup>-1</sup>). (b) LSVs on FeNC<sub>Nescafe-mixed</sub> and commercial FeNC<sub>Pajarito</sub>. (c) Direct hydrazine fuel cell polarization curves with FeNC<sub>Nescafe-mixed</sub> or FeNC<sub>Pajarito</sub> anodes and a PGM-free cathode, at 80 °C (see the SI for details).

incineration with energy recovery, known as waste-to-energy (WtE), with the optimized scenario for producing the FeNC<sub>Nescafe-mixed</sub>. As of today, landfilling remains the primary EoL pathway for food waste globally. While in Europe, approximately 19% of municipal solid waste is composted, in the US, 6–9% of total municipal solid waste is currently composted.<sup>31,32</sup> Emissions from landfilling sites contribute significantly to GHG emissions, including CO<sub>2</sub> and methane (CH<sub>4</sub>), the latter possessing a global warming potential (GWP) of approximately 28 times that of CO<sub>2</sub> over a 100-year period.<sup>33</sup> These EoL options were also evaluated as avoided emissions during the pilot-scenario analysis, assuming 5 kg of coffee waste is diverted from landfill, composting, and waste-to-energy (WtE) processing. In the context of LCA, these avoided emissions are modeled as environmental credits (negative contributions to the global warming intensity, GWI), representing prevented emissions resulting from diverting waste from conventional EoL routes.

A life cycle inventory (LCI) model was developed to account for the collection, transportation and processing of coffee waste and its conversion into an electrocatalyst in the system boundary (Scheme 1) in Europe, with the functional unit defined as the catalyst quantity needed to generate 1000 mW power (per DoE milestones)<sup>34</sup> to operate a hydrazine fuel cell. Background data used in the LCI model were taken from the ecoinvent v3.5 (ref. 35) database and modeled using SimaPro software.<sup>36</sup> The life cycle impact assessment method used to compare the scenarios applied the ReCiPe 2016 midpoint hierarchist method.<sup>37</sup> The fraction of stable (sequestered) organic C was estimated following calculations on CO<sub>2</sub> offset in biochar.<sup>38</sup> For the EoL scenarios, LCI data for landfilling and composting were obtained from previously published work by

Forcina *et al.*,<sup>8</sup> Nordahl *et al.*,<sup>39</sup> and Mayer *et al.*,<sup>40</sup> respectively. The GWI results for the production scenarios of FeNC<sub>Nescafe-mixed</sub> and FeNC<sub>Pajarito</sub> are shown in Fig. 3. The lab-scale scenario for FeNC<sub>Nescafe-mixed</sub> has the highest GWI (12.8 kg CO<sub>2</sub>eq per 1000 mW) primarily due to significant consumption of electricity. Since the lab-scale process is a proof-of-concept to evaluate the electrocatalyst performance, there should be room for optimizing material efficiency and equipment usage at each step. Indeed, the optimized scenario improved results by a factor of 10.9, largely due to reduced electricity consumption, decreasing its process contribution from 96% to 43%. At higher scales, emissions from electrocatalyst production could decrease to 0.51 kg CO<sub>2</sub>eq per 1000 mW since the ethanol solvent can be produced from renewable resources<sup>41</sup> and recovered and recycled within the process. Markers in Fig. 3 indicate the net GWI, obtained by combining emissions from the catalyst production under the pilot scenario, with the emissions credited by avoiding the EoL treatment. They are negative for several scenarios, which is promising. Cradle-to-gate greenhouse gas emissions for ethanol could further decline by including bioenergy with carbon capture and sequestration. While here we assume electricity is supplied by the current average European electricity grid mix, which relies on burning a fraction of fossil fuels and emits 445 g CO<sub>2</sub>eq per kWh, the transition of future electricity grids to renewable sources could reduce electricity inputs to 6–46 g CO<sub>2</sub>eq per kWh (using wind power) or 9–167 g CO<sub>2</sub>eq per kWh (using photovoltaic cells).<sup>42–45</sup> Furthermore, scale-up of FeNC catalyst production would require moving to thermal rather than electrical energy supply, which potentially could use low-carbon biomass resources. Finally, future organic chemical feedstocks and solvents used in the synthesis of FeNC may be produced *via* low-carbon electro-catalytic conversion to further lower energy-related emissions.<sup>46</sup> Comparing the alternative anode electrocatalysts (Fig. 3), the global warming impact of the FeNC<sub>Pajarito</sub> production process is largely attributed to its high electricity

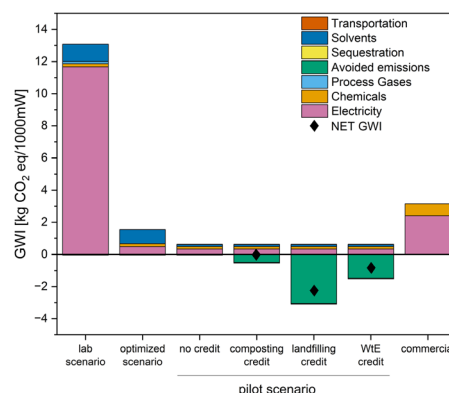


Fig. 3 GWI results for the production scenarios of lab scale FeNC<sub>Nescafe-mixed</sub> (lab scenario), optimized scale (optimized scenario), pilot scale scenarios include consideration of no credit and a credit for avoided GHG emissions from diverting the spent coffee waste from composting, landfilling, or WtE treatment systems, and FeNC<sub>Pajarito</sub> (commercial).



demand during synthesis.<sup>24</sup> Whereas production of **FeNC**<sub>Pajarito</sub> involves two energy-intensive high-temperature pyrolysis steps, its environmental impact is higher than **FeNC**<sub>Nescafe-mixed</sub> due to the high energy demand of the equipment used. Similarly, the production of **FeNC**<sub>Nescafe-mixed</sub> involves two energy-intensive pyrolysis steps with high energy consumption. Compared to **FeNC**<sub>Pajarito</sub> and other metal-based catalysts,<sup>47</sup> using coffee waste as a feedstock is environmentally beneficial, since some of the embodied C in the feedstock would otherwise decompose to CO<sub>2</sub> and CH<sub>4</sub> if landfilled or composted. Instead, the biogenic C in the coffee waste yields valuable electrocatalysts, while storing biogenic C in the **FeNC**, enhancing resource efficiency and reducing reliance on finite resources.<sup>48</sup> There is a declining trend across all ReCiPe categories (Table S13) in optimization scenarios (ii) and (iii) due to energy efficiency and improved product yield, even in relation to the commercial electrocatalyst benchmark. Importantly, the optimization yielded factors of 3–5 improvement in the global warming impact and in all toxicity parameters, when comparing the commercial powder to the pilot scenario. In addition, considering the upgrading and utilization (valorization) of byproducts generated in the process, such as oils, waxes, and gases, to biodiesel,<sup>49</sup> fine-chemicals,<sup>50</sup> and biopolymers<sup>51</sup> could further reduce the emissions of mid-point ReCiPe categories.

Fig. 4 reports the GWI for alternative treatments of spent coffee waste. The GWI values are expressed as emissions of CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O in CO<sub>2</sub> equivalents (CO<sub>2</sub>eq) according to the 100-year GWP of each gas, where 1 g of CH<sub>4</sub> and 1 g of N<sub>2</sub>O correspond to 28 g and 298 g CO<sub>2</sub>eq, respectively.<sup>33</sup> Each scenario in Fig. 4 is presented using disaggregated or aggregated impact, depending on data availability. For landfilling and composting, detailed emission inventories were available, allowing for individual contributions of CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O; however, only aggregated data were available for the WtE. Thus, these are grouped under “upgrading processes” in the figure. Regarding the **FeNC** production process, disaggregated GWI results can be found in Fig. 3. Regarding the EoL options, the environmental impact of emissions associated with landfilling is closely linked with emissions of methane during the degradation of the waste under anaerobic conditions. Furthermore, emissions of NO<sub>x</sub>,

CO, SO<sub>2</sub>, and ammonia from landfilling contribute significantly to other impacts such as acidification, air pollution, and human toxicity. For composting, most GHG emissions originate from emissions of N<sub>2</sub>O (~54%) and CH<sub>4</sub>. These emissions and the ratio between them are mainly controlled by the moisture content, C:N ratio, presence of bulking agents, temperature, pH, availability of oxygen, aeration techniques, *etc.*<sup>52</sup> Due to the fact that CO<sub>2</sub> emissions from upgrading the waste into electrode material, landfilling, and composting result from either thermal or natural organic decomposition of the coffee waste, biogenic CO<sub>2</sub> emissions are considered carbon neutral.<sup>53–55</sup> In all the landfilling, composting and WtE scenarios, CO<sub>2</sub> emissions result from transportation and operation during processing. GW impacts associated with WtE scenarios could vary significantly depending on assumptions regarding process efficiency, feedstock moisture content, calorific value, and how co-products are treated, whether by allocation or substitution (system expansion) applied to recovered energy.<sup>56,57</sup> Furthermore, if using consequential boundaries, an additional environmental burden would be assigned to each conventional EoL scenario for producing synthetic **FeNC**, Pt/C or other commercial electrocatalysts.

We conclude that alternative sources of coffee waste can be sequestered into functional **FeNC** electrocatalysts for a direct hydrazine fuel cell. Analyzing the life cycle environmental impact of these catalysts, relative to a commercial material, highlights the need for process optimization during scale-up, revealing benefits such as reducing landfill emissions, enhancing resource efficiency, and reducing the reliance on finite resources. Finally, we stress that any process developing biowaste into precise electrodes should take into account our findings regarding the coffee waste-derived electrocatalysts; that a precise and well-reported choice of waste precursor (type and region) has a huge effect on performance; and that a full LCA offers actionable directions for further development.

## Author contributions

STA initiated the collaboration and was responsible for conceptualizing the catalysts' research methodology, synthesized, tested, and characterized the catalysts (with SB), and with AS developed the LCA research methodology. AS performed life cycle assessment, SMZ performed fuel cell experiments (supervised by DRD), STA and AS wrote the original manuscript, and TKS analyzed oven power. SS and DE supervised the project and edited the manuscript.

## Conflicts of interest

There are no conflicts to declare.

## Data availability

The data supporting this article have been included as part of the SI. See DOI: <https://doi.org/10.1039/d5su00618j>.

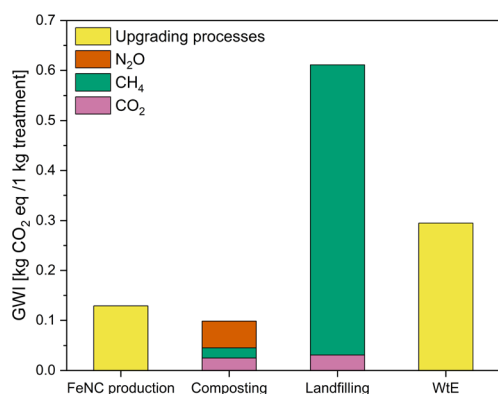


Fig. 4 GWI results for spent coffee waste EoL treatment alternatives benchmarked against **FeNC** pilot scale production.<sup>8,39,40</sup>



## Acknowledgements

We thank Assaf Henia and Noam Shalom (Z"l) of Blooms Coffee Truck and Lahav Bagrish for coffee waste samples, Dr Rachel Edrei and Dr Inna Zeltser (Technion) for material characterization. For partial funding, we thank the Grand Technion Energy Program, the Israel Science Foundation (1484/20), the Israel Ministry of National Infrastructure, Energy and Water (222-11-055, 222-11-064), and the Planning and Budgeting Committee (PBC) of the Council for Higher Education in Israel.

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