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Research Article

Life Cycle Assessment of Supercapacitor Electrodes Based on Activated Carbon from Coconut Shells

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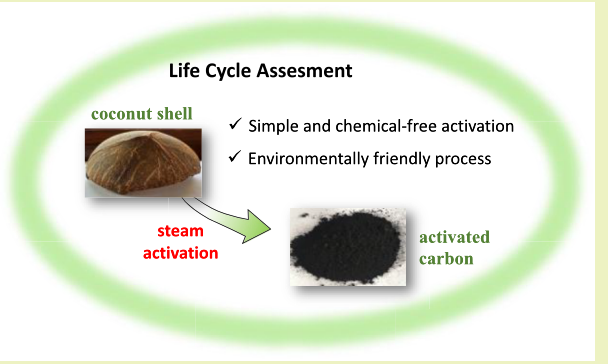
ABSTRACT: Coconut shells can be used to produce high-performance activated carbon (AC) electrodes for energy storage supercapacitors. An incentive to promote this manufacturing route is sought through its anticipated positive impact on the environment and the substitution of non-renewable resources. The present study sets out to assess the environmental performance of a recently developed route for AC manufacturing through a simple activation process using steam. The analysis was carried out using the life cycle assessment (LCA) approach to investigate the production of AC material and resulting electrodes for a broad range of environmental impact categories and energy use. The study was conducted for a hypothetical optimized industrial-scale scenario drawing on experimental observations, literature, and energy and material balance calculations. Impact assessment results were presented both for the functional unit of electrode's capacitance and mass of AC and electrode, and interpreted through comparative analyses with coal-derived AC, reduced graphene oxide, and algae-derived biochar aerogel electrodes. The impact assessment results of the new AC electrode show competitive performance across most of the investigated impact categories and indicators. Larger impacts are mostly only observed for the land and water use categories stemming from the agriculturally intensive practice of coconut production. A total of 5.68 kg of CO_2 and 34.4 MJ of CED kg^{-1} AC are reported, with AC constituting roughly 60% of the total impacts arising in the production of supercapacitor electrodes. The results have to be interpreted with present limitations to data especially considering the potentially high variability of carbon content in coconut shell species.



KEYWORDS: *Activated carbon, Supercapacitors, Electrodes, Ex-ante life cycle assessment, Coconut shells, Bio-based, Emerging technology*

INTRODUCTION

Energy storage systems are deemed essential to decoupling fossil resource use from energy supply and reducing greenhouse gas emissions.[1] In this context, energy storage systems must be cost-effective and built with a low impact on the environment using renewable materials. Recently developed electrodes using an activated carbon (AC) derived from coconut shells, a byproduct of coconut production in Sri Lanka, have a promising application as a bio-based sustainable alternative to fossil-based energy storage electrodes.[2] The new AC has been developed by means of thermal activation using steam as an activation agent to obtain highly porous electrode material with excellent energy storage capabilities.[2]

Environmental performance is seen as pivotal for the proliferation of the new technology given its application for energy storage while simultaneously providing a potentially economically viable solution to utilize agricultural byproducts. The functionalization of the new electrodes employing a simple activation using steam is seen as more environmentally friendly than the use of chemicals, which could create blockages in the pores of AC impeding the self-discharge capabilities, or require a removal, resulting in additional process steps and the generation of wastewater.[3] Moreover, insights into the environmental performance of the AC manufacturing route have a broader interest given the common use of AC for air and water purification, pollution abatement technologies,[4,5] and as an ingredient in personal cosmetics.[6]

The environmental performance of this new technology and its potential benefits could be validated using the life cycle assessment (LCA), a science-based multicriteria assessment method. Specifically, the application of LCA at the early stage of technology development is seen to have advantages for mitigating impacts in comparison to incumbent technologies

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given high design flexibility corresponding to high impact mitigation potential at the low technology readiness level.[7] LCA could be used to realize opportunities and limitations to improve the design of the new AC electrodes and show how they compare with alternative bio-based and fossil-based materials for energy storage.

Design optimization and environmental performance benchmarking of the novel electrodes have important value for energy storage development given limited research in this area. While AC is a common material for electrodes that could be derived from a broad range of bio-based feedstocks and has been researched over a relatively long time,[8−10] there have been very few LCA studies that explore the environmental performance of their use specifically in supercapacitors. Previous studies focused on the use of AC for water and air purification,[11−14] while the environmental assessment of carbon-based electrodes for supercapacitors involved only reduced graphene oxide (rGO)[15,16] and algae-based biochar aerogel (BA) materials.[17]

The present study evaluates the environmental performance of the new AC material and integrating electrodes by quantifying the impacts of their production for a range of impact assessment categories and indicators, and subsequently compares them with the carbon-based electrodes currently reported in the literature.[15,17] This is the first LCA study to evaluate the impacts of AC electrodes from coconut shells on behalf of their capacitance as their principal function in supercapacitors and is meant to provide an important baseline in future technology development and design of bio-based supercapacitors.

METHODS

The analysis has been carried out following recommendations and a framework developed by the International Organization for Standardization (ISO) involving four phases of assessment: goal and scope definition, life cycle inventory analysis, life cycle impact assessment, and interpretation.[18,19] The first two phases are described in the current section, and the last two phases constitute the Results and Discussion section.

Goal and Scope. *Goal Definition.* The present LCA aims to evaluate the environmental impacts of activated carbon (AC) produced from coconut shells and its application as the electrode material for supercapacitors. The purpose of the study is to support the development of new technology and provide a benchmark for future research in this area. Findings from this analysis are meant to benefit the researchers developing new electrodes and the field of renewable energy systems and energy storage in general.

Description of the Product System. AC electrodes evaluated in this study are developed for use in supercapacitors and correspond to the recently published fabrication procedure.[2] Coconut shells are first cleaned and dried in the oven at 100 °C to evaporate the moisture and are subsequently carbonized at 500 °C to convert the coconut shells into char. The char is then ground to 0.5−1.5 mm diameter particles and subjected to a two-step activation process using steam at 900 °C with an intermediate step of grinding to further reduce the particle size to 0.01−0.005 mm. The resulting AC powder is then mixed with carbon black, binder, and solvent to acquire the final paste and printed on the aluminum substrate, followed by annealing to obtain the final electrode. Specific capacitances of electrodes are experimentally measured and reported for two types of electrolytes. A single electrode capacitance of 219.4 F g^{-1} is achieved in 1-methyl-1-propyl-pyrrolidiniumbis(fluorosulfonyl)imide (MPPyFSI) ionic liquid electrolyte using the expanded graphite current collectors for the potential window of 3.5 V, and the capacitance value of 132.2 F g^{-1} is observed in the aqueous potassium hydroxide (KOH) electrolyte using the titanium current collectors for a potential window of 0.9 V.[2] These capacitance values are competitive in comparison to other production methods using coconut precursor, normally in a range 102−268 F g^{-1}.[2]

System Boundaries. The LCA is carried out as a cradle-to-gate analysis. The product system boundary includes all the processes for raw material acquisition and manufacturing of AC material and subsequent electrode fabrication, while potential impacts associated with the use of electrodes and the eventual disposal are not covered in the scope of this analysis. In order to provide a realistic evaluation with LCA, the model emulates process conditions at a prospective industrial level where the use of materials and energy is optimized.[20] This involves adapting system boundaries to exclude several materials or processes while some additional system design aspects are envisioned and added. The cut-off applies to the use of sandpaper for the preparation of coconut shells before cleaning and drying to remove debris. In industrial settings, the sandpaper would likely be substituted by a more resistant and nonabrasive surface grinder and these impacts are generally negligible.[21,22] Impacts of capital goods (e.g., transportation vehicles, machines, etc.) are also excluded since they cannot be adequately anticipated at the current technology maturity level, and are generally low.[23] Moreover, the assessment of electrodes encompasses only an active material component and excludes the impacts of the substrate. This decision was made in order to allow comparison with other electrodes that differ in terms of thickness and active material area mass loading or since their process descriptions do not report the information that is necessary to approximate the surface of the substrate. A similar decision was made in terms of the use of solvent. Dimethyl sulfoxide was initially used in the experiment to dissolve the polyvinylidene fluoride binder but with the aqueous suspension of polytetrafluoroethylene (PTFE) considered in the present model, the use of solvent can be avoided. The prospective system boundaries envision a recovery of energy from flue gases and tar from carbonization and activation processes, analogous to the design of AC production routes from coconut shells reported in the literature.[11,24] The production of AC electrodes is modeled for Sri Lanka, the source region of raw coconut shells.

Characterization and classification of environmental impacts were carried out for the ReCiPe Midpoint (H) life cycle impact assessment method and the indicator of cumulative energy demand (CED) using OpenLCA 1.10.3 open-access LCA software (GreenDelta, Berlin).[25] While all 18 categories of the method are reported, special attention was given to the global warming potential and CED given their important role for the environmental performance and energy efficiency of energy storage systems. The background inventory was populated using the ecoinvent database 3.6 cutoff version.[26,27] The foreground inventory data was derived from experimental work and adapted to resemble industrial-scale operations consistent with the modeling of emerging technology.[20]

New coconut-based AC and electrodes are compared with electrodes based on rGO and BA,[15,17] and commercial AC derived from coal, the current default AC data set of the ecoinvent database.[28] Comparisons with two electrodes are made separately considering that electrodes operate at different voltage conditions and in the context of measurement of their electrochemical properties. The comparison with rGO is made based on a capacitance achieved in ionic liquids at the working potential of 3.5 V, while the comparison with BA electrodes is based on their performance in KOH electrolyte at 0.9 V potential. The comparison with the coal-derived AC is made on a per-mass basis since the specific capacitance values for that material are not available. The current scenario does not take into account potential environmental credits for surplus energy created in the system that exceeds the energetic needs of the process itself and could be utilized in secondary applications. This decision was made amid uncertainties to whether this excess heat would be realistically utilized in other processes (i.e., in industry, office, or residential heating) and counterbalance the fact that some of data assumptions are based on theoretical efficiencies of energy recovery from gases and tar that might be more conservative in practice.

Function and Functional Unit. As a component in supercapacitors, the function of the electrodes is to store and release

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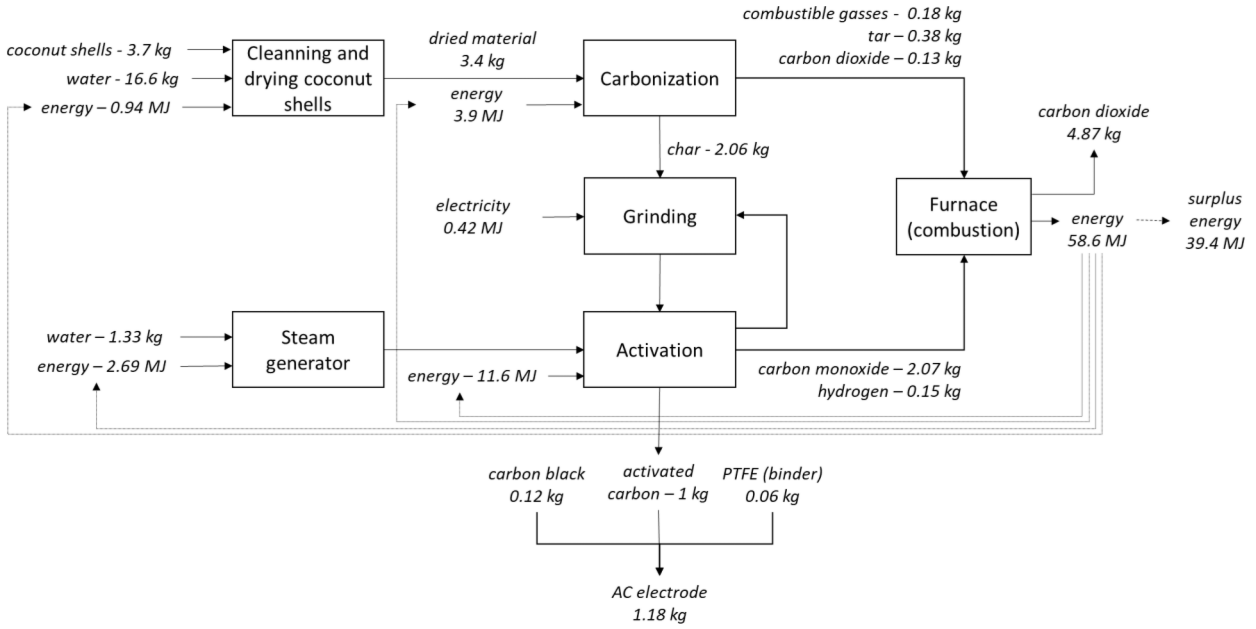

Figure 1. Process flowchart of AC material and electrode production.

electrical charge on demand. The scope of this function is described by the electrode's capacitance which is therefore taken as a functional proxy for the assessment. The functional unit (FU) was to achieve a specific capacitance of 1 Farad (F), which is consistent with the selection of FU in previous studies.[15,17] Given the specific capacitance in an aqueous KOH electrolyte (132.2 F g^{-1}) and specific capacitance in MPPyFSI ionic liquid (219.4 F g^{-1}), we calculated that 7.56 and 4.56 mg of active electrode material are necessary to achieve the capacitance of 1 F. The storage function of the supercapacitor over time is also affected by the electrodes' ability to preserve capacitance over multiple charge−discharge cycles (i.e., capacity fade).[16] However, the influence of these changes on functionality was neglected since AC is proven to have a relatively stable reversible capacity,[29] and given that a lifespan of integrating products (e.g., a car) normally precedes that of a supercapacitor.

Life Cycle Inventory. Data on materials, energy, and emissions arising in the production of AC was based on lab-scale measurements, secondary literature, and mass and energy balance calculations. These flows are approximated for industrial-scale operations consistent with the modeling of emerging technologies in LCA.[20] Primary data consists of material flow measurements of coconut shells, char, and resulting AC material while an inventory of the flue gases was based on anticipated elemental compositions of the raw coconut shells, char and AC using the secondary literature.[11,30] Industrial-scale energy approximations for the processes of drying, grinding and steam production were obtained by applying process engineering calculations and assumptions as detailed in Piccinno et al.[31]

The flows of materials, energy, and emissions in the production of AC material and electrode are depicted in Figure 1. The flowchart describes the foreground system showing the principal processes of cleaning and drying coconut shells, carbonization, grinding, and activation, and two supporting processes of generating the steam for activation and the capture and recovery of energy from flue gases and tar. For the sake of easier visual representation, the energy recovered in the furnace and CO_2 emissions from the combustion furnace are shown as aggregate, and the figure does not include emissions of water vapor that arise in processes of drying (0.3 kg), carbonization (0.6 kg), and combustion (1.6 kg). From the total of 4.87 kg of CO_2 emissions, 1.47 kg originate from carbonization (of which 0.13 kg are direct emissions from pyrolysis and 1.34 kg from burning combustible gases and tar), and 3.26 kg of CO_2 arise in the combustion of gases from activation. From 58.6 MJ of the energy recovered from the furnace, 38.9 MJ originate from gases fed from the process of activation, and 19.7 MJ from burning carbonization gases (5.9 MJ) and char (13.8 MJ). Detailed calculations underpinning each of the material and energy flows are disclosed in the Supporting Information (SI).

Production, Cleaning, and Drying of Coconut Shells. Impacts of coconut shells production are modeled using the ecoinvent data set "coconut production, dehusked". The impacts represented in the data set are partitioned between the coconut, coconut shell, and coconut husk using the economic allocation while considering the following approximate market values per tone of each byproduct: coconut, 550 €, coconut shell, 200 €, and coconut husk, 520 €. Factoring in the mass ratios between three fractions of the coconut and that around 6.7 kg of coconut is needed to produce 1 kg of coconut shells,[32] the ratio of environmental burdens from the production of virgin coconut allocated to coconut shells is estimated at around 6%. The energy for separating shells from other parts of the coconut is considered small and neglected. Cleaning of the coconut shells is carried out using water and sandpaper. Water is taken as measured in the laboratory and the use of sandpaper is excluded as explained earlier. The energy for drying coconut shells was calculated following recommendations from the literature considering the energy required to evaporate water at 80% of the drier's efficiency, as reported in Table S1.[31] The amount of moisture removed by drying was calculated by the difference in mass between wet and dried coconut shells.

Carbonization of Coconut Shell Powder. Environmentally relevant material and energy flows in the process of carbonization consist of energy use for the pyrolysis process and gaseous emissions created during the decomposition of cellulose, semicellulose, and lignin contained in coconut shells, leading to evaporation of water and partial oxidation of carbon.[33] The inventory for carbonization was based on experimental measurements of material flows of coconut shells and char while relying on secondary literature to approximate gaseous emissions. The experimental procedure reports a large conversion of dry raw coconut shells into char (around 61%) with the remainder consisting of volatiles in the form of flue gases, tar, and water vapor. The composition of volatiles was adopted from Arena et al. while the further breakdown of pyrolysis gases is adopted from Fagbemi et al.[11,30] The energy to heat the biomass was calculated and reported in Table S2, and the outputs of volatiles and gases are reported in Table S3.

Energy for the pyrolysis reaction was excluded from the study since the original composition of coconut shells is not known but previous studies show that these energy requirements are quite small.[34] For

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example, the energy necessary to pyrolyze oat hulls and pine varies between 0.2 and 1.6 MJ/kg, which is comparatively small with the energy needed to heat the reactor (in our case, 3.9 MJ) and potential energy that can be recovered from pyrolysis gases and tar (19.7 MJ).

Grinding. Grinding is employed before and after the first activation step. Grinding energy is estimated considering the initial and final size of the ground material, following recommendations for the grinding of soft ore (Table S4).[35] In the first grinding step, the particles of biochar are reduced from 40 to 1 mm. Grinding after the first activation step is carried out to further reduce the particle size to 7.5 μm and corresponds to the grinding of 75% of the original material mass (due to partial oxidation of carbon during the first activation process).

Activation. The activation is carried out by injecting a flow of steam into the chamber with char to increase the porosity and pore volume and eliminate impurities. The principal reaction that takes place is between char and superheated steam resulting in the generation of carbon monoxide and hydrogen: $C_{(s)} + H_2O_{(g)} \rightarrow CO_{(g)} + H_{2(g)}$. The carbon monoxide could further react with water vapor to create carbon dioxide and hydrogen following the reaction: $CO_{(g)} + H_2O_{(g)} \rightarrow CO_{2(g)} + H_{2(g)}$.[36] However, we assume that the process would be designed to minimize the second reaction so that carbon monoxide and hydrogen gases would be preserved, separated, and utilized as an energy fuel for heating the steam and other processes. The inputs of water and outputs of gases are calculated considering the amount of volatilized carbon material after the conversion of char into AC (roughly 50% as observed experimentally) while taking that the content of carbon would increase from 87% to 91% as observed in the literature.[11] Theoretical quantities of water had to be adopted since in the laboratory settings the steam is used in excess (149 L per kg of AC). However, it is plausible to assume that in practice, the water would be reused within the system. Inputs of 1.33 kg of water and outputs of 2.07 kg of carbon monoxide and 0.15 kg of hydrogen gases accompany the production of 1 kg of AC.

The energy demand for the activation process consists of the energy consumed by the activation reaction, the energy needed to heat the steam, the energy needed to heat the material to 900 °C, and the energy to compensate for the heat losses in the reactor. The material is assumed to be heated from the carbonization temperature (i.e., 500 °C), consistent with the design of a continuous industrial-scale setup. In this scenario, coconut shells are ground immediately after the carbonization step and fed into the activation reactor without cooling down between the carbonization and activation steps. The energy for raising the temperature of the reactor is excluded as the system is assumed to run in a continuous fashion. The activation reaction is endothermic consuming 9.73 MJ to produce 1 kg of AC. The energy required to generate the steam was calculated by considering the specific heat capacities of water and steam. The energy uses to produce the steam and maintain the temperature of the reactor are reported in Tables S5 and S6, respectively.

Combustion of Tar and Output Gases from Carbonization and Activation Processes. In the prospective scenario of AC production, the tar and combustible fraction of gases from pyrolysis and activation (hydrogen, methane, ethylene, and carbon monoxide) are burned to produce the energy to generate the steam and heat for the processes of drying, carbonization, and activation. Recoverable energy quantities are approximated by multiplying the mass of each of these fractions by their lower heating value (LHV) considering their reaction with oxygen and conversion into carbon dioxide and water. LHV represents the amount of heat released during the combustion of a substance, including the energy losses needed to vaporize the water. Combustion reactions, LHVs of tar and gases, energy quantities, and CO_2 emissions from carbonization and activation processes are reported in Tables S7 and S8, respectively. CO_2 emissions from burning the tar are approximated by taking the total calorific value of tar (36 MJ) and associated CO_2 emissions (0.083 kg CO_2 per 1 MJ of energy).

Manufacture of AC Material and Electrode. The final life cycle inventory for the AC is shown in Table 1, and the inventory of the integrating electrode is in Table 2. The inventory for AC electrode is provided for both 1 kg of electrode and reference flow of 1 F (as per the FU). The final inventory for AC reflects the notion that the heat energy used in most processes would be compensated within the system. A total of 19.1 MJ of energy needed in the system would be easily supplied by 58.6 MJ of the energy derived from the combustion of flue gases and tar from carbonization and activation processes. The observation that the system is energetically self-sustaining is consistent with the assumption made in previous works.[11]

Material inputs of AC, carbon black, and PTFE binder to produce the final electrode are measured experimentally. The carbon material is simply mixed with the additives and pressed onto the metallic substrate. The data set for carbon black is derived from the ecoinvent database and the data set for PTFE is from a recently published inventory.[37] Inventories of reduced graphene oxide (rGO) and biochar aerogel (BA) electrodes which are used for comparison are taken from previous studies[15,17] and detailed in Tables S9–S11. The data for rGO were based on a combination of process simulation software and GaBi database, while data for BA is derived from experimental observations and practical assumptions related to transport, harvesting of algae, and their conversion to aerogel.[15,17,38] The single electrode capacitance values are reported to be 83 F g^{-1} for rGO and 260 F g^{-1} for BA electrodes.

Table 1. Inventory for 1 kg of AC

input/output	value	unit
inputs		
coconut shells	3.65	kg
water (for cleaning and steam in activation)	17.90	kg
electricity (for grinding)	0.42	MJ
outputs		
carbon dioxide	4.87	kg
waste (organic from cleaning)	0.30	kg

Table 2. Inventory for the AC Electrode

input/output	for 1 kg of electrode	for 1 F	unit
inputs			
activated carbon	0.85	0.0221	kg
carbon black	0.10	0.0026	kg
PTFE	0.05	0.0013	kg

RESULTS AND DISCUSSION

Contribution Analysis of AC Material and Electrode. Impact assessment results of AC material and AC electrode are shown in Table 3. Additionally, the relative impact contributions between different components in the electrodes are shown in Figure 2. The synthesis of 1 kg of AC is associated with 34.4 MJ or cumulative energy use and 5.68 kg of CO_2 emissions. About 86% of CO_2 is generated during the production and activation of char, with the remaining 14% coming from coconut farming and electricity use for grinding. Relatively large impacts on the categories of land and water use attributed to the production of coconut shells are accompanied by relatively small eutrophication and acidification effect, which often times accompany agriculturally intensive production practices.

On the level of the electrode, AC dominates most of the categories with an average contribution of 60%. Secondary in the contribution are the impacts from the use of PTFE binder which is used in only a small amount (5% of the total mass of electrode) but is largely responsible for the impact in the categories of ionizing radiation and eutrophying emissions to freshwater ecosystems. The impacts of carbon black are small

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Table 3. Impact Assessment Results for AC and AC Electrode

category	1 kg of AC	1 kg of electrode	1 F electrode	unit
land use	1.93×10^{00}	1.65×10^{00}	7.54×10^{-03}	m^2a crop eq
marine eutrophication	4.54×10^{-03}	3.88×10^{-03}	1.77×10^{-05}	kg N eq
marine ecotoxicity	1.04×10^{-01}	1.30×10^{-01}	5.91×10^{-04}	kg 1.4-DCB
freshwater eutrophication	3.15×10^{-04}	5.50×10^{-04}	2.51×10^{-06}	kg P eq
stratospheric ozone depletion	7.39×10^{-06}	6.66×10^{-06}	3.04×10^{-08}	kg CFC11 eq
freshwater ecotoxicity	1.51×10^{-01}	1.60×10^{-01}	7.27×10^{-04}	kg 1.4-DCB
human noncarcinogenic toxicity	1.71×10^{00}	1.92×10^{00}	8.76×10^{-03}	kg 1.4-DCB
global warming	5.68×10^{00}	5.63×10^{00}	2.57×10^{-02}	kg CO_2 eq
ionizing radiation	1.64×10^{-02}	9.79×10^{-02}	4.46×10^{-04}	kBq Co-60 eq
fossil resource scarcity	1.32×10^{-01}	4.24×10^{-01}	1.93×10^{-03}	kg oil eq
fine particulate matter formation	2.09×10^{-03}	3.33×10^{-03}	1.52×10^{-05}	kg PM2.5 eq
ozone formation, human health	2.92×10^{-03}	4.04×10^{-03}	1.84×10^{-05}	kg NO_x eq
mineral resource scarcity	5.50×10^{-03}	7.57×10^{-03}	3.45×10^{-05}	kg Cu eq
human carcinogenic toxicity	3.72×10^{-02}	5.48×10^{-02}	2.50×10^{-04}	kg 1.4-DCB
terrestrial acidification	1.03×10^{-02}	1.19×10^{-02}	5.44×10^{-05}	kg SO_2 eq
terrestrial ecotoxicity	2.27×10^{00}	3.63×10^{00}	1.65×10^{-02}	kg 1.4-DCB
ozone formation, terrestrial ecosystems	3.11×10^{-03}	4.23×10^{-03}	1.93×10^{-05}	kg NO_x eq
water consumption	2.91×10^{-01}	2.57×10^{-01}	1.17×10^{-03}	m^3
cumulative energy demand	3.44×10^{01}	4.58×10^{01}	2.09×10^{-01}	MJ

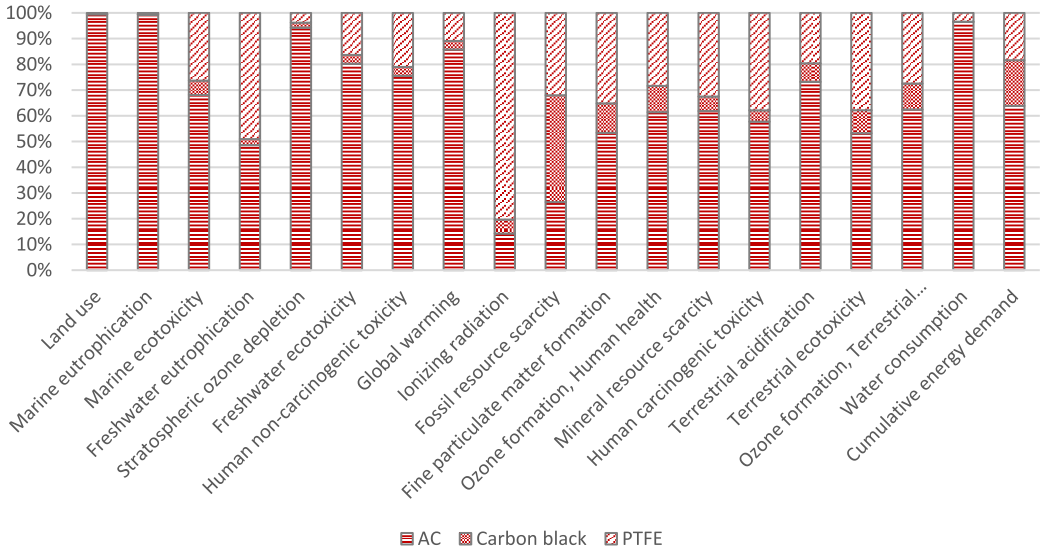

Figure 2. Contribution analysis of AC electrode.

on average, but notable in the categories of fossil resource scarcity and cumulative energy demand.

Comparison with rGO and BA Electrodes. The comparisons of the new AC electrode with the algae-based BA and the coal-derived rGO electrodes are shown in Figures 3 and 4, and the absolute impact assessment results are detailed in SI, Table S12. The comparison shows that the new AC electrodes are associated with significantly lower environmental impacts with a single trade-off to the marine eutrophication category when compared with the BA electrode. The better comparative performance is a result of less chemical-intensive fabrication and energetic neutrality of AC synthesis combined with relatively high specific capacitance.

Comparison with Coal-Derived AC and Other Bio-based AC. The comparison between the new AC and the coal-derived default ecoinvent AC is shown in Figure 5. The comparison is carried out using the proxy of mass without considering the functional performance of two ACs. Figure 5 demonstrates clear trade-offs among impacts arising in the production of new AC material versus the coal-derived alternative. The impacts of new AC are lower for half of the impact categories and the preference for AC material would come down to prioritizing specific impact categories. Using the new AC over coal-derived alternative would mean reducing ecotoxicity impact burdens on terrestrial ecosystems but increasing those burdens on the freshwater and marine ecosystems. Similarly, reducing the eutrophying impacts to the freshwater would increase their potential impacts on marine ecosystems. Using the new electrode material would also mean lowering cumulative energy use but in turn, emitting greenhouse gases with larger global warming potential.

Impacts on the global warming category (5.68 kg CO_2-eq) of AC explored in this study are generally low when compared on a per-mass basis with ACs based on fossil and other bio-based feedstocks reported in the literature (Figure 6). From ACs produced specifically for energy storage applications, Wang et al. report 62 kg CO_2-eq kg^{-1} for AC derived from

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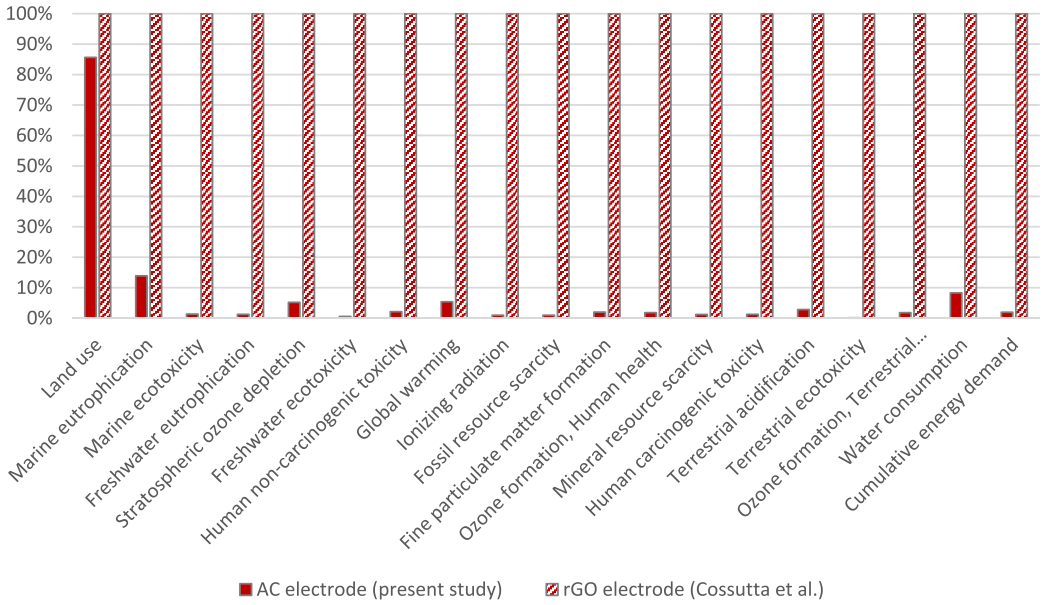

Figure 3. Relative comparison with rGO electrode.

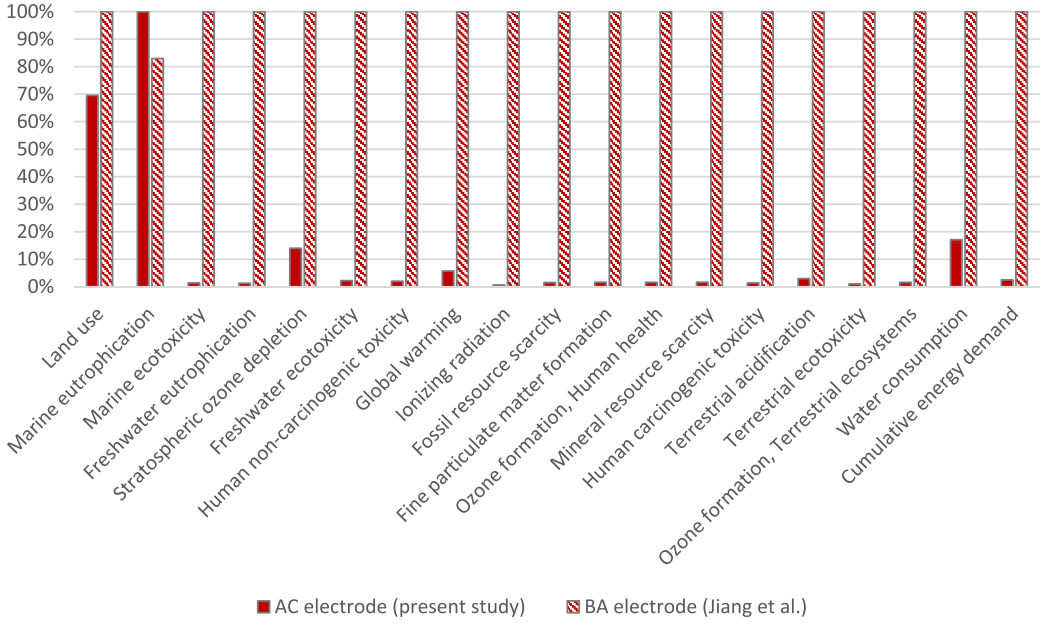

Figure 4. Relative comparison with BA electrode.

lignocellulosic biomass or 10.2−15.4 kg given specific design changes.[39] From other studies on AC production, Heidari et al. report 5.5−8.5 kg CO_2-eq emissions arising in the production from the eucalyptus tree by chemical activation.[40] The CO_2 equivalent emissions appear to be also lower than the hard coal and charcoal-derived AC of 7.8 and 5.8 kg CO_2-eq , respectively,[41] and 18.28 kg CO_2-eq reported elsewhere.[12] The impacts arising in the production of AC specifically for water treatment and pollution abatement are generally between 10 and 20 kg CO_2-eq kg^{-1} of AC,[12,13,28,42] but can be much higher when based on laboratory-scale data.[43] In terms of AC derived specifically from coconut shells, impacts are similar to those reported in Arena et al. (3.78−6.46 kg of direct CO_2 emissions depending on the scenario),[11] but higher than the 1.15 kg reported elsewhere.[24] Present AC electrodes report higher CO_2 emission equivalents when compared to AC derived from residual biomass −0.2−0.43 kg CO_2-eq ,[44] wood waste −0.01 kg CO_2-eq ,[24] and secondary AC recovery -2.45 kg CO_2-eq.[45]

CED is less commonly reported in previous studies, but appears to be generally larger than observed for AC electrodes in this study (34.4 MJ). Hjaila et al. report 167.63 MJ to produce 1 kg of AC from the olive waste cake and 17−51 MJ kg^{-1} AC from olive waste biomass.[13] A broad range of values are also reported for the AC from eucalyptus tree, 16 and 153.8 MJ, depending on the chemical activation route.[40]

However, it should be noted that a mass-based comparison among different AC materials has a limited value as it neglects the actual functionality of the material. While raw materials and synthesis techniques are generally applied to enhance physical characteristics such as surface area, pore volume, and morphological and crystalline properties, which are shared

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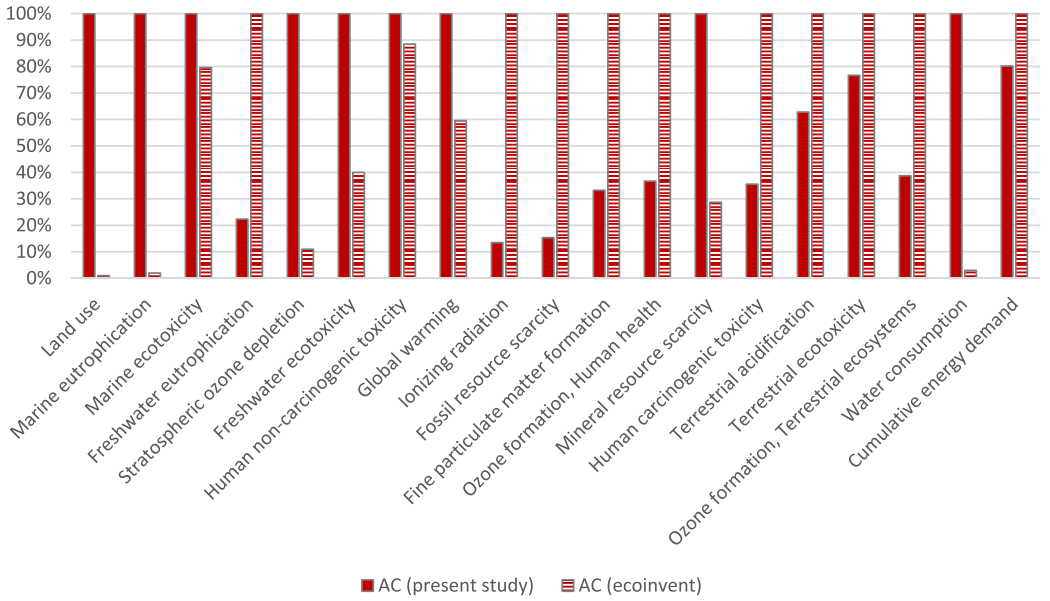

Figure 5. Relative comparison with coal-derived AC (default ecoinvent data set).

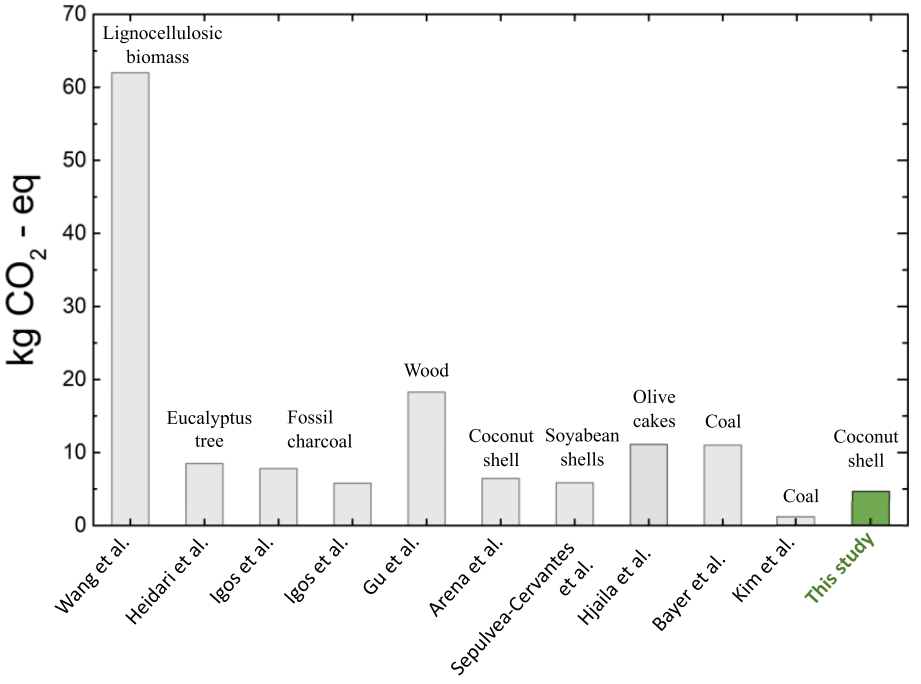

Figure 6. Greenhouse gas emissions for producing 1 kg AC reported in literature and the present study.

among different functional uses, the properties will differ between ACs used for energy storage and pollution abatement. Hence, the more plausible benchmark pertains to comparisons based on the electrochemical properties.

Implications to Technology Development and Future Assessment Opportunities Considering Data and System Boundary Limitations. A detailed breakdown of various energy and material flows presented in this study should benefit future process design and optimization of the AC manufacturing route. Technology developers and industry should design the system to reflect on the flows of material and gases and their composition to maximize a useful material output and ensure that the process is energetically self-sufficient. Since the most energy is consumed in the activation stage, it would be important to control the process to minimize the input of steam and reactivity of generated carbon monoxide in order to capture and utilize the energetically potent gas. The same applies to the carbonization process where changing process conditions would affect the composition and content of gases and tar.[30,46,47]

Results in this study need to be interpreted considering occasional data uncertainties arising from calculations and the use of secondary literature to estimate how much energy is used and available in the system. Data estimates are partially based on stichometry and theoretical efficiencies to calculate the inputs of water in the activation stage and the composition of energy-potent flue gases, their recovery and conversion into energy. Additionally, the current analysis relies on secondary

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literature to fill in the data gaps pertaining to the composition of coconut shells which may vary among different plant species. The choice of coconut plant may affect the content of cellulose, lignin, and moisture and the resulting quantities and composition of char, tar, and pyrolysis gases. According to the literature, the concentration of carbon in coconut shells can vary widely (27.2−53.5%),[48,49] which also reflects on a wide range of reported calorific values (17.39−30.4 MJ/kg).[50] In our case, the material recovery from carbonization is much higher (61%) in comparison to the literature (e.g., 26%),[11] which might suggest a higher concentration of carbon and lower concentration of water or higher stability of carbon in raw coconut shells.

While addressing those uncertainties would likely draw a more precise representation of energy flows in the system, they are unlikely to challenge the overall conclusion of energetic self-sufficiency drawn in this study given that a lot more energy can be recovered (58.6 MJ) than is consumed in the system (19.4 MJ). Hence, even if the practical conditions would somewhat deviate from the current scenario, resulting in higher energy use and more conservative recovery than presently estimated, the system is likely to remain energetically self-sufficient. In addition, the current observation of high energetic recovery potential from output flows of flue gases and tar in comparison to the potential energy derived from the direct combustion of coconut shells is in agreement with previous studies.[51] We observe that the surplus energy from the system (39.4 MJ) is only 35% lower than the energy that can be obtained by the direct burning of coconut shells (60.4 MJ, assuming LHV of 16584 kJ kg^{-1}).

Assessment of the environmental impacts of AC electrodes could be extended to account for the impacts arising during the device use and end-of-life phase, particularly in view of the specific capacitance and its effect on the size of the supercapacitor. The electrodes with lower specific capacitance would be bulkier given the larger size of the electrode and corresponding increase in size of other components in a supercapacitor, and may result in higher impacts in the use phase if the device weight is associated with energy usage (e.g., if the supercapacitor is used in a vehicle). Such shifting of environmental burdens between the production and use phase is commonly observed for the systems that use or create energy during their use phase.[52,53] If we are to consider the correlation between specific capacitance and weight of electrodes compared in this study, we would observe that the rGO electrode would be roughly 50% larger than the new AC electrode, while the AC electrode would be twice the size of the BA electrode. Therefore, the influence of other materials and the final applications of the supercapacitor has to be taken into consideration before making definite assertions. In addition, it is important to recognize that some of the aspects influencing any comparative preferences are not adequately captured in LCA but that might have important implications for sustainability or end-of-life management. For example, the use of algae as a naturally grown material for BA electrodes represents the true utilization of unwanted waste material that has value for a circular economy and sustainable resource management, but it has a limited influence on LCA results.[54]

Future assessments could also consider the possible indirect effects of diverting coconut shells from their alternative uses. The use of coconut shells in supercapacitor applications may result in increased market demand for their material substitutes in their conventional applications such as a fuel or a filler in concrete and composites, resulting in a positive or negative indirect consequence on the environment. An assessment of these indirect environmental impacts could be investigated using the consequential LCA approach.[55]

CONCLUSIONS

Energy storage systems built with low impacts on the environment are seen as one of the necessary conditions for alleviating the energy-induced climate crisis. According to the results of this LCA, electrodes based on AC derived from coconut shells through simple and chemical-free pyrolysis and activation process is a more eco-efficient substitute to a majority of ACs and carbon-based electrode materials reported in the literature. The study provides a detailed account of different energy and material flows to show that the manufacturing process could be designed in a way that is energetically self-sufficient, resulting in a material with low environmental impacts. Comparisons with electrodes based on rGO and biomass from algae show lower impacts of new electrodes in the majority of the environmental impact categories and cumulative energy use. Relative to the coal-based AC (a default ecoinvent data set), the new AC shows advantages for the terrestrial ecotoxicity and freshwater eutrophication impact categories and cumulative energy demand. Greenhouse gas emissions are also in a lower range in comparison to other coal-derived and bio-based ACs reported in literature.

The current study internalizes some of the data uncertainties characteristic of the analysis of emerging technology to approximate energy inputs and recovery in the system since the concentration of carbon and other elements in coconut shells and its derivates (i.e., char and AC) could vary among species of coconut and since these concentrations are not customarily measured when technology is developed in a laboratory. The environmental assessment of new electrodes could be extended to capture the indirect impacts of using coconut shells for energy storage electrodes and compare new material with other AC alternatives in the use and end-of-life of supercapacitors. But overall, the high energetic efficiency of the production process, which provides a value-added material from an agricultural byproduct, presents a strong argument for utilizing coconut shells as a raw material for supercapacitor electrodes.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acssuschemeng.2c03239.

Calculations and life cycle inventory of activated carbon (AC) material and electrode: Table S1, Energy for drying coconut shells; Table S2, Energy for carbonization; Table S3, Composition of volatile byproducts from carbonization; Table S4, Energy for grinding; Table S5, Energy for activation (steam); Table S6, Energy for activation (reactor); Table S7, Energy and CO_2 emissions from the combustion of flue gases and tar from carbonization; Table S8, Energy and CO_2 emissions produced from the combustion of gases from activation. Life cycle inventories and impact assessment results for electrode materials used for comparison: Table S9, Inventory for rGO; Table S10, Inventory for BA; Table S11, Inventory for rGO and BA

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electrodes; Table S12, Impact assessment values for rGO and BA electrodes, and coal-derived AC (PDF)

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Notes

The authors declare no competing financial interest.

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