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Environmental footprint of voltammetric sensors based on screen-printed electrodes: An assessment towards "green" sensor manufacturing

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Abstract

Voltammetric sensors based on screen-printed electrodes (SPEs) await diverse applications in environmental monitoring, food, agricultural and biomedical analysis, owing to their simple design, miniature size, and low cost. However, due to the single-use and disposable characteristics of SPEs and the scale of measurements these voltammetric sensors are used for, their environmental impacts should be considered. A life cycle assessment was conducted to evaluate the environmental footprint of SPEs manufactured using various substrate materials (SMs: cotton textile, HDPE plastic, Kraft paper, graphic paper, glass, and ceramic) and electrode materials (EMs: platinum, gold, silver, copper, carbon black, and carbon nanotubes (CNTs)). The greatest environmental impact was observed when cotton textile was used as SM. HDPE plastic

demonstrated the least impact (13 out of 19 categories), followed by ceramic, glass, and paper. However, considering the end-of-life scenarios and release of microplastics into the environment, ceramic, glass, or paper could be the most suitable options for SMs. Amongst the EMs, the replacement of metals, especially noble metals, by carbon-based EMs greatly reduces the environmental footprint of SPEs. Compared with other materials, carbon black was the least impactful on the environment. On the other hand, copper and waste-derived CNTs (WCNTs) showed low impacts except for terrestrial ecotoxicity and human toxicity (non-cancer) potentials. In comparison to commercial CNTs (CCNTs), WCNTs demonstrated lower environmental footprint and comparable voltammetric performance in heavy metal detections, justifying the substitution of CCNTs with WCNTs in commercial applications. In conclusion, a combination of carbon black or WCNTs EMs with ceramic, glass, or paper SMs represents the most environmentally friendly SPE configurations for voltammetric sensor arrangement.

Keywords: voltammetric sensor; screen-printed electrode; life cycle assessment; environmental footprint; climate change; carbon nanotubes

1. Introduction

The field of chemical sensing using voltammetric sensors have grown rapidly over the past decades as these types of sensors are suitable for on-site determination of target analytes with a portable, user-friendly, and field-deployable electrochemical instrumentation (Dhanjai et al., 2018; Holmes et al., 2019; Lisak et al., 2013; Kotani et al., 2019). Moreover, voltammetric sensors are widely adopted for the determination of analytes, such as metal ions, drugs, toxins, and pesticides in trace levels for environmental monitoring, food safety, agricultural, and biomedical analysis, owing to

their high sensitivity (Hayat & Marty, 2014; Lipskikh et al., 2018; Ciepiela et al., 2013; Gupta et al., 2011). Most of the voltammetric measuring setups employ a three-electrode system, comprising of a working electrode (WE), a reference electrode (RE), and a counter electrode (CE) (Jin et al., 2011; Tyszczuk-Rotko et al., 2019). Among the three electrodes, the WE is used as the principal electrode to conduct the electrochemical reactions, while the RE and CE are used to complete the electronic circuit and to measure or control either potential or current flowing in the system. In principle, voltammetric sensors register the change in the current between WE and CE owing to a redox reaction at the WE that involves the target analyte from the electrolyte, with the application of a controlled change of potential between WE and RE (Gupta et al., 2011; Scholz, 2015). For stripping voltammetry, an additional step of analyte preconcentration at the surface of the WE is performed, e.g. by electrodeposition of the target ion at the WE and its subsequent oxidation to register the current peak of preconcentrated analyte as the analytical signal for ion determination (Lovrić, 2005).

The electrochemical performance of voltammetric sensors is largely dependent on the type and quality of electroactive materials used to prepare them. In the past, liquid drop mercury electrode was considered as a suitable electrode material (EM) owing to its highly reproducible, renewable, and smooth surface (Choi et al., 2001). However, growing concerns about health hazard makes mercury electrodes inappropriate for on-site monitoring in the environment (Ahmad et al., 2020). With the advancements in electroanalytical science, various non-mercury materials, including noble metals (such as gold, silver, and platinum) and carbon materials (such as carbon black and various carbon nanomaterials) are widely used as EMs (Ariño et al., 2017; Dhanjai et al., 2018; Economou, 2018).

Screen-printing methodology offers the most promising approach to produce the new generation of voltammetric sensors (Metters et al., 2011; Lezi et al., 2012; Li et al., 2012). With an appropriate stencil design, the electrodes for voltammetric sensors based on screen printed electrodes (SPEs) can be fabricated by applying the paste consisting of electrode material through a patterned metal mesh. The screen-printing machine supports the mass production of disposable and single-use voltammetric sensors at low cost and high reproducibility (Morrin et al., 2003; Li et al., 2012). A typical design of the three-electrode system of SPEs, including WE, CE, and RE printed on the same substrate surface is shown in Figure 1. The use of such disposable SPEs for voltammetric sensors presents an attractive alternative to the conventional electrode substrates, which could significantly reduce the amount of sample required, avoid pre-treatment procedures (e.g. cleaning and polishing), and provide high flexibility with a wide range of EM configurations (Wang et al., 2001; Metters et al., 2011; Maczuga et al., 2013). With the rapid development of screen-printing technology, SPEs have become easier to manufacture and more economical. These advantages cause them to be commonly used as disposable electrodes (Medina-Plaza et al., 2015). Recently, SPEs have also been widely used in clinical applications, including early detection of microorganisms (Alonso-Lomillo et al., 2010), detection of antibiotics (Munteanu et al., 2018), and detection of drugs (Zhang et al., 2019). In addition, SPEs are a suitable platform for detection via electrochemical impedance spectroscopy (EIS). Numerous EIS sensors based on SPEs have been developed, including aptasensor for lysozyme detection (Rohrbach et al., 2012), impedimetric immunosensors for detection of anti-transglutaminase antibodies (Balkenhohl and Lisdat, 2007) and capsaicin sensor (Randviir et al., 2013).

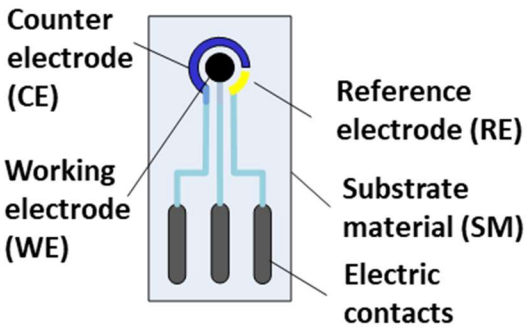


Figure 1. A typical design of disposable SPE based voltammetric sensors

As shown in Figure 1, the two major components in SPEs are substrate materials (SMs) and EMs. The SMs serve as the supporting platform for the electrode components of SPEs, thereby, accounting for the highest weight fraction of the SPEs in most cases. A number of chemical and physical properties, mainly including chemical inertness, non-conductivity, and temperature stability are generally required for selecting suitable SMs in SPE fabrications (Hayat & Marty, 2014; Metters & Banks, 2013; Altay et al., 2018). The typical SMs used in the SPEs are cotton textile (Xu et al., 2019; Xu et al., 2020), plastic (Jović et al., 2017), paper (Ferreira et al., 2018; Pungjunun et al., 2019), glass (Karuwan et al., 2017; Ghosale et al., 2018) and ceramic (Obaje et al., 2016; Adley, 2014).

For SPE based voltammetric sensors, silver is predominantly used to produce an inexpensive and stable Ag/AgCl RE, while carbon pastes are most commonly used to form the CE and WE, due to the low cost, low background currents, and broad potential windows (Wring et al., 1991; Morrin et al., 2003). In addition, silver is also widely used as electric contacts for SPEs, due to the superior conductivity. Significant enhancement in the sensor signal could be achieved using electrocatalytic materials to modify the surface of WEs with a reduced cost. A variety of metal nanoparticles (gold, silver, copper, bismuth, etc.), carbon materials (carbon nanotubes, carbon black, graphene, etc.),

or conductive polymers (polypyrrole, polythiophene, polyaniline, etc.) can be easily deposited on the surface of WEs to achieve a higher surface-to-volume ratio, superior conductivity, lower background current, improved adsorption properties, and enhanced signal intensities of voltammetric sensors (Rico et al., 2009; Bernalte et al., 2012; March et al., 2015; Waheed et al., 2018). Among carbonaceous nanomaterials, carbon nanotubes (CNTs) have attracted considerable attention in the scientific community, due to the outstanding properties in the electrochemical detection of various analytes (Jacobs et al., 2010; March et al., 2015; Arduini et al., 2016). Furthermore, CNTs can be enriched with functional groups (carboxylic, carbonyl, and hydroxyl) to improve their physicochemical properties and achieve favorable applications (Vaseashta & Dimova-Malinovska, 2005; Zhao et al., 2021). Besides, CNTs have been reported to be synthesized from waste resources (Wu et al., 2014; Veksha et al., 2020a; 2020b) and has been successfully demonstrated in applications including as electrode in electrocatalytic oxygen reduction reaction and as nano-adsorbent for enhanced heavy metals adsorption (As(V), Mn(VII), Pb(II) and Ni(II)) from industrial wastewaters (Moo et al., 2019; Egbosiuba et al., 2020; 2021).

Owing to the single use and disposable characteristics of SPEs and the scale of measurements employed using such type of sensors, the environmental impacts of SPEs should be considered. The selection of appropriate materials can impact the sustainability and environmental footprint of SPEs. Factually, the current environmental footprint of humankind is unsustainable in the context of limited natural resources and assimilation capacity (Hoekstra & Wiedmann, 2014). The usage of the limited resources is ever increasing, as we progress through the information age. SPEs are one such forthcoming application, where scarce resources are widely used. Life cycle assessment (LCA) determines the global environmental impacts of production, use, and disposal of goods and evaluate them from a systems perspective, identifying strategies for improvement without burden

shifting (Hellweg & Canals, 2014). The studies on LCA are increasing to aid in addressing the global sustainability and environmental challenges. Studies by Walser et al. (2011), Pourzahedi et al. (2017), and Pourzahedi & Eckelman (2015) reported that the major contributors to the environmental impacts were the production processes including metal mining (raw material extraction) and energy use. Moreover, the economic aspects of the sensors predominantly determine the shift to industrial production, while the environmental impacts are seldom evaluated. Hence, the objective of this study is to evaluate the environmental impacts of (i) the production of various material choices in SPEs application, including SMs (cotton textile, HDPE plastic, Kraft paper, graphic paper (recycled), glass, and ceramic) and EMs (platinum, gold, silver, copper, carbon black, commercial multi-walled CNTs (CCNTs), and waste-derived multi-walled CNTs (WCNTs)) and (ii) the end-of-life scenarios in order to investigate which combinations of SM and EM are the most eco-friendly. The study provides recommendations based on the projected environmental implications in 19 impact categories and qualitative end-of-life assessment. Furthermore, a comparison of the electrochemical sensing performance of CCNTs and WCNTs is included to validate the functionality of WCNTs in SPEs that is essential to affirm the EM suitability. Finally, the limitations and prospects of this study are discussed. Therefore, this study will identify the environmental impacts of a typical SPE and assist the research and development teams, industrial manufacturers, and decision-makers to adopt environmentally beneficial material choices prior to commercial product development.

2. Materials and methods

2.1. Life cycle assessment methodology

ISO 14040 standards were followed for the LCA study (ISO, 2006). The goal of the LCA is to compare various material choices of SPE components. The scope of the LCA includes all the unit

operations involved in the production of the materials, including resource extraction, energy consumption, and associated emissions. The functional unit of the study was fixed as 1 kg of the material. System boundary included the production process of each studied material. Life cycle inventory was used from the matching datasets in the ecoinvent3.5 database for the respective materials (Weber et al., 2018). GaBi professional software was utilized as the modelling platform and ReCiPe midpoint (H) impact assessment method was used to generate the results of the environmental impact indicators.

In this study, the assumption of a hypothetical scenario was disregarded to avoid the associated uncertainties. For instance, Weber et al. (2018) reported the LCA of vanadium redox flow batteries by developing hypothetical inventory models with data from various sources and involving numerous assumptions and simplifications of the model. This hypothetical scenario and assumptions made the uncertainty of the study to be high, and therefore interpretation of the results needed consideration as per the authors. The focus of this study was on the assessment of production process. According to many reported studies, the production process carried the highest burden in similar LCAs and represented the primary concern (Walser et al., 2011; Ahamed et al., 2021a; Pourzahedi et al., 2017; Pourzahedi & Eckelman, 2015).

The production process of the final product, including modification of the EM by nanomaterials, was excluded due to the variety of manufacturing methods employed for different materials and the non-availability of the process data from the manufacturers owing to private licenses. Additionally, one of the challenges in conducting LCA for the nanomaterials is the difficulty in quantifying the actual functional benefits due to improved material properties, extended lifespan of the products/materials, inconclusive toxicological effects, and avoided products/burdens. Direct substitution can be assumed for the conventional materials based on the functional unit, whereas

only anticipated benefits can be estimated for nanomaterials (Pourzahedi & Eckelman, 2015) increasing the uncertainties of the study. Further, the manufacturing process of the different materials also varies based on applications. The downstream processes, such as transport of the product to the end-user, and work involved in the application or use of the product are excluded, since they are either insignificant, non-specific to the use of the materials, or common for all the materials.

2.2. Chemicals and reagents

The following chemicals and reagents were used in the voltammetric measurements using SPEs incorporated with CCNTs and WCNTs. Analytical grade reagents and high purity solvents were used throughout the study. Sodium acetate, acetic acid (glacial, 100%), isopropanol and *N,N*-dimethylformamide (DMF) were purchased from Merck Pte. Ltd. (Singapore). The CCNTs (95%) were purchased from Nanografi Nano Technology. Deionized water (DI water) from a Millipore Milli-Q purification system was used in all the experiments. Acetate buffer solution (0.1 M, pH 4.5) was prepared by adjusting the different ratios of 0.1 M acetic acid and 0.1 M sodium acetate with a pH meter. Acetate buffer solution was used to dilute 1000 ppm standard solutions of Ag, Cu, Cd, Pb, and Hg from PerkinElmer Inc. (Waltham, MA, USA) to the concentration of 50 ppb.

The WCNTs were obtained from laboratory experiments reported by Veksha et al. (2020b). In brief, the synthesis of WCNTs was carried out in a vertical quartz reactor at 700 °C in N_2 atmosphere, where the calcined Ni-Ca catalyst (size 100-315 μm) was contacted with the non-condensable pyrolysis gas (50 mL min^{-1}). A plastic packaging waste mixture (53% polyethylene, 28.2% polypropylene, 11.8% polyethylene and the remaining fraction composed of polyaniline, inks, adhesives, and aluminum) was pyrolyzed at 550 °C, the pyrolysis vapors were catalytically reformed at 400 °C, and condensed to obtain the pyrolysis oil and the non-condensable pyrolysis

199 gas. The prepared WCNTs were purified by acid and water washing and dried overnight at 105
200 °C.

201 *2.3. Experimental preparation of WEs and electrochemical measurements*

202 The voltammetric sensors were prepared with disposable SPE chips (DRP-C110, Metrohm) by
203 modifying the WEs with CNTs. In the disposable SPE chips, a ceramic substrate was used for
204 electrode printing. WE and CE were printed with carbon paste, while RE and electric contacts
205 were printed with a silver paste. The process of WE modification was as follows. WCNTs or
206 CCNTs (5 mg) were added into a 10 mL mixture solution containing 8 mL DMF and 2 mL
207 isopropanol, and then sonicated for 10 minutes. The suspension (10 μL) was dropped on the WEs
208 of SPE chips, then the SPE chips were dried in the oven at 60 ℃ for 7 min. The SPEs modified
209 with WCNTs and CCNTs were labeled as WCNTs-SPCE and CCNTs-SPCE, respectively. The
210 electrochemical behavior of heavy metal ions on the prepared voltammetric sensors was studied
211 by differential pulse anodic stripping voltammetry (DPASV) using a handheld potentiostat (μStat
212 300 Bipotentiostat, Metrohm). Typically, 100 μL of sample solution was dropped onto the surface
213 of the SPE chip for the detection of heavy metal ions. The details of the developed testing protocols
214 are shown in Table 1.

215 Table 1. The detection protocols of heavy metal ions with prepared voltammetric sensors

Detection Protocols				
Method	DPASV			
Ions	Ag^{+}	Cu^{2+}	$Cd^{2+} + Pb^{2+}$	Hg^{2+}
Initial Potential (V)	−0.6	−0.5	−1.4	−0.1

Final Potential (V)	0.4	0.1	−0.5	0.5
Deposition Potential (V)	−0.5	−0.7	−1.2	−0.1
Deposition Time (s)	120	120	240	120
Equilibrium Time (s)	3	3	3	30
Time Pulse (ms)	0.008	80	0.005	0.002
Step Potential (V)	0.05	0.008	0.1	0.05
Pulse Potential (V)	80	0.01	80	80
Scan Rate (V/s)	0.04	0.02	0.01	0.04

3. Results and Discussion

The impact assessment results for the construction of the voltammetric sensors based on SPEs are presented for six SMs (including cotton textile, HDPE plastic, Kraft paper, graphic paper, glass, and ceramic) and for seven EMs (including platinum, gold, silver, copper, carbon black, CCNTs, and WCNTs). Although the application of these materials in SPEs are interchangeable, the production processes of the materials vary from agricultural land cultivation to mining and extraction. The comparison of the electrochemical performance of the WCNTs and CCNTs is provided, in order to justify the equivalent application. Subsequently, the end-of-life fate of all the SMs and EMs are discussed.

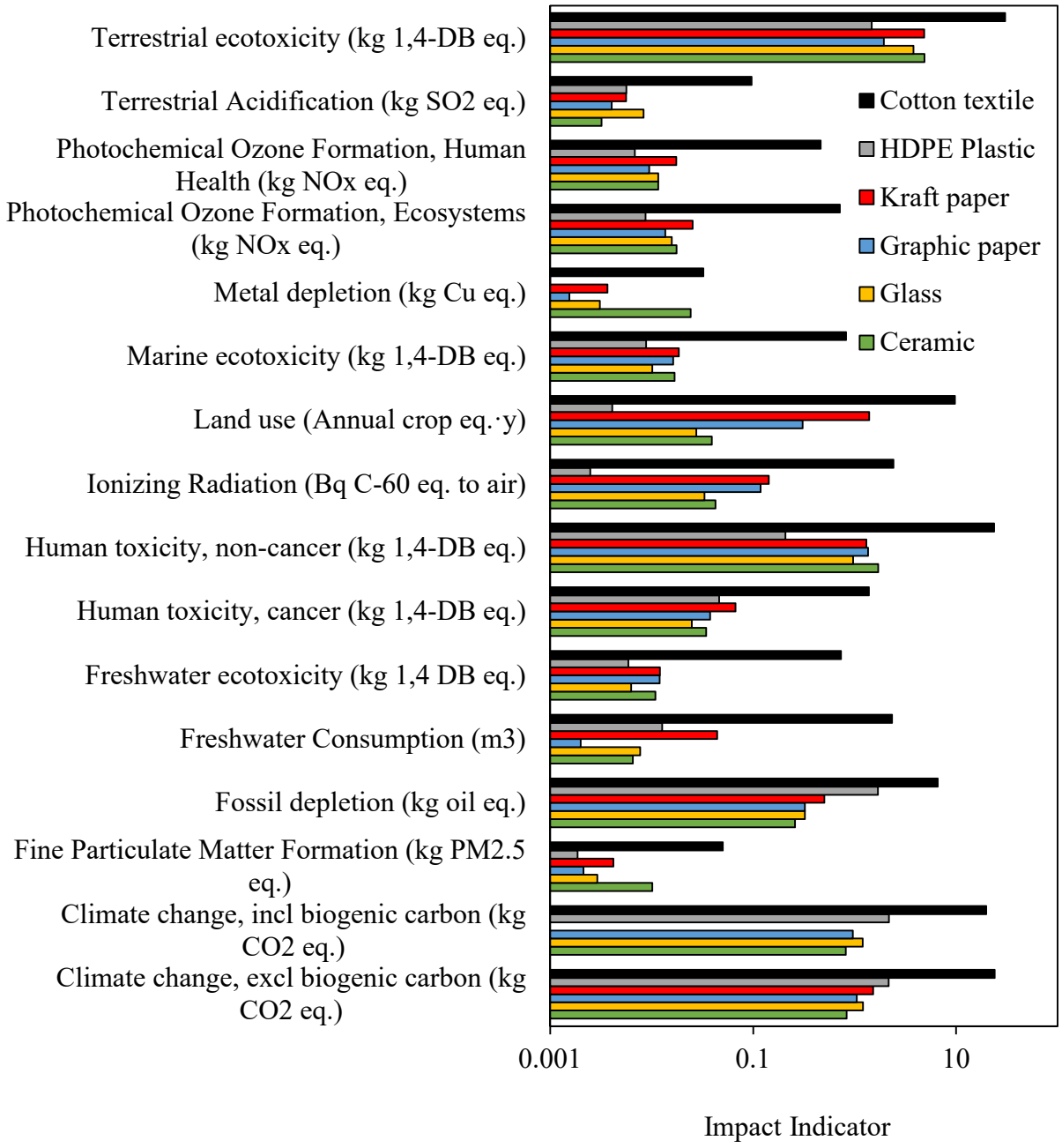
3.1. Impact assessment results

227 The nineteen impact indicators under the ReCiPe midpoint method was assessed for the SMs and
228 EMs. The following sections discuss the impact indicators that showed a significant effect on the
229 environment.

230 *3.1.1. Substrate materials for voltammetric sensors*

231 Figure 2 displays the impact assessment result for the production process of cotton textile, HDPE
232 plastic, Kraft paper, graphic paper, glass, and ceramic. Among the SMs, cotton textile
233 demonstrated the highest environmental implications for all the impact categories, when compared
234 to the other SMs. The causes are associated with the elaborate production process involving
235 intensive cultivation and allied manufacturing activities. Due to its superior toughness, comfort
236 and tight conformal contact with the skin, the cotton textile is particularly suitable to be used as a
237 SM for the electrodes in various wearable devices, which can be fabricated by a screen printer with
238 the similar procedures for the other SMs (Xu et al., 2019; Xu et al., 2020). The primary contributor
239 is the inorganic emissions to air that accounted for over 85 and 105% of the impact scores for
240 climate change potentials – excluding and including biogenic carbon, respectively. An offset of
241 24% was observed due to the biogenic sources. In all the SMs, the consumption of non-renewable
242 energy resources contributes entirely to the fossil depletion potential. It can be overcome by opting
243 for renewable energy resources throughout the upstream production stages. Emissions to
244 freshwater in cotton textile production accounted for 94 and 86% of the contribution in the case of
245 human toxicity - cancer and non-cancer, respectively. The cause of terrestrial ecotoxicity was the
246 heavy metals emission to air that amounted to approximately 90% of the overall indicator score.
247 These emissions are associated with the use of fertilizers and pesticides (Ahamed et al., 2021a).
248 HDPE plastic showed the second highest implications for climate change and fossil depletion
249 categories due to the use of fossil fuels for the material extraction and manufacturing processes.

Kraft paper showed the third highest climate change potential (excluding biogenic carbon) of 1.5 kg-CO_2 eq., while in the case of climate change potential (including biogenic carbon) the implications were positive (−0.52 kg-CO_2 eq.) to the environment. This can be attributed to the biological origin of the paper that absorbs CO_2 from the atmosphere, however, sustainable production practices need to be followed to accrue the benefits. In the case of human toxicity – cancer and non-cancer impact categories, Kraft paper and ceramic demonstrated the second highest implications, respectively, while both reported a similar terrestrial ecotoxic effects.



257

258 Figure 2. The impact assessment results of the major impact categories for the production

259 process of the substrate materials for voltammetric sensors. Freshwater eutrophication, marine

260 eutrophication, and stratospheric ozone depletion potentials were below 0.001 for all the SMs.

261

In the impact categories of freshwater consumption, ionizing radiation, and land use change, cotton textile showed the highest impacts because of the requirement of large areas of land for cultivation and voluminous consumption of water, unlike the other five materials. The second highest impact was observed in Kraft and graphic papers in the impact categories of ionizing radiation and land use. The ionizing radiation effects are associated with the material consumption such as fertilizer for cultivation and fuel for the processing stages. The land use reflects the requirement of land area for the crop cultivation and timber production. Furthermore, Kraft paper showed the second highest freshwater consumption, while recycled graphic paper showed the least due to the avoidance of timber cultivation and primary production processes. Compared to paper, the effects of plastic, glass, and ceramic were minor in these impact categories.

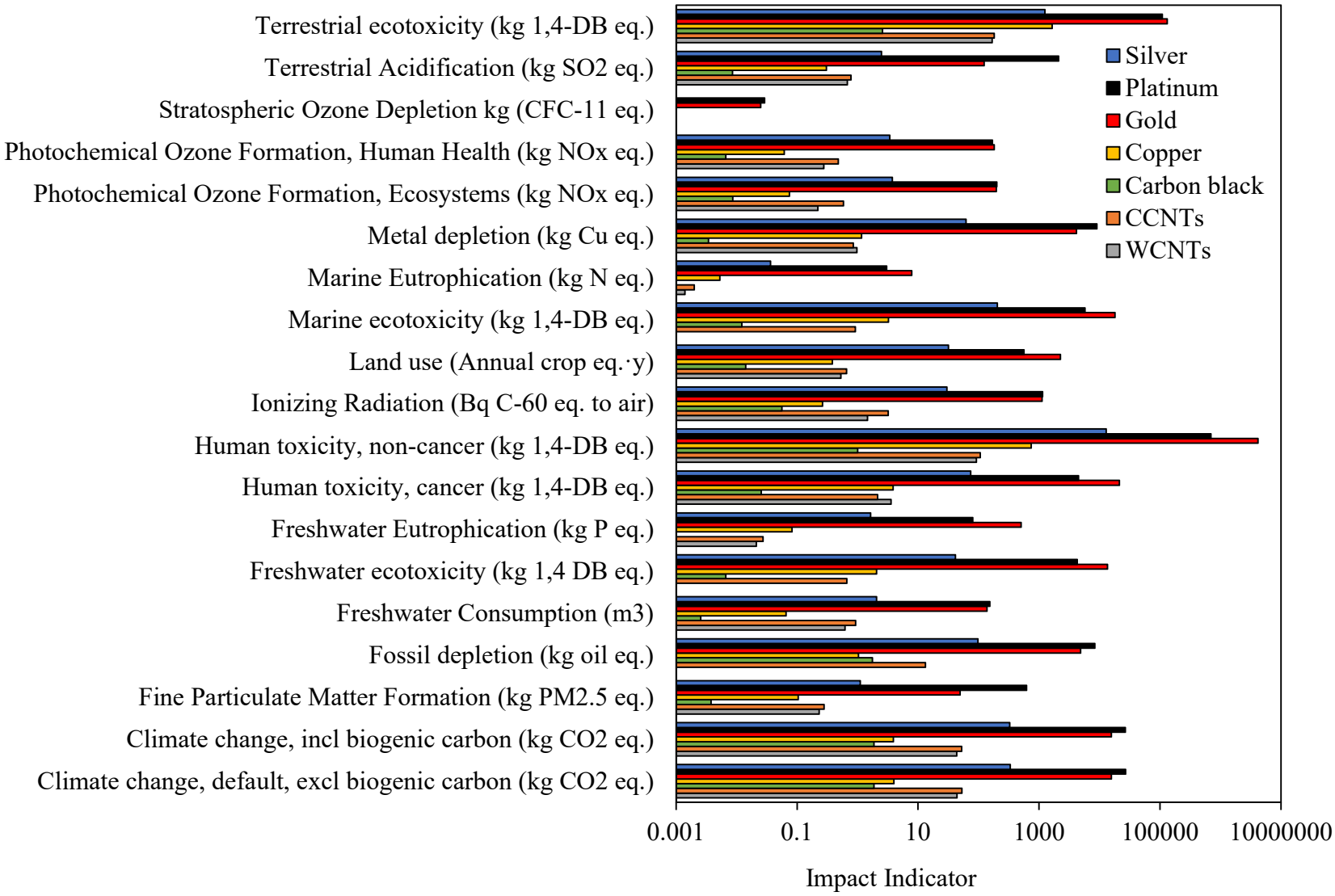
The remaining impact categories demonstrated similar trends in the environmental implications with cotton textile contributing to the highest in all the cases. Ceramic exhibited notable metal depletion (0.0242 Kg Cu eq.) and fine particulate matter formation (0.0102 Kg $PM_{2.5}$ eq.) potentials. From the overall results, HDPE plastic exhibited the lowest implications in 13 out of the 19 impact categories evaluated, while the remaining materials showed minor implications when compared to cotton textile. Hence, among the studied SMs, plastic carry an environmental advantage based on the upstream production process. The order of preference from this assessment is plastic > ceramic/glass > paper > cotton. In current practice, ceramic is the predominantly used material for the SMs in commercial SPEs. The use of plastic, ceramic, glass, and paper can be interchangeable, depending on the specific requirements in the application of SPEs. For instance, plastic or paper SMs could be used in applications that require flexible SPEs instead of ceramic or glass and vice versa.

3.1.2. Electrode materials for voltammetric sensors

Figure 3 presents the environmental implications of EMs in the 19 impact categories under the ReCiPe midpoint method. Among all the EMs, platinum exhibited the highest implications in the impact categories - climate change (excluding (27017 kg CO_2 eq.) and including (26839 kg CO_2 eq.) biogenic carbon), metal (9072 kg Cu eq.) and fossil depletion (8408 kg oil eq.), terrestrial acidification (2113 kg SO_2 eq.), and fine particulate matter formation (625 kg $PM_{2.5}$ eq.) potentials followed by gold. The effects are due to the inorganic emissions to air and use of non-renewable energy resources and elements during the production process. Gold exhibited the highest implications in the human toxicity (cancer and non-cancer (21328 and 4167907 kg 1,4-DB eq., respectively)), freshwater, marine, and terrestrial ecotoxicities (13521, 18079, and 132101 kg 1,4-DB eq., respectively), freshwater (505 kg P eq.) and marine eutrophications (8 kg N eq.), and land use (2254 annual crop eq.·y). The implications are attributed to the long-term inorganic and heavy metal releases to freshwater, and heavy metal emissions to air. Gold content in the ore is usually low. It requires enormous quantities of ore to be mined and processed to yield one unit of metal, making it one of the most resource intensive materials (Mudd, 2007). The largest contributor to ecotoxicity was related to environmental releases from bulk processing, including the disposal of sulfidic tailings from mining and refining processes (Pourzahedi & Eckelman, 2015). Ionizing radiation, freshwater consumption, photochemical ozone formation (ecosystem and human health), and stratospheric ozone depletion potentials had similar contributions from gold and platinum. Silver exhibited comparatively limited effects due to the greater availability and higher content in ore. The stratospheric ozone depletion and marine eutrophication effects were minimal for all the materials. Among the metals, copper presented the lowest implications in all the impact categories attributed to the relative abundance of the material. Apart from human toxicity (non-

cancer) and terrestrial ecotoxicity (747 and 1645 kg 1,4-DB eq., respectively), the remaining impact categories displayed minimal implications. However, copper metal is not as resistant to oxidation and other chemical interactions as the other EMs discussed, has exhibited considerable leaching potential (Slijepcevic et al., 2021), hazard risk and toxicity potential (Muhammad et al., 2020), which diminishes its scope of application in SPEs. Hence, metal-free materials should be considered for SPEs that could come in the various forms of carbon.

The metal-free materials, particularly the carbon-based materials possess some attractive characteristics, including moderately high porosities, large specific surface areas, and excellent electrical conductivities for construction of voltammetric sensors (Pistone & Espro, 2020; Kalinke et al., 2019). Although noble metals (e.g. gold, silver, or platinum) could offer better performance in some cases, the high cost and insufficient resources limit their applications in fields requiring mass or large-scale production. Furthermore, the carbon-based materials can be decorated with noble metal modifier(s) (e.g. metal or metal oxide nanoparticles) to further improve the performance of voltammetric sensors for various applications (Xiang et al., 2018).



322

323 Figure 3. Impact assessment results of all the impact categories for the production process of electrode materials.

Carbon black exhibited the lowest implications in all the impact categories except fossil depletion (1.8 kg oil eq.) among the EMs. Notably, WCNTs demonstrated a positive effect on the environment in fossil depletion (−22.13 kg oil eq.), freshwater (−18.21 kg 1,4-DB eq.) and marine (−21.64 kg 1,4-DB eq.) ecotoxicities (not shown in Figure 3). The positive effects are associated with the greater offsetting effect of the byproduct (pyrolysis oil) than the resource consumption during the WCNTs production process. Detailed production process and environmental impact assessment of WCNTs production was reported in Veksha et al. (2020b) and Ahamed et al. (2020). The CCNTs reported ca 50% higher implications on an average in all the impact categories excluding stratospheric ozone depletion when compared to WCNTs. Furthermore, the electrochemical performance of WCNTs in comparison with the CCNTs are reported in Section 3.1.3 to justify the material choice as replacement. Hence, carbon black followed by WCNTs is recommended as the EMs for SPE applications. More specifically, carbon-based materials are applicable for the WE in SPEs. On the other hand, Ag is an essential material for REs in most cases, except where the application of pseudo-RE is possible. However, WE and CE could prioritize carbon-based electrodes to minimize the use of metals where applicable.

Moreover, the metals are finite resources that eventually deplete over the long term especially gold and platinum. Arvidsson & Sanden (2017) listed fourteen metals as scarce including the European Union's list of critical metals, the United States' list of conflict materials, and those extracted from high-grade ores after screening sixty elements from the periodic table. Gold, platinum and silver, all of which are commonly used in SPEs, were included under these fourteen metals. Resource depletion-driven scarcity can become permanent and pose a potential threat to modern civilization (Söderholm & Tilton, 2012). Hence, avoidance of scarce resources at the early stage of a product development is crucial for progress towards the sustainable development goals. On the other hand,

carbon-based materials can be categorized as a comparatively infinite resource, if sustainable practices are adopted.

3.1.3. Comparison of electrochemical performance of CCNTs and WCNTs

Although the WCNTs were found to be more environmentally friendly than the CCNTs, their electrochemical performance needs to be substantiated. A consistent performance is essential to ensure favorable policy measures in material choices (Söderholm & Tilton, 2012). Heavy metal detections are of vital importance in environmental quality monitoring, owing to the toxic and non-biodegradable nature (Ding et al., 2021; Zhou et al., 2020a; Khan et al., 2020; Kong et al., 2020; Liao et al., 2021). Therefore, voltammetric sensing using SPEs were conducted for heavy metal detection to evaluate the use of WCNTs as a substitute for CCNTs. Interestingly, compared to CCNTs-SPCE, WCNTs-SPCE achieved superior signal intensity as well as lower background current for the heavy metal ions tested, i.e., Ag, Cu, Cd, Pb, and Hg (Figure 4). The possible reasons are that the modification with WCNTs made a smooth WE surface with a higher surface-to-volume ratio, boosting electron transfer kinetics, adsorption properties, and signal intensities of the voltammetric sensors. The CCNTs aggregate and are inhomogeneous when compared to WCNTs, which resulted in higher background current and unstable response signals of the voltammetric sensors. Hence, the WCNTs exhibited viable electrochemical performance and demonstrated potential to replace the CCNTs. Furthermore, the material characterization of CCNTs and WCNTs using X-ray photoelectron spectroscopy (XPS), Fourier transform infrared spectroscopy (FTIR), and field emission scanning electron microscopy (FESEM) coupled with energy dispersive spectroscopy (EDS) are presented in the Supplementary information. The XPS spectra revealed the elemental composition (at.%) from the surfaces of the two CNTs as follows, CCNTs: 98.61% C; 1.31% O; 0.08% Si and WCNTs: 98.97% C and 1.03% O (Figure S1), which

indicated that both CCNTs and WCNTs are almost free of impurities. Similarly, the EDS maps also showed the composition of CCNTs: 98.5% C, 0.9% Fe and 0.4% Al and WCNTs: 99.4% C, 0.2% O and 0.2% S (Figure S3). Furthermore, the XRD spectra (Figure S4) confirmed the lack of impurities. Therefore, according to the XPS, EDS and XRD spectrum, the purity of WCNTs is comparable to or even higher than the purity of CCNTs. The FTIR profiles depicted the presence of C=C, C-H, C-O, C=O, C-N and O-H characteristic peaks in both the CNTs (Figure S2). The XPS and FTIR spectra of the CCNTs and WCNTs were analogous with the presence of similar elemental composition and functional groups. The FESEM micrographs presented the surface morphologies with the average sizes of CCNTs (10-30 nm) and WCNTs (30-50 nm), while the FESEM-EDS mapping indicated the predominance of carbon (ca 99 wt.%) and trace level of impurities in the CNTs (Figure S3). Additionally, the cyclic voltammograms (CVs) and electrochemical active surface area (ECSA) were measured, and the graphs are provided in Figures S5 and S6. The CVs were comparable between CCNTs and WCNTs, while the calculated ECSA of WCNTs and CCNTs were 3.41 and 6.14 μF/cm. Although the ECSA was of the same order of magnitude, the minor difference in the ECSAs can be attributed to the different size ranges between the CNTs as inferred from the FESEM images. These results suggest that the WCNTs are comparable, effective and environmentally friendly substitute of CCNTs from fossil resources.

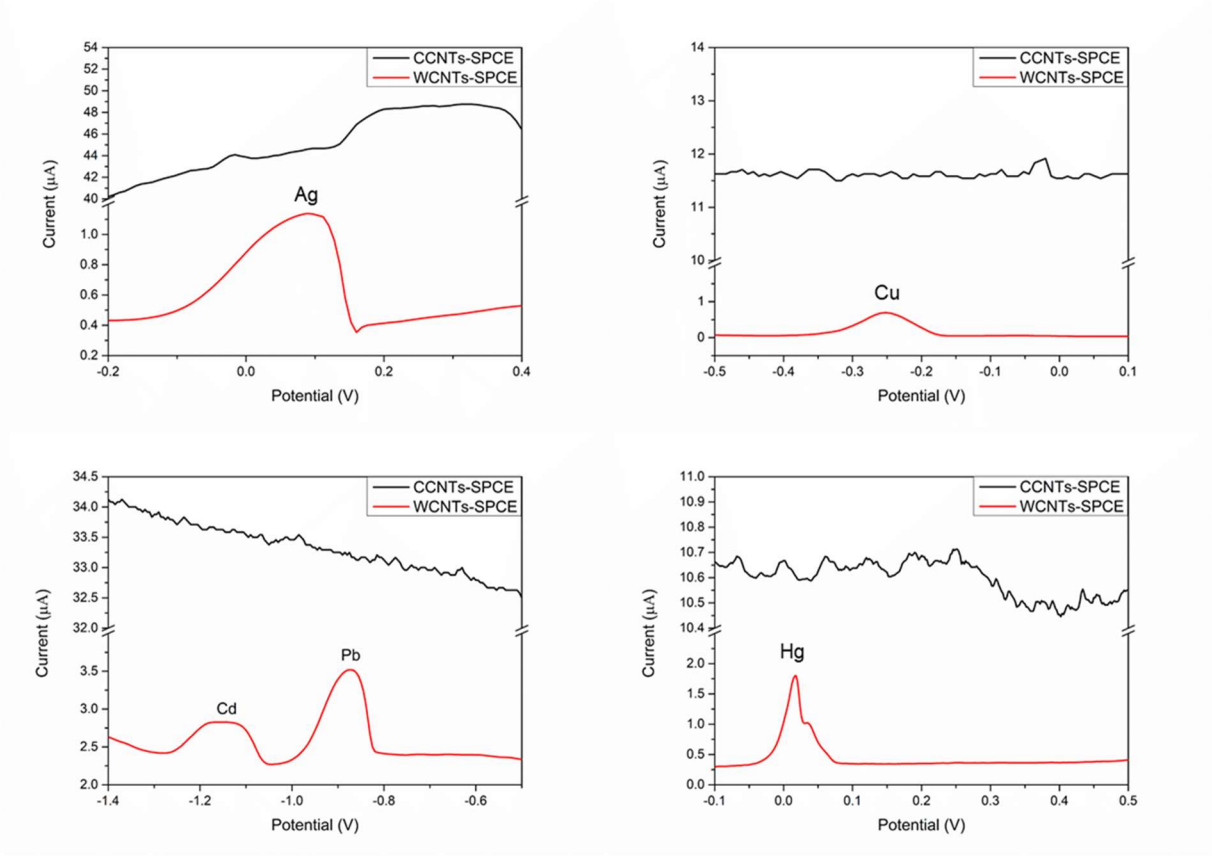


Figure 4. Detection of heavy metal ions with voltammetric sensors: Typical DPASVs of heavy metal ions (50 ppb, in 0.1 M acetate buffer, pH=4.5) with CCNTs and WCNTs modified SPE chips.

3.2. End-of-life assessment

The inclusion of end-of-life release is particularly relevant for the single-use materials/products (Pourzahedi & Eckelman, 2015), which are disposed afterwards. The predominant end-of-life fate for the SPEs (including SMs and EMs) are landfilling, open burning, incineration, or leakage into the natural environment, based on the prevalent international practices of solid waste management. We emphatically assess the physical fate of the SMs and EMs in these environmental sinks (Table

2). The airborne particle suspension was excluded as the presence of the materials in the atmosphere is short-lived.

The course of the SMs and EMs in incineration and landfill are well contemplated, compared with the other environmental sinks. The end-of-life scenario for carbon-based materials are expected to undergo near complete combustion during incineration. For example, carbon black is very likely to be completely combusted and eliminated during waste incineration under optimal stoichiometric conditions (Mueller et al., 2013; Johnson, 2016). Under oxidative conditions, carbon-based nanomaterials degrade completely at 740°C (Yang et al., 2004; Wang et al., 2006). The CO_2 emissions of the SMs and EMs from complete combustion are 1.62, 2.24, 1.51, 3.67, and 3.67 Kg-CO_2 eq./Kg of cotton textile, HDPE plastic, Kraft paper, carbon black, and CNTs, respectively (Ahamed et al., 2021a). The rest of the materials are non-carbon in origin, hence, no direct CO_2 is released from the combustion process. The metal particles were reported to be deposited in the bottom ash, slag, or filtered from airborne exhaust with up to 1% being released into the atmosphere as particulate emission (Keller et al., 2013; Johnson, 2016). Glass and ceramics are non-combustible and inert, hence, remain predominantly in the slag or bottom ash. In the case of landfill, the materials are deposited indefinitely at the landfill site and undergo the transition as depicted in Table 2. The weathered fragments of the materials may penetrate to the soil and surface waters via leachate run off and groundwater percolation possibly leading to toxic effects on living organisms. The SMs and EMs undergo similar transitions in open burning as in the case of incineration. The direct air emissions are high due to the lack of exhaust treatment system that result in increased air pollution effects including climate change, fine particulate matter formation, photochemical ozone formation, and terrestrial acidification potentials (Forbid et al., 2011;

419 Oguntoke et al., 2019). In addition, open burning might result in incomplete combustion products,
420 which complicate the understanding of the associated environmental implications.

421

422 Table 2. The physical fate of SMs and EMs in various environmental sinks.

SM/EM	**Landfill**	**Open burning**	**Incineration**	**Soil**	**Surface water**
Cotton textile	Biodegrade	Combust	Combust	Biodegrade	Biodegrade
HDPE plastic	Inert*	Combust	Combust	Inert*	Inert*
Kraft paper	Biodegrade	Combust	Combust	Biodegrade	Biodegrade
Graphic paper	Biodegrade	Combust	Combust	Biodegrade	Biodegrade
Glass	Inert	Inert	Inert	Inert	Inert
Ceramic	Inert	Inert	Inert	Inert	Inert
Platinum	Inert	Inert	Inert	Inert	Inert
Gold	Inert	Inert	Inert	Inert	Inert
Silver	Inert	Inert	Inert	Inert	Inert
Copper	Inert	Inert	Inert	Inert	Inert
Carbon black	Inert	Combust	Combust	Inert	Inert
Carbon nanotubes	Inert	Combust	Combust	Inert	Inert

†Biodegrade: can decompose in <1 year; Inert: remain physically inert for >100 years.

*Owing to the weathering effect there is a possibility of plastic fragmentation and microplastic formation.

Note: Residue will be present in most of the combustible materials depending on the ash content of the respective materials.

Release into the natural environment may pose unknown consequences in the cases of plastics and nanomaterials, especially the ecotoxic effects, which are still being studied. In the case of plastics, Lithner et al. (2012) studied the acute toxicity (short term: 24-72h) of five plastic type leachates in 26 commercial products to *Daphnia magna*. Polypropylene, ABS, and rigid PVC exhibited insignificant leachate toxicity, HDPE products caused acute toxic effect in 1 out of 5 samples and all the plasticized PVC and epoxy products exhibited acute toxicity in the tested samples, possibly due to the hydrophobic organics and cationic metals. Bejgarn et al. (2015) studied the acute toxicity of various types of ground plastic leachates from commercial products in *Nitocra spinipes,* with leachate from tire as a positive control. The results showed 38% of the plastics induce toxicity at high concentration (liquid/solid ratio: 10). However, no common trend in toxicity was observed as a function of artificial sunlight irradiation time (765 W/m^2; 300–800 nm). A study by Nolte et al. (2017) investigated the influence of plastic nanoparticle functionality and water hardness on cellular adsorption by algal cell walls. The results suggested that the usage of appropriate dosimetry, including material properties, agglomeration, cellular attachment, and adsorption could better describe the toxicity of nanoparticles. Hence, different types of plastics demonstrated the varied levels of toxicities.

A 90-day intravenous toxicity study by Lee et al. (2010) reported no observed systemic toxicity from a high dosage of $CaO\text{-}SiO_2\text{-}P_2O_5\text{-}B_2O_3$ glass-ceramics administered to male and female Sprague-Dawley rats. However, a 30-day intraperitoneal administration of calcium phosphate glass containing various concentrations of silica administered to C57BL/6 mice resulted in higher mortality with increasing concentration of silica in glass (Nagase et al., 1992). The toxicity was

attributed to the dissolution of Si^{4+}. Hence, the toxic effect of glass and ceramic is dependent on the dissolution of SiO_2. On the other hand, the remaining SMs (paper and cotton) are naturally compatible with the environment and undergo degradation or remain inert.

Particles of smaller size exhibit greater toxicity towards organisms (Park et al., 2011; Tuttle, 2012). However, the toxic effects of nanomaterials on the environment and human health have been difficult to predict (Foldbjerg et al., 2015; Ahamed et al., 2021b). The influence of environmental factors, such as weathering further complicates the understanding of the toxic effects. The toxicity of nanomaterials, such as gold (Khlebtsov & Dykman, 2010; Jia et al., 2017), silver (Du et al., 2018; Foldbjerg et al., 2015; McShan et al., 2014; Akter et al., 2018), and carbon nanomaterials (Berlo et al., 2012; Dong & Ma, 2015; Peng et al., 2020) have been widely studied and reviewed. In a study by Dahlben et al. (2013) of electronic memory devices enabled with CNTs, insignificant environmental impacts of CNTs were reported when compared to both the metals (particularly gold) and the process inputs used during production, indicating negligible nano-related impacts of nano-enabled devices. Eckelman et al. (2012) reported that an LCA on the aquatic ecotoxicity of non-nano emissions from CNT synthesis and upstream production of materials and energy resulted in ecotoxic impacts that were several orders of magnitude greater than those from CNT releases. Hence, the toxic effects of the nanomaterials reported are contentious and inconclusive at present. In addition, the research on the toxicity of platinum (Asharani et al., 2011; Sørensen et al., 2016; Hlavkova et al., 2018) and copper nanomaterials (Hua et al., 2014; Gosens et al., 2016; Meng et al., 2007; Ye et al., 2017) were minimal.

As an increasing number of nano-enabled products enter commercial applications, the concerns of the associated environmental implications are intensifying. Grillo et al. (2018) highlighted the need to establish the control of waste management processes for nanomaterials, as the amount of

467 nanomaterials in the environment increases gradually. Abbas et al. (2020) recommended that
468 robust standardized chemical extraction methods, analytical techniques (including imaging
469 technique and mass spectroscopy), long-term exposure studies, trophic level transfer and
470 transgenerational effects of transformed nanomaterials, extraction of nanomaterials from waste
471 before disposal techniques, and social awareness programs are required.

472 Hence, ceramic, glass or paper substrates are recommended as low-risk environmentally friendly
473 SM options over plastic, as the toxicity implications of microplastics in the environment are yet to
474 be fully quantified. Similarly, carbon black and WCNTs are recommended as suitable EMs for
475 SPE applications. A combination of either of these materials as SM and EM would represent an
476 environmentally friendly option for SPEs in voltammetric applications. Despite the plastic
477 demonstrating one of the lowest environmental footprints for production among SMs, it was
478 disregarded in the final recommendation due to the associated end-of-life consequences. However,
479 in cases where the end-of-life treatment options are favorable such as thermal treatments, the
480 application of plastic SM could be considered. The preference of ceramic, glass or paper for SMs
481 and carbon black or WCNTs for WEs are open, depending on the case specific requirements and
482 experimentally optimal conditions in the applications of SPEs. For example, the combination of
483 ceramic SM with WCNTs WE as demonstrated in Section 3.1.3 can be applied for heavy metal
484 analysis in environmental samples, while for wearable sweat analysis, a more flexible SM such as
485 paper or plastic may be preferred. The RE in most instances requires Ag, excluding the possible
486 application of carbon-based pseudo-RE. Hence, the CE and WE could adopt to carbon-based
487 electrodes to minimize the use of metals, thereby reducing their environmental footprint. Further
488 advancement of SPEs incorporating an environmentally sensitive design and material choice is
489 essential to alleviate the associated environmental impacts.

3.3. Limitations and prospects

The LCA result is restricted to the primary production process in the ecoinvent database, due to (i) the preliminary stages of development of the products, (ii) non-availability of the production data for finished products, (iii) private licenses, and (iv) variance in the sub-processes with respect to specific applications and manufacturers. Universal production datasets from the ecoinvent database were adopted in this study. Data gaps for novel technologies present a challenge in conducting the full LCA studies. Hence, studying the entire process flow and evaluating the respective environmental impacts of each of the compared products was beyond the scope of this study. Furthermore, special treatments before applications were excluded. For instance, paper-based SMs are exposed to various treatments, such as acid to improve the intrinsic qualities including hardness, purity, and chemical resistance of the material that were not accounted. In addition, the end-of-life interactions of EMs with the molecules in the environment were disregarded. For instance, the interaction of metal nanoparticles with various exhaust gases after incineration process could alter the toxicity potential of the emitted metal nanoparticles (Arvidsson et al., 2011). However, the impact assessment categorically highlights the impending negative effects of the material choices for SPEs.

Plastics showed one of the lowest footprints from the production process in SMs. However, other biodegradable materials (such as paper), or inert materials (such as ceramic or glass) are prioritized considering the possible end-of-life implications. Material selection and research addressing the development of improved and biodegradable SM is limited. Specific focus on the SMs would facilitate the advent of a superior SM in the future. For example, the use of biodegradable plastic produced from renewable resources would serve as a superior SM and overcome the shortfalls of fossil-based plastics. Polylactic acid (PLA) and polyhydroxyalkanoate (PHA) are the two

predominant bioplastics currently available as material options. The suitability of these bioplastics could be tested in future studies for application in SPEs. However, it is important to ensure that these alternative materials are sourced from sustainable practices and are biodegradable in the natural environments and biogenic in origin. Similarly, cotton and paper from sustainable resources and farming practices can act as an alternative. However, a full LCA of the biodegradable materials is essential before application.

In the case of EMs, metal-based EMs exhibited greater negative environmental implications in most instances, especially the noble metals. The impact assessment results suggested that the shift towards carbon-based EMs is essential to minimize the environmental footprint of SPEs. Although metals are completely recyclable natural resources, the use of metals in single-use miniature-sized product will make the recycling of the material challenging. The existing electronic waste recovery and treatment primarily involves mechanical and electromagnetic separation and extractive metallurgy. Furthermore, novel adsorption techniques are being developed for their recovery (Zhou et al., 2020b; Qin et al., 2020). Hence, the energy intensive production process of extracting these natural resources is considered underutilized, if the multiple reuses of the metal is not favored. Furthermore, SPEs are more likely non-recyclable due to the complex nature of materials used, low concentration of recoverable metals/materials, scattered distribution of the product, and impurities of the tested sample. On the other hand, the abundance of the element carbon makes the shift from scarcer resources beneficial (Arvidsson & Sanden, 2017). Hence, carbon-based nanomaterials attract a wide range of applications due to the abundant and inexpensive nature of its precursors. However, only the most common carbon-based materials were investigated in this study, while it has to be noted that currently various forms of carbon materials, including graphene, fullerene, carbon nanosheets, mesoporous carbon, and carbon dots, are available and could

potentially be used for preparation of the electrodes (Chen et al., 2020; Veksha et al., 2020a; Maharjan et al., 2019; Chen et al., 2019). Similar conclusions can be extended for the other carbon nanomaterials provided the production footprint is comparable. Additionally, the end-of-life incineration of carbon-based EMs offer a complete decomposition of the spent EMs.

Although the focus of this study is limited to voltammetric sensors, the results can be extended to other sensing techniques including potentiometry, coulometry and electrochemical impedence spectroscopy provided that the electrochemical performance is validated for different materials. The development of the SPEs market is inevitable with rising number of applications. The market projections remain positive and complacent. Mordor Intelligence report (2020) valued the electrochemical sensors market at US$ 6.19 billion in 2019. Furthermore, it is expected to attain US$ 11.83 billion by 2025 at a compound annual growth rate of 11.4%. The market projections provide a strong perspective of the likelihood of the electrochemical sensors in the commercial production and, thereby, leading to the release into the environment after its use phase. For instance, millions of people use the blood glucose monitors throughout the world daily (Carrell et al., 2019). Similarly, millions of units are expected to be used on a daily basis for sweat analysis alone in point-of-care healthcare, professional sporting, and recreational activity applications (He et al., 2019; Lee et al., 2017; Chen et al., 2017; Nyein et al., 2019), which creates a possibility of an enormous amount of resources being consumed and sensor waste being generated. It is an important juncture to decide the best material candidate for mass fabrications. Among the carbon-based EMs, carbon black and WCNTs demonstrated to be the least impactful to the environment. The conventional fossil-based carbon nanomaterial synthesis can be further averted by adopting to the waste-derived carbon nanomaterials as highlighted in this study. On the other hand, the utilization of waste-derived materials would boost the technology implementation of the waste-to-

559 materials initiatives that are in research stage to industrial-scale development. This in turn would
560 facilitate to achieve higher recycling and productive waste management technologies. Resorting
561 to materials with lower embodied energy of extraction and refining and possessing equivalent
562 functional properties as noble metals would be appropriate to minimize the environmental
563 implications (Dahlben et al., 2013; Pourzahedi & Eckelman, 2015). Nevertheless, further research
564 in specific applications, commercial scale production, and full life cycle environmental impact
565 evaluation of waste-derived carbon nanomaterials are recommended as the future directions. Life
566 cycle impacts will decline as nanomaterial production volumes increase and nanotechnologies
567 become more efficient and mature (Pourzahedi & Eckelman, 2015).

568 Furthermore, the economic cost of materials increases exponentially as the resource depletes over
569 time. Thereby, economic depletion occurs long before the actual physical depletion of the
570 resources (Allwood et al., 2011; Söderholm & Tilton, 2012). A shift towards renewable material
571 resources and supply chain is critical to ensure unhindered long-term application of any product.
572 Moreover, the metals are reliant on the availability of metal ores, which are restricted to certain
573 geographical locations that can be affected by geopolitical causes. On the other hand, carbon-based
574 materials can actually be free from such externalities.

575 LCA provides potential insights for areas of improvement in product manufacturing (Pourzahedi
576 et al., 2017). LCA has particularly greater influence at the early stage of product and process
577 design, when there is a freedom to implement substantial changes (Hellweg & Canals, 2014).
578 Switching materials/technologies due to the environmental implications after fully developing
579 products from extensive research and development would be a mistreatment of resources and result
580 in economic distress. Hence, evaluating the environmental impacts at the early research stage of
581 the product/technology development is ideal and proactive. It helps to save enormous economic

582 expenditures, including cost, labor, time, and infrastructure spending. However, the LCA involves
583 simplifications and standardization to facilitate consistent and accessible use in practice before
584 mainstream applications (Baitz et al., 2013).

585 The environmental impact assessment is of paramount importance from a global perspective. The
586 World Economic Forum predicted the evolving risk landscape for 2020 on a global scale. The
587 environmental risks were among the highest concerns in terms of likelihood and impact, when
588 compared to economic, geopolitical, societal, and technological risks (The global risks report
589 2020, 2020). Hence, minimizing the environmental footprints of every product is essential to
590 minimize the rising concerns and achieve the sustainable development goals.

591 **4. Conclusion**

592 Among the studied SMs, cotton demonstrated the highest environmental footprint for SPE
593 applications. However, ceramic, glass, or paper substrates are recommended as a low-risk
594 environmentally friendly SM option over plastic substrates, as the toxicity implications of
595 microplastics in the environment have not been fully quantified yet. The environmental
596 implications of EMs were several orders of magnitude greater than the SMs when assessed per
597 unit of the material. Gold and platinum exhibited the highest environmental impacts among all the
598 studied materials. The involvement of intensive mining and processing activities for these metals
599 contribute immensely to the negative environmental effects. The environmentally friendly option
600 would be to avoid the usage of metals as highlighted in this study. Especially, if the performance
601 is not compromised, carbon-based electrodes are superior to metal electrodes. The order of priority
602 can be as follows, Carbon > Copper > Silver > Gold/Platinum. A combination of either ceramic,
603 glass, or paper SM and carbon-based EM would represent the most environmentally friendly
604 option for SPEs in voltammetric sensing applications. Depending on the case specific requirements

and the optimal experimental conditions in the SPE applications, the final selection is open among the above-mentioned SMs and EMs. Although RE requires Ag in most instances, WE and CE could consider carbon-based electrodes in order to minimize the use of metals, thereby reducing their environmental footprint. Notably, WCNTs are produced from resources that are abundant, inexpensive, bear low or negative environmental footprint, and exhibited comparable electrochemical performance to the conventional fossil-derived materials. In this study, WCNTs achieved superior signal intensity and lower background current for all the studied heavy metal ion detections when compared to CCNTs, suggesting that WCNTs can be a viable option to CCNTs in SPEs. Further optimization of the WCNT synthesis with industrial-scale production methods could additionally reduce the environmental footprint of the EMs. Moreover, the waste management of carbon-based electrodes is uncomplicated with the existing options of incineration or other thermal treatment being most suitable. Environmentally sensitive design and material selection in SPEs are important to alleviate the associated environmental impacts and achieve the sustainable development goals.

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Declaration of competing interest

The authors declare no competing interests.

CRediT authorship contribution statement

Ashiq Ahamed: Conceptualization; Methodology; Software modelling; Investigation; Data analysis; Validation; Writing - original draft; Writing - review & editing. **Liya Ge:** Experimental investigation; Data analysis; Writing - review & editing. **Ke Zhao:** Experimental investigation; Materials characterization. **Andrei Veksha:** Writing - review & editing. **Johan Bobacka:** Supervision; Writing - review & editing. **Grzegorz Lisak:** Supervision; Conceptualization; Project administration; Funding acquisition; Resources; Writing - review & editing.

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