Research article

Design and operation of pilot-scale microbial electrosynthesis for the production of acetic acid from biogas with economic and environmental assessment

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A B S T R A C T

The embryonic technology of microbial electrosynthesis (MES) possesses the potential to alleviate global CO_2 concentration with concomitant recovery of valuables. However, due to the significant bottlenecks of inferior yield of valuables and higher capital cost, its potential has not been fully realized at a larger scale till date. With the aim of bridging this lacuna, a first of its kind pilot-scale MES (PSMES) was designed and operated to yield acetic acid from biogas. The PSMES was able to produce 70.55 g m^{-2}.day of acetic acid in its extraction chamber with the coulombic efficiency of 77.8 % for an imposed cathode potential of −1.0 V vs. standard hydrogen electrode. Moreover, life cycle assessment (LCA) and economic analysis of the PSMES was also conducted to elucidate the economic and environmental feasibility of the same. From the LCA and economic analysis of the PSMES, it was inferred that acrylic sheet and carbon felt used during the fabrication of PSMES were the major culprit in terms of both environmental and economic sustainability and thus should be replaced with greener but cost-effective materials. Therefore, these results would guide the budding scholars in designing more economical and environment friendly scaled-up MES, thus paving towards the commercialization of this ingenious technology.

1. Introduction

Increasing global atmospheric CO_2 concentration is a major environmental issue and to mitigate the same, scientists are toiling hard to develop efficient carbon capture and sequestration technologies [1]. Bioelectrochemical CO_2 sequestration through microbial electrosynthesis (MES) is one such futuristic technology, which employs microbes as catalysts to sequester CO_2 and produce multi-carbon organic compounds, primarily acetic acid [2]. The technology of MES is at the embryonic stage and hence, researchers are toiling hard to overcome the hurdle associated with this innovative technology so that it can be efficaciously applied in the field to alleviate real world problems [3]. The inventive technology of MES can produce value-added chemicals through carbon sequestration employing self-sustainable and cost-effective microbes as biocatalysts [4]. However, due to the major bottlenecks like inferior production rate of valuables and exorbitant fabrication cost associated with this technology, it has not been successfully commercialized or employed in the field conditions to deal with real life situations [5].

Generally, scaling-up of bioelectrochemical systems is a major challenge as ohmic, concentration, and activation losses of these systems cumulate, when a lab-scale setup is scaled-up and implemented in the field, thus significantly diminishing the yield [6,7]. Also, the capital expenditure required for the construction of field-scale bioelectrochemical systems (BESs) is the major limitation, which prohibits its scale-up and further commercialization [8,9]. Previously also, researchers have tried to scale-up BESs like microbial electrolysis cell (MEC) and microbial fuel cell (MFC); however, due to their inferior performance in the field-scale demonstrations, these technologies have not been commercialized till date [10–12]. However, to the best of our knowledge, scaling-up of MES for acetic acid production from lab to field or pilot-scale application has not been conceptualized in the literature till date. Operation of a scaled-up MES setup would provide key insights of the critical areas/parameters, where the researchers should focus on so that this technology can be successfully applied in the real field and simultaneously commercialized. Also, the environmental and economic assessment of a operational scaled-up MES setup is yet to be documented; however, environmental and economic evaluation of hypothetical full-scale MES has been presented previously [13,14]. The major drawback of such hypothetical investigations on an embryonic technology is that it fails to draw the complete picture by inaccurate assumptions made to simplify the process. Also, it is very difficult to predict the exact path this technology will be heading towards due to the infancy nature of MES.

This research has significant implications for the future commercialization and scaling-up of MES. By demonstrating the successful operation of a pilot-scale MES for acetic acid production from biogas-derived CO_2, the study provides a critical proof of concept for larger-scale applications. The technology's potential for carbon sequestration could be a viable method for reducing atmospheric CO_2 levels, directly contributing to mitigating global warming and advancing efforts toward net-zero emissions. The insights gained from the life cycle and economic assessments highlight areas where material improvements can enhance both environmental and economic sustainability, thus paving the way for more cost-effective solutions at commercial scales. As the world increasingly focuses on carbon reduction technologies, the scalability of MES could become a cornerstone in sustainable industrial practices, turning CO_2 into valuable products while addressing the urgent need for climate action [15].

With the aim of bridging this gap, a modular pilot-scale MES (PSMES) was formulated and operated, which synthesized acetic acid in the cathodic chamber, simultaneously separated it from the catholyte and furthermore acetic acid was accumulated in a separate extraction chamber. In this research, the novel modular PSMES was designed based on the results obtained from our past experiments [16–18] and its performance was also assessed. The environmental impact assessment of the PSMES setup was also carried out using life cycle assessment (LCA) by considering the environmental impact generated by the PSMES due to the exhaustion of materials, energy and resources, and emission of gases during the synthesis of acetic acid [19,20]. Moreover, the economic evaluation of PSMES was also accomplished by explaining the major components affecting the cost of the setup [21]. Moreover, ways to further enhance the revenue generation from the PSMES were also explained. Thus, the results obtained and lessons learnt during the design and operation of this PSMES would provide valuable information to the researchers, which would be helpful to them in spanning out their research for overcoming the hurdles encountered in the present demonstration.

2. Material and methods

2.1. Pilot-scale MES design and fabrication

A novel modular PSMES was designed and fabricated using acrylic sheets, which was not only to synthesize acetic acid using CO_2 as substrate, however it also separated the produced acetic acid from the catholyte and the same was collected in the extraction chamber. The designed PSMES consisted of five chambers, namely two extraction chambers, two anodic chambers, and a central cathodic chamber (Fig. 1a). The peripheral extraction and anodic chambers were having volume of 3.8 L each and the central cathodic chamber was having volume of 2.2 L (Fig. 1b). Thus, the total volume of the PSMES setup was 12.6 L and it was square in shape with each side of
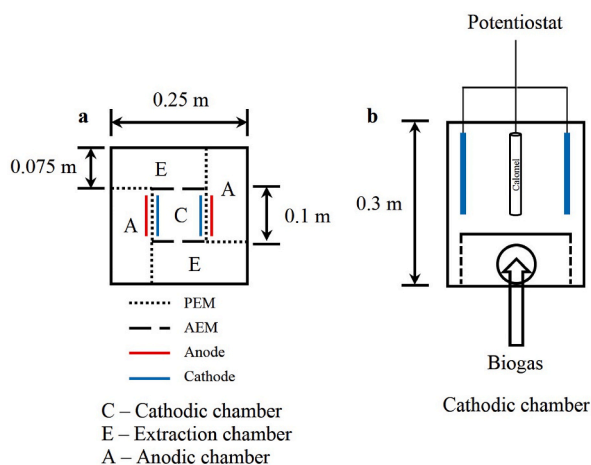

Fig. 1. (a) Top view of the pilot-scale MES setup and (b) side view of the cathodic chamber.

25 cm and height of 30 cm. Moreover, the PSMES setup was designed accordingly so that it can successfully extract acetic acid from a mixture of other components present in the cathodic chamber. The principles and mechanisms behind the exaction and purification of acetic acid synthesized in the cathodic chamber of MES, employing a three chamber modular reactor using similar reactor at the lab-scale was demonstrated earlier by the authors [17]. In the employed three-chamber MES reactor, acetic acid was produced in the cathodic chamber and extracted through an anion exchange membrane (AEM) into an adjacent extraction chamber. The acetate ions formed in the cathodic chamber passed through the AEM and combined with protons migrating from the anodic chamber via a proton exchange membrane (PEM), forming acetic acid in the extraction chamber. This setup enabled continuous separation and purification of acetic acid from the catholyte, ensuring a cleaner product without the presence of microbes or other organic compounds. However, the very first of its kind demonstration of three chamber MES reactor was performed by a different research group [22].

Based on the principles presented earlier the different chambers were parted using different types of membranes [23]. The extraction chamber was separated from the cathodic chamber and anodic chamber by AEM and PEM, respectively, so that anionic acetate ions and protons can migrate into the extraction chamber from the cathodic and anodic chamber, respectively. Likewise, the anodic chamber was separated from the extraction and cathodic chamber by PEM, so that only protons can transfer from the anodic chamber to these chambers. In the extraction chamber of PSMES, purified acetic acid was produced due to the combination of anionic acetate ions transported from the cathodic chamber through the AEM and cationic protons moving from the anodic chamber through the cation exchange membrane. For this setup, clayware ceramic and Ralex AM-PES (Mega, Czech Republic) were used as PEM and AEM, respectively [24]. Previously the lab-scale MES was operated with Nafion 117 (DuPont, USA) as the PEM; however due to the high cost of Nafion membrane, clayware-based PEM was used during the fabrication of PSMES [23]. Thus, this novel design of PSMES employing two different types of membrane ensured that only acetic acid is recovered in the extraction chamber of the setup, where protons and acetate traversing from the anodic and cathodic chamber, respectively, combine to form acetic acid.

Carbon felt with projected surface area of 116 cm^2 and dimension of 29 cm × 4 cm each was used as the electrode material for both anodic and cathodic chambers, whereas no electrode was present in the extraction chamber. Further, the cathodic chamber consisted of two pieces of carbon felt electrode placed adjacent to the PEM that is separating the anodic and cathodic chamber. Moreover, both the anodic chambers housed a piece of carbon felt each of same dimension as mentioned previously. No catalysts coating was used for the electrodes, as coating of expensive metal catalysts on the electrodes would be economically infeasible, given the higher surface area of the electrodes used in the present pilot setup. Both the anode and cathode were placed in close proximity to each other in respective chambers only to be separated by the PEM. Each panel of membrane separating the adjacent chambers was of 120 cm^2 of projected surface area. Also, provision was made to place the reference calomel electrode in the middle of the cathodic chamber so that potential can be imposed on the cathode.

2.2. Operation of the pilot-scale MES

To facilitate the development of mature biofilm, the pre-treated inoculum was acclimatized on the carbon felt, prior to the operation of the PSMES as explained earlier [18]. After the formation of biofilm, general acetogen medium used as the catholyte till then was replaced with a mineral solution containing (in g L^{-1}): 0.33 KH_2PO_4, 0.45 K_2HPO_4, 1 NH_4Cl, 0.1 KCl, 0.8 NaCl and 0.2 $MgSO_4 \cdot 7H_2O$ with pH preserved at 5.0 ± 0.2 [25,26]. The catholyte pH of 5.0 was found to improve the efficacy of MES considerably [27]; hence, it was maintained during the operation of the PSMES. Thus, after the formation of biofilm, biogas purging was initiated in the cathodic chamber of PSMES and the amount of acetic acid produced in both the chambers was measured daily. During the operation of the PSMES, distilled water was used as the electrolyte for both the extraction and anodic chambers. Daily samples were collected for analysis from the extraction chamber and subsequently, it was entirely refilled with distilled water so that the concentration gradient of acetic acid between the extraction and cathodic chamber remains high enough, which was imperative for the effective separation of acetic acid from the catholyte. Furthermore, the amount of anolyte reduced due to uninterrupted water splitting was replenished with distilled water as and when required.

Biogas acquired from an up-flow anaerobic sludge blanket (UASB) reactor was employed as the source of CO_2 and as the lone carbon source for the PSMES cell to produce acetic acid. Biogas majorly containing CH_4 (53 ± 4 %) and CO_2 (41 ± 2 %) and negligible quantity of other gases, like H_2S (<1 %), NH_3 (<1 %), CO, etc., was purged intermittently in the cathodic chamber at the rate of 7 L min^{-1} for a period of 3 h daily. The composition of both biogas used as feedstock for the PSMES and cathode-off gas released from the PSMES was quantified using gas chromatography (GC) following the methods as explained previously [28]. Intermittent supply of biogas was used instead of continuous supply as intermittent supply was found to enhance the yield of acetic acid as explained previously [16]. The pilot-scale MES reactor was operated in a fed-batch mode as stated previously [29]. Daily samples were collected from the extraction and cathodic chambers and were examined to detect the quantity of acetic acid present in them through gas chromatography coupled with mass spectroscopy (Agilent Technologies MS-5977B, Penang, Malaysia) as per the methods explained previously [17].

The catholyte pH and the pH of liquid samples of the extraction chamber were examined daily using a pH probe connected to a water quality analyser (Thermo Fisher Scientific, USA). After every three days, 30 % of the catholyte was substituted with a newly made mineral solution, which provided necessary nutrients to the cathodic microbiome required for their growth. The PSMES setup was operated at an ambient temperature of 29 ± 3 °C for more than 60 days and at the fixed operating parameters until stable acetic acid production rate (similar acetic acid production for three consecutive days) was obtained.

2.3. Life cycle assessment

The LCA of PSMES was performed to elucidate the degree of environmental sustainability of this novel PSMES and plausible ways to further alleviate the environmental impacts associated with the fabrication and operation of the PSMES. The LCA was performed in four major steps as suggested in ISO 14040–44:2006 [30]. These four steps involve defining the goal and scope, preparation of life cycle inventory (LCI), life cycle impact assessment (LCIA) and the interpretation of results. The goal of the present LCA was to perform environmental analysis and determine the environmental impact associated with the fabrication and operation of PEMES setup. The primary goal of the investigation was to determine the components of PSMES setup that were creating the major impact on the environment and ways to mitigate the same. The optimized parameters of the PSMES setup, as explained previously were used for the present LCA. Only the processes involved with the operation and fabrication of PSMES are considered; hence, this LCA can be classified as a gate-to-gate approach [31]. Production processes of the materials used during the fabrication and operation of the PSMES setup are considered during the present LCA. The treatment processes involved in treating water used as anolyte during the operation of the PSMES setup is not considered; however, only water as material input was considered during the LCA. The functional unit for this LCA was selected to be 1 g of acetic acid produced by PSMES. Moreover, the target audience for the present LCA would be researchers, scientists, and decision and policy makers. Also, the geographic and temporal scope of the investigation was India and 2017 to 2022, respectively.

2.3.1. Definition of system boundary

The system boundary of the present LCA involves only the processes involved in the operation and fabrication of PSMES. The processes associated with the manufacturing of the materials required for the operation and fabrication of PSMES are considered for the LCA (Fig. S1). However, the production process of biogas used as feedstock during the operation of PSMES setup, and downstream processes involved in the transportation, further purification (if required) and utilization of acetic acid was not considered during the LCA. Electricity was consumed during the operation of PSMES setup, hence the processes involved in the production of electricity in the context of India were considered. Electricity in India is provided using a mix of resources: 62 % fossil, 13 % hydro, 4 % nuclear, 9 % solar, and 12 % wind [32]. The cathode off-gas emitted from the PSMES setup was considered as emission and its impacts on the environment were considered in the present LCA.

2.3.2. Life cycle impact assessment

The environmental impacts of the PSMES were analysed by converting the formulated LCI into the related environmental impact categories using IMPACT 2002+® method using SimaPro 8.0.3 inbuilt Ecoinvent® version 3.1 datasets, available in the SimaPro software [33]. The characterisation of the environmental impacts was selected for eight mid-point impact categories, namely: (1) carcinogens; (2) non-carcinogens; (3) aquatic ecotoxicity; (4) terrestrial ecotoxicity; (5) terrestrial acidification; (6) aquatic acidification; (7) aquatic eutrophication; (8) global warming (GW). Three end-point damage assessment categories, namely: (1) human health; (2) eco-system quality, and (3) climate change, were also evaluated using the same dataset. The reference time horizon for global warming potential is considered as 100 years, as it is a standard reference time frame for climate change policy [34]. However, for impacts associated with the acidification, toxicity impact categories have a large magnitude of impacts on the future generations over a long-time horizon (>100 years). Also, each end-point damage assessment category is allotted an equal weightage of 33 % each in India and same was also done in the present investigation. Furthermore, sensitivity analysis was also performed to determine the reduction in GW potential if a portion of the methane from the cathode off-gas was captured and reused instead of emitting it into the atmosphere.

2.4. Economic assessment of pilot-scale MES

The economic valuation of the PSMES setup was performed to determine the major components affecting the operating and fabrication cost of PSMES, and ways to reduce these costs are also being highlighted. Moreover, the degree of revenue generation from the PSMES was also estimated in addition to the breakeven analysis. Also, sensitivity analysis was conducted to determine the change in the breakeven time with the alteration in the production rate of the PSMES setup. Thus, the current economic evaluation expects to assist the researchers in developing more economical MES setups to achieve higher degree of economic sustainability associated with this novel technology.

The fabrication and operating cost of the PSMES setup was determined by considering the cost of the materials used for the same. As it was done during the LCA of the PSMES setup, no pre- and post-processes of PSMES setup were considered during the present cost analysis. Therefore, the cost of UASB reactor, and downstream and transportation cost of acetic acid were out of the scope of the present cost analysis. Materials procurement cost from international suppliers was used during these calculations. The capital cost was defined as the money invested in the fabrication of the PSMES setup, which included the cost of different materials and manpower required during the fabrication of the PSMES setup (Table S2).

Revenue was estimated only considering the cost of the acetic acid synthesized in the extraction chamber due to the purified nature of the acetic acid produced in this chamber. Moreover, the upgraded biogas leaving the cathodic chamber was not considered as the source of revenue as it was presently not reused for cooking or electricity generation; however, it was flared. The cost of acetic acid with average concentration of 372 mg L^{-1} produced in the extraction chamber was estimated as per the cost of commercial grade acetic acid (99 % pure with concentration of 1039.8 mg L^{-1}). The cost of acetic acid produced in the extraction chamber of the PSMES was estimated based on the cost of this dilute acetic acid solution produced in PSMES as per the cost of the concentrated commercial grade

acetic acid required to prepare it through dilution. Moreover, the lifespan of acrylic sheet was considered as 3 years, whereas for all the other materials it was considered to be two years.

3. Results and discussion

3.1. Electrochemical performance and overpotential losses

The overpotential loss for different imposed potentials was measured regularly and it was found that the overpotential loss increased with the application of further negative imposed potential (Fig. 2). The overpotential losses in a BES are affected by many parameters, like charge transfer resistance, ohmic losses, activation losses, properties and characteristics of electrode, electrolyte and membrane used, distance between the electrodes and the nature of the biofilm present on the cathode [35]. The losses arising due to the use of different materials during the fabrication of PSMES will be constant for different imposed potential; however, other factors affecting overpotential losses would change, when the imposed potential is changed [4]. Also, these factors do not affect the overpotential losses linearly, which can be easily ascertained by the non-linear increment in overpotential losses with the application of further negative imposed potential. For instance, the overpotential losses were 0.157 V and 0.334 V for an applied potential of -0.6 V and -1.2 V vs. SHE, respectively, which evidently demonstrates that the overpotential loss increased 2.13 times, when the imposed potential was doubled (Fig. 2).

Due to the non-linear increment in the overpotential losses with the application of further negative applied potential, overpotential loss per unit of imposed potential was estimated to ascertain the optimum potential required to minimize the overpotential losses encountered during the operation of PSMES. It was found that the overpotential loss per unit of imposed potential (0.219) was least for the applied potential of -1.0 V vs. SHE, which increased to 0.278 V for the imposed potential of -1.2 V vs. SHE (Fig. 2). Therefore, with the application of 0.2 V more negative imposed potential, the overpotential loss per unit imposed potential increased by 1.27 times and hence, it can be said that in terms of overpotential losses, -1.0 V vs. SHE was the optimum imposed potential for the PSMES setup used in the present investigation.

3.2. Operation of PSMES and acetic acid production rate

During the operation of PSMES, the acetic acid production rate for both extraction and cathodic chamber gradually increased as the microbiome got better acclimatized to the electron supply and biogas supplied to them [36]. Once stable production rate was reached for every imposed potential, the potential was shifted to a more negative potential and in this way the different performance parameters of PSMES were assessed under different applied potential. During the operation of PSMES, it was found that the concentration of acetic acid in the cathodic chamber of PSMES was always lesser than that found in the extraction chamber at all the applied potential. Similar observation were also made when a triple chamber MES was employed at a lab-scale to separate acetic acid from the catholyte [22]. This signifies that the acetate produced in the cathodic chamber of PSMES successfully migrated through the AEM and it was finally getting accumulated in the extraction chamber. Analogous observation was also noted during the operation of the modular lab-scale MES [23].

As mentioned previously, the concentration of acetic acid was always observed to be higher in the extraction chamber when compared to that of cathodic chamber of PSMES. Therefore, the accumulation rate of acetic acid was also higher for the extraction chamber in comparison to the cathodic chamber of the PSMES setup under a fixed imposed potential. In this regard, the concentration and production rate of acetic acid in the extraction chamber of PSMES was 134 ± 8 mg L^{-1} and 2.27 ± 0.13 mM day^{-1}, respectively, for an imposed potential of -0.6 V vs. SHE (Table 1). Moreover, when the imposed potential was doubled to -1.2 V vs. SHE, both the concentration and the acetic acid accumulation rate in the extraction chamber increased more than three-folds to 411 ± 21 mg L^{-1} and
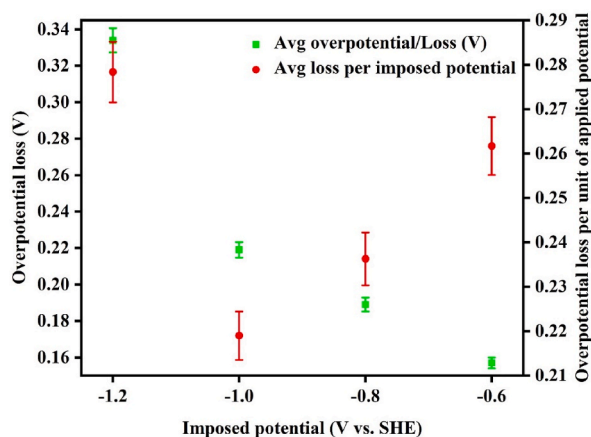

Fig. 2. Overpotential loss and overpotential loss per unit applied potential for different imposed potential as observed in PSMES setup.

Table 1

Production rate acetic acid for extraction and cathodic chamber of PSMES setup.

Imposed potential (V vs. SHE)	Extraction chamber			Cathodic chamber		
	Concent-ration (mg L^{-1})	Production rate (mM day^{-1})	Normalized production rate (g m^{-2}.day)	Concent-ration (mg L^{-1})	Production rate (mM day^{-1})	Normalized production rate (g m^{-2}.day)
−0.6	134 ± 8	2.27 ± 0.13	25.41 ± 1.31	104 ± 6	1.76 ± 0.09	17.17 ± 0.85
−0.8	335 ± 17	5.68 ± 0.29	63.53 ± 3.19	271 ± 14	4.59 ± 0.21	44.74 ± 2.21
−1.0	372 ± 19	6.31 ± 0.31	70.55 ± 3.64	291 ± 16	4.93 ± 0.26	48.04 ± 2.42
−1.2	411 ± 21	6.97 ± 0.33	77.95 ± 3.89	331 ± 17	5.61 ± 0.27	54.54 ± 2.87

6.97 ± 0.33 mM day^{-1}, respectively. Likewise, similar acetic acid concentration of 330 mg L^{-1} was also observed in lab-scale MES operated with carbon felt cathode operated at −0.85 V vs. SHE and inoculated with domestic wastewater sludge [37]. The concentration and acetic acid production rate in the cathodic chamber of PSMES setup was 104 ± 6 mg L^{-1} and 1.76 ± 0.09 mM day^{-1}, respectively, for an applied potential of −0.6 V vs. SHE. These values increased by 3.2 times to 331 ± 17 mg L^{-1} and 5.61 ± 0.27 mM day^{-1} for the concentration and acetic acid production rate, respectively, when the imposed potential was doubled to −1.2 V vs. SHE. Therefore, these findings again corroborate the fact that the production rate of acetic acid can be enhanced with the application of more negative imposed potential in PSMES setup as observed during the operation of a lab-scale MES.

3.3. Carbon recovery efficiency, coulombic efficiency and energy requirement

Coulombic efficiency (CE) is a vital parameter for any BES and hence for MES, as it signifies the efficacy of microbes to use electrons as reducing equivalents and convert CO_2 into organic chemicals. The CE of a MES is majorly affected by the production rate of organic compounds and the amount of electricity supplied to it measured by the current density, given that all other operating parameters remain constant. During the operation of PSMES setup at different imposed potentials, both the acetic acid production rate of and current density increased with the application further negative imposed potential and simultaneously CE of PSMES setup was also found to surge with the application more negative applied potential (Table 2). The highest CE of 86.76 ± 4.39 % was observed for the applied potential of −1.2 V vs. SHE, which was 28 % higher than that obtained at the applied potential of −0.6 V vs. SHE (67.88 ± 3.41 %). The increase in CE and acetic acid production rate with the application of further negative imposed potential can be ascertained to the fact that at a higher imposed potential, more electrons were available to the electrotrophs for the reduction of CO_2, and hence, both current density and production rate of acetic acid was enhanced. Moreover, similar CE of 89.5 % was observed for a lab-scale MES operated with carbon felt poised at a potential of −0.9 V vs. SHE and inoculated with sewage sludge [38].

3.4. Mid-point impact assessment

Impact of PSMES fabrication and operation was analysed based on eight mid-point impact categories and it was found that carbon felt and acrylic sheet used for the fabrication of PSMES setup were generating the maximum impact in majority of the mid-point impact categories. In the category of carcinogens, acrylic sheet was having the maximum impact of 0.263 kg C_2H_3Cl eq., which can be attributed to the release of aromatic hydrocarbons and 2,3,7,8-tetrachlorodibenzo-p-dioxin, both known carcinogens into the atmosphere during the production procedure of acrylic sheet (Fig. 3a). Moreover, carbon felt and clayware membrane used in PSMES were also having significant impact of 0.071 kg C_2H_3Cl eq. and 0.117 kg C_2H_3Cl eq., respectively, which was due to the release of benzo(a) pyrene and arsenic, and aromatic hydrocarbons and 2,3,7,8-tetrachlorodibenzo-p-dioxin, respectively, during the manufacturing of these materials.

In terms of the category of non-carcinogens, carbon felt was generating the maximum impact of 0.441 kg C_2H_3Cl eq. followed by acrylic sheet, which was having an impact of 0.404 kg C_2H_3Cl eq. (Fig. 3b). This can be again attributed to the release of 2,3,7,8-tetrachlorodibenzo-p-dioxin and arsenic into the atmosphere during the production of acrylic sheet and carbon felt, respectively. Other than that, clayware membrane and electricity were also possessing significant impact of 0.021 kg C_2H_3Cl eq. and 0.014 kg C_2H_3Cl eq., respectively, in the category of non-carcinogens. This can be again attributed to the release of both 2,3,7,8-tetrachlorodibenzo-p-dioxin and arsenic into the atmosphere during the production of clayware membrane and electricity.

In terms of aquatic ecotoxicity, carbon felt was having the highest impact of 778.23 kg triethylene glycol (TEG) water followed by acrylic sheet (191.70 kg TEG water) and electricity (108.09 kg TEG water) (Fig. 3c). This can be again accredited to the release of aluminium into the atmosphere and water during the production of both carbon felt and acrylic sheet. For electricity production,

Table 2

Performance parameters of PSMES setup at different imposed potentials.

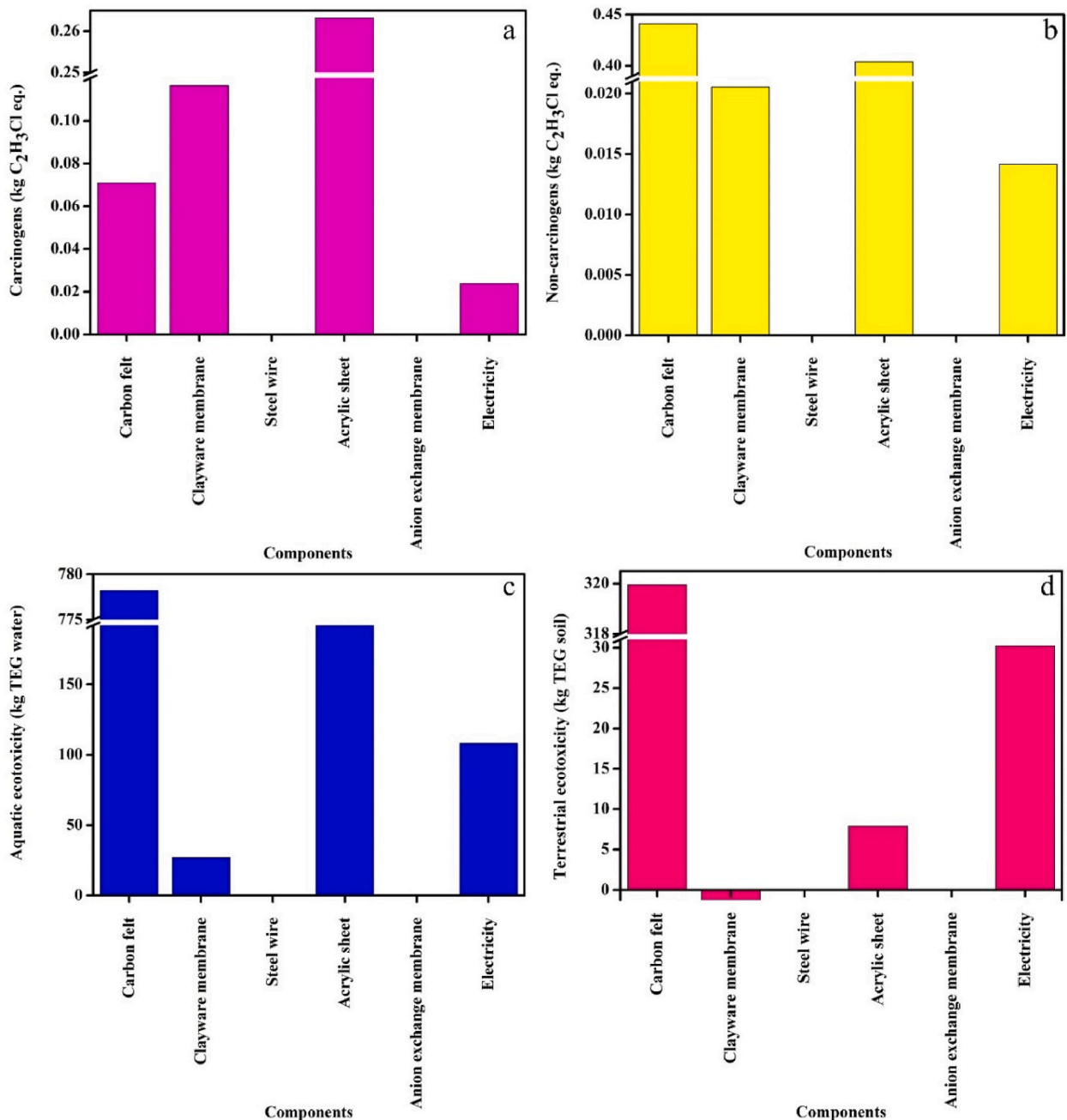| Imposed potential (V vs. SHE)	Current density (A m^{-2})	Coulombic efficiency (%)	Carbon recovery efficiency (%)	Energy requirement (W.h M^{-1})
−0.6	1.00 ± 0.05	67.88 ± 3.41	1.34 ± 0.05	32.04 ± 1.65
−0.8	2.31 ± 0.12	76.57 ± 3.85	3.49 ± 0.18	22.40 ± 1.14
−1.0	2.44 ± 0.13	77.84 ± 3.91	3.74 ± 0.19	18.95 ± 0.97
−1.2	2.49 ± 0.13	86.76 ± 4.39	4.26 ± 0.21	29.66 ± 1.47

Fig. 3. Categorized impact results for (a) Carcinogens, (b) Non-carcinogens, (c) Aquatic ecotoxicity, and (d) Terrestrial ecotoxicity.

aluminium is released into the air during its production process, thus it is having a significant impact in terms of aquatic ecotoxicity. Moreover, in the category of terrestrial ecotoxicity, carbon felt was having the highest impact of 319.95 kg TEG soil followed by electricity (30.25 kg TEG soil) and acrylic sheet (7.90 kg TEG soil) (Fig. 3d). This can be again ascribed to the release of aluminium and nickel into the air during the production of carbon felt, acrylic sheet and electricity. Also, it is worth noting that the use of clayware membrane reduced terrestrial ecotoxicity by 1.23 kg TEG soil as during its production process, zinc from the environment was offset, thus reducing terrestrial ecotoxicity.

In the category of aquatic acidification, again acrylic sheet used during the fabrication of PSMES setup was having the highest impact of 0.115 kg SO_2 eq. followed by electricity (0.030 kg SO_2 eq.) and carbon felt (0.026 kg SO_2 eq.) (Fig. 4a). This can be again accredited to the release of SO_2, NO_x and H_2SO_4 into the air during the production of carbon felt and acrylic sheet. Also, clayware membrane was functioning in reducing aquatic acidification, which was due to the offset of SO_2 and HCl emission during the production of this material. Similarly for terrestrial acidification, acrylic sheet was generating the maximum impact of 0.321 kg SO_2 eq.
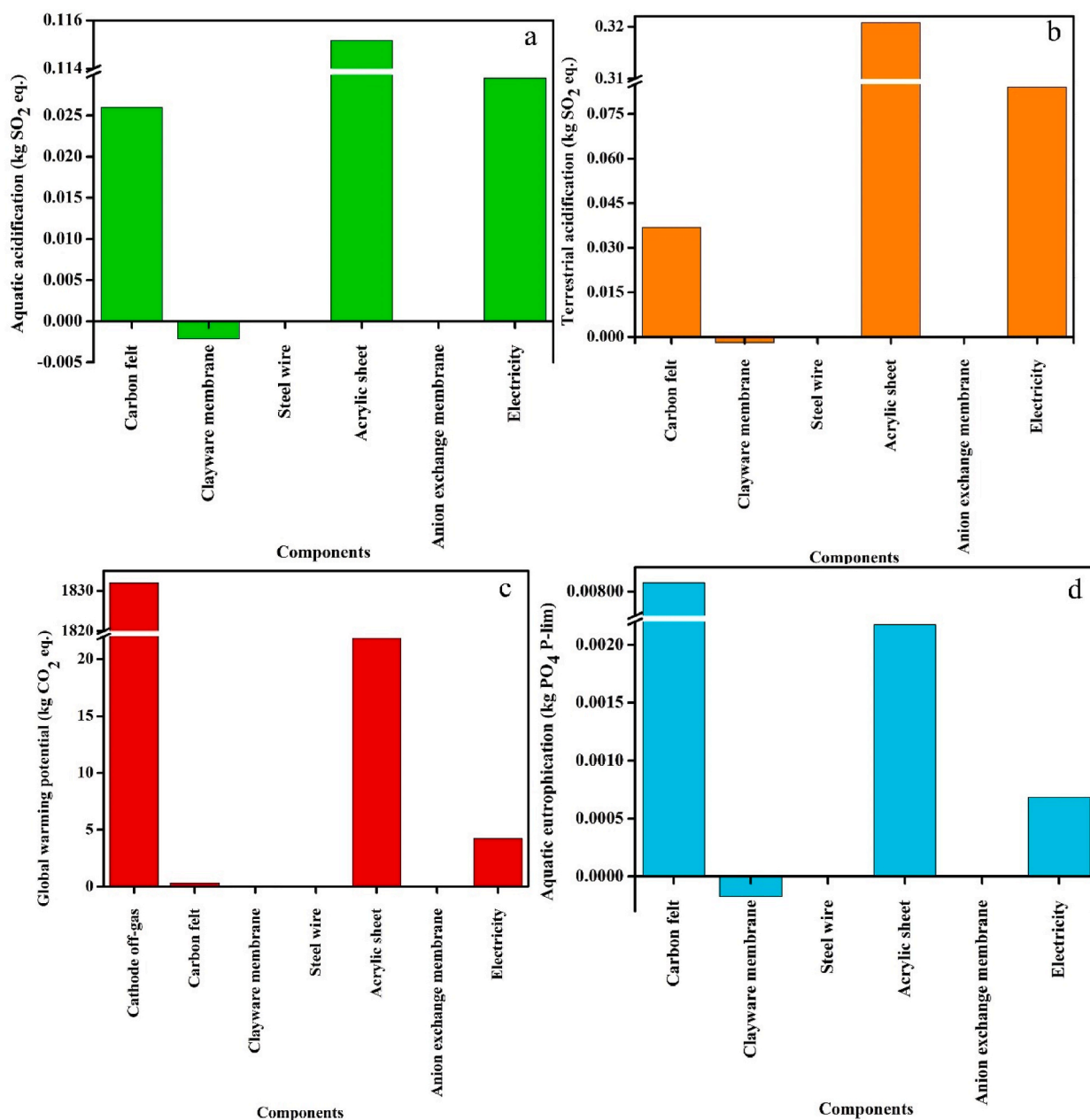
Fig. 4. Categorized impact results for (a) Aquatic acidification, (b) Terrestrial acidification, (c) Global warming potential, and (d) Aquatic eutrophication.

followed by electricity (0.084 kg SO_2 eq.) and carbon felt (0.037 kg SO_2 eq.) (Fig. 4b). This can be again accredited to the release of SO_2 and NO_x into the atmosphere during the production of carbon felt, acrylic sheet, and electricity. In this category also, clayware membrane demonstrated a beneficial impact on the environment due to the environmental offset of SO_2 and NO_x emission during the production of clayware membrane.

The major impact on the environment can be ascertained by estimating the GW potential of a process. In this regard, the cathode off-gas containing majorly CO_2 and CH_4 was possessing the highest GW potential of 1832 kg CO_2 eq (Fig. 4c), if it is released into the atmosphere without being used as energy source. These results suggest that the cathode off-gas should be reused rather than emitting it into the atmosphere, which would considerably reduce the GW potential of cathode off-gas of the PSMES. Other than that, acrylic sheet (22 kg CO_2 eq.) and electricity (4 kg CO_2 eq.) were also having significant impact in terms of GW. This can again be attributed to the release of CO_2 and CH_4 into the atmosphere during the production of acrylic sheet and electricity. For the impact category of aquatic eutrophication, carbon felt was having the highest impact of 8.04×10^{-3} kg PO_4 P-lim followed by acrylic sheet (2.17×10^{-3} kg PO_4 P-

lim) and electricity (0.68×10^{-3} kg PO$_4$ P-lim) (Fig. 4d). The release of phosphorus and phosphate into the water during the manufacturing of acrylic sheet and carbon felt is the reason for such a high impact of these components in the category of aquatic eutrophication. Moreover, the use of clayware membrane demonstrated a reduction in aquatic eutrophication due to the environmental offset of phosphorus and phosphate into the water during its production process.

3.5. Cost analysis, revenue generation and breakeven analysis

The capital expenditure linked with the construction of PSMES was 57.54$, among which more than 26 % was incurred by the cost of acrylic sheet used in its fabrication (Fig. S7). The next major component of fabrication cost was for manpower with 23 % of total capital cost, which was used for the PSMES fabrication. The manpower cost can vary based on the geographical location of the fabrication site and accordingly the capital cost and net revenue generation of the PSMES would vary. However, acrylic sheet, which was the major contributor of capital cost, can be replaced with other low-cost fabricating materials, which would reduce the fabrication cost of PSMES and in turn would increase the revenue generation of PSMES. Also, electrode materials incurred around 25 % of the capital cost, which is comparatively lower than the economic assessment performed by Jourdin et al. where they estimated it to incur around 59 % [39]. This can be ascertained to the fact that in the previous refereed investigation, metal based costly anodes were considered, whereas in the present PSMES, low-cost carbon-based electrodes were used.

From the cost of acetic acid produced in the extraction chamber of PSMES at the optimized operating condition, the daily revenue generation was 0.08$. Therefore, considering the daily operating cost of 0.03$, the net daily revenue generation was 0.05$. Thus, the yearly net revenue generation from the PSMES at optimized condition was 17.37$ that amounts to 69.47$ in four years. This shows that within four years, i.e., 3.32 years, the capital cost of the PSMES setup (57.54$) can be recovered back. Therefore, from four year onwards the PSMES would start generating revenue.

3.6. Impact of the present research and future directions

The research on PSMES system for producing acetic acid from CO_2 present in the biogas addresses two critical challenges: CO_2 sequestration and the simultaneous generation of value-added products. The PSMES was able to achieve a production rate of 70.55 g m^{-2} per day of acetic acid with a coulombic efficiency of 77.8 %. The system's life cycle and economic assessments revealed that while this technology has potential, there are key bottlenecks, particularly in terms of the environmental and economic costs associated with materials like acrylic sheets and carbon felt. Replacing these materials with greener, cost-effective alternatives could significantly improve the sustainability of this technology. The broader impact of this research lies in its ability to provide a scalable solution for CO_2 capture, turning a major greenhouse gas into a commercially valuable product. The pilot-scale operation offers crucial insights into the scalability of MES and identifies areas for improvement, potentially guiding future developments in bioelectrochemical systems. By bridging the gap between lab-scale setups and practical real-world applications, thus this research paves the way for more sustainable and economically viable carbon capture technologies.

To enhance the robustness of the current research on the design and operation of PSMES, insights from prospective life cycle assessment (LCA) and patent analysis techniques, can be incorporated [40]. Using prospective LCA would allow the study to assess the environmental impacts of PSMES not only based on present data but also by forecasting future developments in material and process innovations. This would address the uncertainties inherent in static, traditional LCA, providing a dynamic view of the potential environmental benefits as the technology matures. For instance, eco-design strategies could be applied to replace materials like acrylic sheets and carbon felt, which were identified as significant contributors to the environmental footprint, with more sustainable alternatives. By considering such changes early in the design phase, the environmental performance of PSMES could be significantly improved over time. Further, the robustness of the present research could be bolstered by integrating patent analysis to forecast future trends in technology and cost reduction. By analysing patent literature, it is possible to anticipate innovations in materials and design improvements, which could lower production costs. This method also provides a clearer picture of the economic scalability of PSMES by predicting future price trends for materials and equipment. Moreover, patent analysis could reveal industry trends that indicate the potential for broader commercialization, further supporting the financial viability of this technology. By leveraging these forward-looking assessment techniques, the study would provide stronger justification for both the environmental and economic feasibility of PSMES at larger scales, positioning it as a viable solution for carbon sequestration and sustainable chemical production in the future.

4. Conclusion

A first-of-its-kind PSMES was designed and operated for the concurrent production and separation of acetic acid from biogas-derived CO_2. This innovative system demonstrated the potential for addressing both carbon sequestration and value-added chemical production. The novelty of this research lies in the modular design, enabling simultaneous synthesis and extraction of acetic acid, which is a significant advancement over previous MES cells. Additionally, a comprehensive LCA and economic analysis were conducted, highlighting the importance of material selection for improving the system's environmental and economic sustainability. The main features of this research include the modular PSMES design, which offers flexibility in operation and parametric optimization, and the integrated environmental and economic evaluations, which provide valuable insights for future improvements. However, there are limitations that must be addressed for further scale-up and commercialization, which include the high environmental impact of specific materials used in construction, such as acrylic sheets and carbon felt that contribute significantly to both the economic and

environmental costs. Additionally, further research is required to enhance the overall efficiency and product yield of the system, particularly at larger scales. Despite these limitations, the study provides critical groundwork for the advancement of MES technology and its potential applications.

CRediT authorship contribution statement

Sovik Das: Writing – original draft, Visualization, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Venkata Ravi Sankar Cheela:** Writing – review & editing, Writing – original draft, Validation, Software, Formal analysis. **B.K. Dubey:** Writing – review & editing, Validation, Supervision, Software. **M.M. Ghangrekar:** Supervision, Resources, Project administration, Funding acquisition.

Data availability

Data will be made available on request.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Sovik Das is serving as the Associate Editor for Heliyon If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.heliyon.2024.e39950.

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