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Development of an MFC-biosensor for determination of Pb^{+2}: an assessment from computational fluid dynamics and life cycle assessment perspectives

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Abstract Microbial fuel cell (MFC)-based biosensor sensing has emerged as an innovative approach to in situ and immediate monitoring of substrate concentration. MFC-biosensor uses bioanode as a sensing element. In this study, the performance of MFC-biosensor, operated with Pb^{+2}, was studied at different hydraulic retention times (HRTs). The HRT ranges were 0.5, 1, and 2 days. The power density generation increased with the decreasing HRT. The highest achievable power density was obtained at HRT of 1 days with the density value of 597 mW/m^2. The power density produced in the MFC system was stored in the energy storage system. The computational fluid dynamics (CFD) method simulates detailed three-dimensional flow and heat transfer properties in reactors and provides information about potential reactor design. CFD was chosen to simulate the concentration distribution of the substrate in the MFC in different reactor type and different HRTs. It was observed that there was good turbulence in the reactor in a two day HRT and the reactor volume was used effectively. Life cycle assessment (LCA) was performed at 1 day with the highest power density. An LCA was implemented to the production and operation processes of a microbial fuel cell. According to the results, these two processes caused 4.23×10^{-6} loss of healthy years, extinction of 1.3×10^{-8} species in a year and loss of $ 0.33 source availability. The emissions to air, water, and soil were also calculated. These results showed that MFC-biosensor provided information on the rate of biodegradation processes.

Keywords Microbial fuel cell · Toxicity sensor · HRT · CFD · Life cycle assessment

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Introduction

Nowadays, conventional wastewater treatment plants (WWTPs) are being widely operated based on biological treatment processes. Heavy metals and toxic substances, which are industrial wastes, threaten natural habitats due to their high toxic content. Discharged toxic water including heavy metals or other toxic substances can bring about serious interruptions or serious damage to the biological process in WWTPs (Liu et al., 2014, 2021). The design and development of biosensors to determine the amount and type of toxicity in water is attracting importance due to the increased water pollution. Traditional monitoring of water quality is typically done by physicochemical methods. Currently, the chemical experiments for wastewater quality monitoring are conducted off-site and toxic substances cause detoriation in the microbial media of WWTPs until the results of the experiments are acquired. The development of online sensors become critical to detect toxic matter in wastewater and to ensure online early warning systems for timely prevention and resolution. For this reason, a sensor capacity of providing online monitoring of heavy metals and organic toxic substances should urgently be used as an early warning system to ensure the operation of WWTPs (Stewart et al., 2021; Tran et al., 2015). In order to preserve human health and ecosystems from hazardous waste water contaminations, early warning systems are continuously established to monitor water quality (Stein et al., 2010; Çetinkaya & Özkaya et al., 2019). Compared to the conventional water quality detection methods, prescreening of biotoxins in wastewater with microbial fuel cell (MFC) is rather quick, easy, and can easily be performed online as an early warning device. MFC technology provides an approach to the development of an inexpensive measurement (Yi et al., 2019; Zhou et al., 2018). MFC is a device that converts chemical energy in organics directly into electrical energy through the metabolic processes of microbes, and has been studied as a self-contained biosensor to monitor waste water quality (Munoz-Cupa et al., 2021). MFCs seems to be a potential solution to harvest electrical energy and neutralize organic wastes (Cetinkaya et al., 2017). Any toxic material can adversely affect the activity of anode eco-electrogens and cause a sudden drop in MFC voltage output. The conventional MFC consists of anode and cathode parts, which are established as independent regions divided by the proton exchange membrane (PEM). In the anodic compartment, organic compounds are oxidized by microorganisms and produce electrons and protons (Di Lorenzo et al., 2009). The performance of the MFC is pretented by many factors like configuration types, substrate source, temperature, pH, electrode materials, and HRT. HRT plays an important role in the design of wastewater treatment plants, the operating/investment cost, and energy requirements. In addition, HRT has a important impact on the power generation and wastewater treatment properties of the MFC (Cetinkaya et al., 2016).

Lead (Pb) is a heavy metal pollutant commonly found in industrial wastewater. High concentrations of Pb can cause important health hazards to living organisms such as anemia, hemolysis, and liver dysfunction (Jarup, 2003). According to the World Health Organization (WHO) and the US Environmental Protection Agency (US EPA), the permitted limit for lead in drinking water is 0.01 and 0.015 mg/L, repectively (Samuel et al., 2018).

The computational fluid dynamics (CFD) methodology is applied in diverse engineering practices for various purposes.The most common reason to use it in wastewater engineering is to resolve the behavior of the fluid within a chamber. Several similar studies utilized CFD to simulate the fluid flow inside thr test chambers (Toparlar et al., 2017; Kim et al., 2014; Yi et al., 2020). Several hypothetical application alternatives can be detected by CFD calculations and their pros and cons can be determined.

LCA is a holistic approach utilized for mainly environmental analysis of products and services (Cetinkaya & Bilgili, 2021). In 2006, International Organization for Standardisation (ISO) issued a series of standards and technical reports for LCA. ISO14040 and ISO 14044 consist of Principles and Framework and Requirements and Guidelines, respectively (Heijung & Guinee, 2012). The calculations are realized in the impact assessment phase.

In this study, the relationship between the power density and Pb^{+2} concentration and HRT on was investigated. Pb^{+2} concentrations in syntetic wastewater were measured using an improved MFC-based biosensor to demonstrate the accuracy of the self-contained device. The results were compared with experimental data and validated. CFD simulation was applied to model the fluid dynamics within the MFC-biosensor channel to gain insight into the relationship between HRT and sensor performance. The environmental effects of MFC-biosensor were investigated by LCA.

Materials and methods

MFC-based biosensor

The MFC-biosensor was made of kestamid that had anaerobic and aerobic compartments. The aerobic compartment was continuously ventilated with an air pump to supply oxygen. An external electrical resistence was connected to the anode and the cathode compartments and power density was measured through the resistance. The MFC reactor is made of Kestamide material with an analyte volume of 123 ml, and the compartments of the fuel cell are physically separated by a cation selective membrane (CMI7000 Membrane International INC.). The setup procedure of MFCs was explained in a previous study (Cetinkaya et al., 2017). Two following tests were performed on the same set of reactors. The MFC-biosensor was operated at room temperature as around 20–22 °C and 200 rpm using a magnetic stirrer.

Standard solution and anaerobic inoculum

A stock solution of Pb^{+2} (1000 mg/L) was prepared by dissolving the suitable amount of lead nitrate ($Pb(NO_3)_2$) in distilled water, was purchased from Merck (Darmstadt, Germany). The double-distilled water was used in the preparation of all the solutions. The concentration of Pb^{+2} was 500 mg/L. Synthetic wastewater was prepared with the following constituents: NH_4Cl, 6.4 mg dm^{-3}; KH_2PO_4, 30 mg dm^{-3}; $MgSO_4 \cdot 7H_2O$, 0.5 mg dm^{-3}; $CaCl_2 \cdot 2\ H_2O$, 50 mg dm^{-3}. Glucose was utilized as the carbon source (Do et al., 2021). The concentrations of Pb^{2+} were determined by atomic absorption spectrometry (Analytik Jena NovAA 300). MFC-biosensor were inoculated by mixing 10 mL taken from Golden Horn in Istanbul. All the cultures were carried out at room temperature of 25 °C.

Electrochemical analyses

Linear sweep voltammetry (LSV) of all the MFC-biosensor were tested by a potentiostat (Gamry interface 1000). Determination of Pb^{+2} was carried out by an atomic absorption spectrometry (Perkin Elmer Analyst 400).

CFD methodology

CFD modeling was executed by ANSYS Flent 2019 R2. The CFD calculations utilize Navier–Stokes equations to solve the flow characteristics. It must satisfy the convervation of mass and the conservation of momentum, which are given in Eqs. (1) and (2), respectively (Celikten et al. 2021).

$$\frac{\partial(\rho u)_i}{\partial x_i} = 0 \tag{1}$$

$$\frac{\partial \rho u_i u_j}{\partial x_j} = \frac{\partial P}{\partial x_i} + \frac{\partial}{\partial x_j}\mu_{eff}\left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i}\right) - \frac{2}{3}\mu_{eff}\frac{\partial u_k}{\partial x_k} + \rho g_i \tag{2}$$

The model parameters are given in Table 1.

The model generated 33,053 elements for actual cell and 35,344 elements for hypothetical cell.

The LCA

The products used in the system were selected from Ecoinvent database of SimaPro. In this study, SimaPro 8.0.2 version and ReCiPe impact assessment method was used. There are three endpoint categories: human health, ecosystem, and resources. Each midpoint category affect an endpoint category. The impacts of the product or system to human health are estimated by the help of unit disability-adjusted loss of life years (DALY), which means the loss of healthy years due to the various processes (Zhang et al., 2019). Among midpoint categories are climate change, ozone depletion, human toxicity, photochemical oxidant formation, and ionizing radiation affect human health (Cetinkaya et al., 2018; Cetinkaya & Bilgili, 2021).

Results and discussion

Response of current density to various HRT

LSV tests were carried out to evaluate the internal resistance composition of the anode to explore the

Table 1 Model options

Mesh options	
Element size	5×10^{-3} m
Max size	1×10^{-2} m
Model options	
Viscous model	Ꝁ-ε realizable
Near wall treatment	Enhanced wall treatment
C2-Epsilon	1.9
TKE Prandtl number	1
TDR Prandtl number	1.2
Convergence criteria	10^{-5}
Solution methods	
Pressure–velocity coupling scheme	Simple
Gradient	Least squares cell based
Presure	Second order
Momentum	Second-order upwind
Turbulent kinetic energy	Second-order upwind
Turbulent dissipation rate	Second-order upwind

main factors for diversities in anode performance. The current density produced at different HRT is shown in Fig. 1.

As shown in Fig. 1, the responses of MFC-biosensors operated on different HRTs (0.5, 1, and 2 days) showed significant differences. This results showed that the differences in anode performance cause the MFC-biosensor performance to be different. According to these results, it has been determined that the sensor responds to the Pb^{+2}. Figure 1 shows that the peak power density generation of 597 mW/m^2 occurred at HRT 1 day. As can be seen from Fig. 1, it is also seen that every change in HRT cause a spike in power density. Results of this study indicated that the optimal HRT was 1 day. The lowest performance was seen in the MFC operating HRT for 0.5 days.The sudden increase in power density may be due to the presence of some components in the spent inoculated which are easily degreaded by bacteria, thus causing an increase in the electron transfer rate. Higher power generation at lower HRT indicated that the MFC reactor was predominantly enriched with active electrogenic microorganisms which contributed to better voltage generation at lower HRT (Fazli et al., 2018). The highly electrochemically active bacteria resulted in increased electron transfer efficiency in the MFC reactor. The results were similar to that of other literature studies carried out by Liu et al. (2011) and Fazli et al. (2018).

Energy storage system

The biologic battery is not only energy-saving but also biodegradable and non-toxic. Glucose batteries that produce 20 times more energy than lithium batteries and do not damage the environment so systems that can use biological batteries in mobile phones should be developed. DC power distribution systems have become widespread as a result of improvements in semiconductor power electronics (Deeke et al., 2012). Given the advantages of DC power distribution and storage systems, it can be predicted that power distribution and management will be provided by DC systems in the future (Yu et al., 2015). Electric energy can be stored in the capacitors. Capacitors are devices that store energy by separating positive and negative charges. The capacitors consist of two conducting plates and the insulators which separate them and are called dielectrics (Wang et al., 2019). A typical MFC system produces about 0.7 V of energy. In studies with MFC system, a system with a power density of 10 µW/cm^2 produces approximately 10 s µW of energy. The operating voltage of the MFC system was determined from the polarization curve. The proposed controller keeps the energy storage system operational near the MFC's maximum power point. Figure 2 shows the energy storage system.

The generated energy is stored in the energy storage system. Then, the LED lamp was lit with this stored energy.

CFD analysis

The aim of this study was to analyze the flow distribution within the MFC under different flow conditions. Apart from the actual cell, we prepared a hypothetic cell with a different secondary influent option. We applied CFD model to visualize and

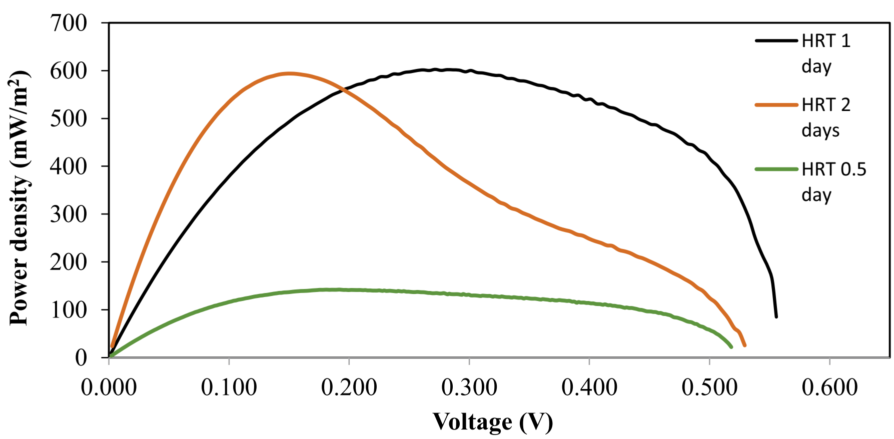

Fig. 1 Polaridation curves of MFC-biosensor at different HRTs

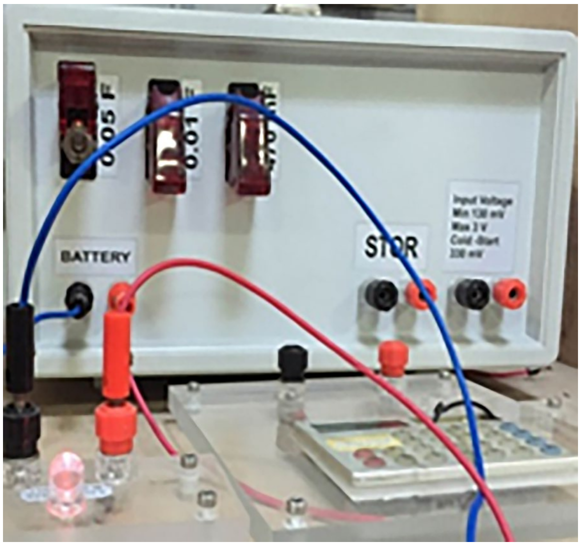

Fig. 2 Energy storage system

determine the best flow options. Figure 3 shows the streamlines and isosurface of 0.0001 m/s flow velocity for the actual cell at three different HRT.

CFD results indicated that the HRT 0.5 day was not enough to produce turbulance within the cell and most of the volume remained as dead zone. The dead zones were reduced with the increasing flowrates. However, it was observed that there was good turbulence in the reactor in HRT 2 days and the reactor volume was used effectively.

The current analysis was retested after the reactor modification. Another inlet was generated which is symetrical to the center of the cell according to the present inlet. The flowrate was divided identically into two. The results are shown in Fig. 3.

As with the simulation of the original cell, HRT 0.5 day was not enough to produce turbulance inside the volume. The solid volume with blue color in Fig. 4c shows the flow velocity having a value higher than 0.0001 m/s. The volume in the hypothetical reactor was lower than the physical reactor.

LCA results of MFC-based sensor

Three perspectives were identified for different time ranges. These time ranges were determined to understand the short-, medium-, and long-term impacts of the LCA analysis. These perspectives are used to realize an uncertainty and sensitivity analysis (Cetinkaya & Bilgili, 2021). ReCiPe also provides a wide range of emission types to air, water, and soil.

Table 2 presents the materials, which are used in LCA calculations, of MFC system.

Table 3 presents the results of damage assessment category of I (Individualist), H (Hierarchist), and E (Egalitarian) perspectives.

The calculations were repeated for different perspectives to realize a sensitivity analysis. While the H perspective is accepted as the default and base conclusion, E and I perspectives are used for verification and comparison (Cetinkaya & Bilgili, 2021).

Table 2 shows that the production and operation of a microbial fuel cell caused 0.000004228 years of healthy life loss for H perspective, which is developed for a 100-year period. Thus, it can be noted that the life cycle of a microbial fuel cell decreases the healthy years of a human by 0.000004228 years when the life expectancy is accepted as 100 years. Similarly, the life cycle of a MFC may threat 0.000000013 species in a year. According to the H perspective, which covers a 100-year period, 0.0000013 species may be lost during 100 years. Considering the resources, the life cycle of a microbial fuel cell cause $ 0.327 loss in resources' availability during 100 years.

I and E perspectives cover 20 and 1000 years, respectively. It can easily be seen in Table 3 that the values of I perspective are less for human health and resources; however, the impacts to the ecosystem are almost the same. The impacts to the resources of H and E perspectives are the same. On the other hand, it is clear that the harmful impacts to the ecosystem and human health is much higher in E perspective.

Table 4 presents the amount of the selected emissions to air, water, and soil. These emissions were selected due to their significant impacts among many other.

Carbon dioxide (CO_2), nitrous oxide (N_2O), and methane (CH_4) are known as greenhouse gases (IPCC, 1992). It is also known that nitrogen oxides (NO_x), particulate matter, carbon monoxide (CO), and non-methane volatile organic compounds (NMVOC) have adverse effects on human health and environment (Andreoni et al., 2008; AEA Energy & Environment, 2008; Kågesson, 1999; Haglind, 2008; Lauer et al., 2007; Schreier et al., 2007; Corbett et al., 2007). Besides, it is well investigated that waste heat also contribute to the global warming (Zhang et al., 2019). Arsenic, cadmium, chromium, lead, and mercury are known as the

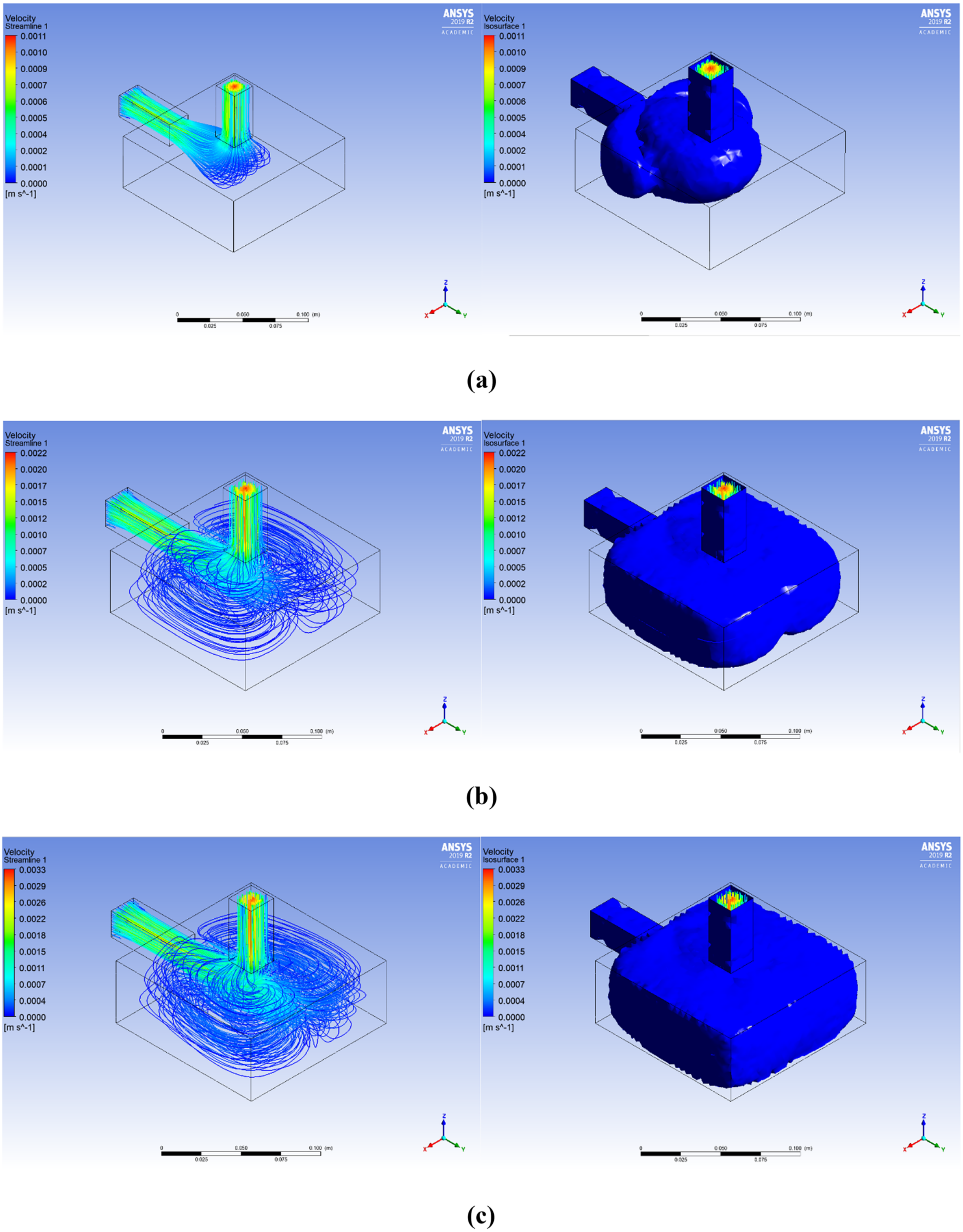

Fig. 3 Streamlines and isosurface in actual cell with **a** HRT 0.5 day, **b** HRT 1 day, **c** HRT 2 day

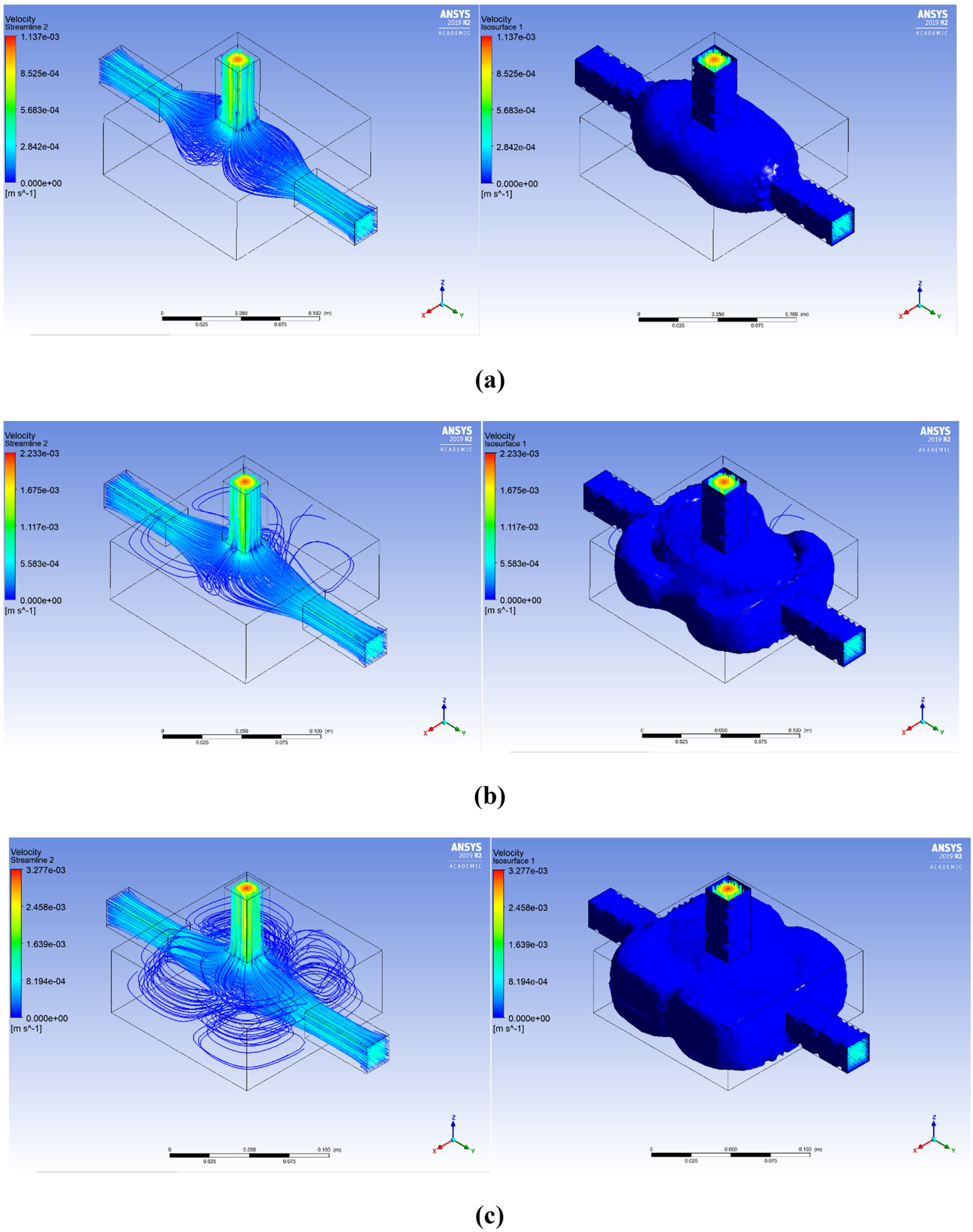

Fig. 4 Streamlines and isosurface in hypothetical cell with **a** HRT 0.5 day, **b** HRT 1 day, **c** HRT 2 day

Table 2 Materials used in MFC systems

Material	Amount	Unit
Titanium	0.0018	g
Silicone	13.75	g
Chromium steel	272	g
Carbon black	0.15	g
Polyethylene	36.89	g
Electricity production	18.72	Wh
Wastewater	86.4	dm^3

Table 3 Damage assessment results for I perspective

Damage Assessment	I	H	E	Unit
Human health	0.000003735	0.000004228	0.000019039	DALY
Ecosystem	0.000000014	0.000000013	0.000000065	species*yr
Resources	0.284047805	0.327778511	0.327778511	$

Table 4 The amount of the selected inventories

Compartment	Substance	Production and operation of a microbial fuel cell	Unit
Air	Carbon dioxide, fossil	1.26	kg
	Carbon monoxide, fossil	5.18	g
	Dinitrogen monoxide	29.14	mg
	Heat, waste	1.47	kJ
	Methane, fossil	4.10	g
	Nitrogen oxides	4.03	g
	Non-methane volatile organic compounds	1.02	g
	$Particulates_{2.5}$	2.94	g
Water	Arsenic	3.27	g
	Cadmium	1.15	mg
	Chromium	103.76	µg
	Lead	2.19	mg
	Mercury	93.76	mg
	Oils, unspecified	80.50	mg
	Solids, inorganic	171.21	mg
	Suspended solids, unspecified	3.09	g
	Water	49.04	mm^3
Soil	Arsenic	–4.44	µg
	Cadmium	–4.13	µg
	Chromium	–239.03	µg
	Lead	–327.99	µg
	Mercury	–6.15	µg
	Oils, unspecified	41.93	mg
Raw	Natural gas	142.56	l
	Forest occupation	784.95	cm^2
	Permanent crop occupation	16.56	mm^2
	Oil, crude	99.02	g

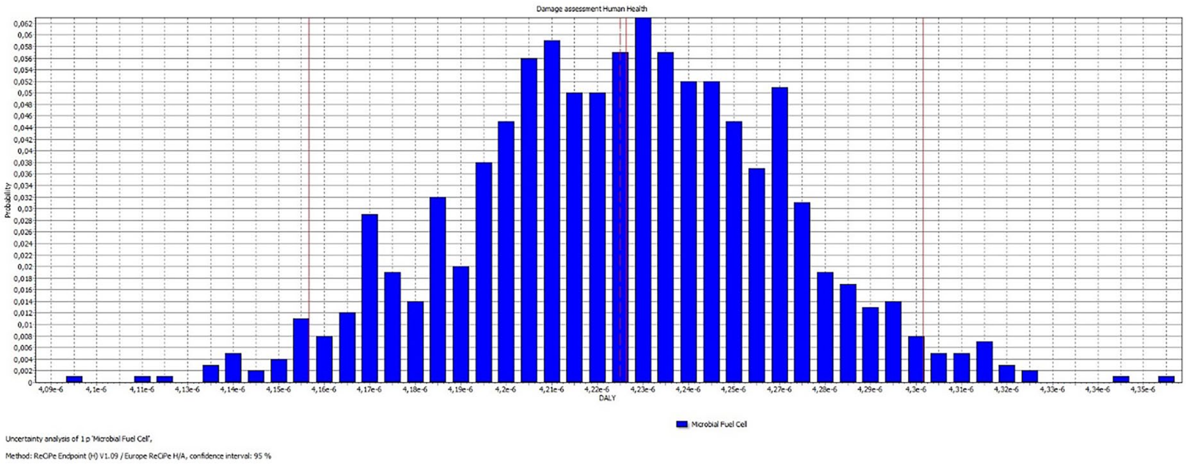

Fig. 5 Monte Carlo uncertainty analysis for human health

most hazardous heavy metals among all. The adverse effects of these metals were investigated in various studies (Aseth et al., 2018; Basketter et al., 2001; Gebel, 2001; IPCS, 1992; Taylor et al., 2019). Additionally, natural gas and crude oil consumption and occupation of forest and permanent crop area were also calculated.

Table 4 shows that the amount of emissions to air, water, and soil are very small; however, especially CO_2 production of the system during its life cycle is significant with a value of 1.26 kg. Besides, the electricity production of the system provides reduction of arsenic, cadmium, chromium, lead, and mercury emissions to soil. The land occupation and raw material consumption amounts are also very small.

In addition to these results, an uncertainty analysis was realized for damage assessment of H perspective. Monte Carlo method, which is already identified in SimaPro, was used for the analysis. The reason of utilization of uncertainty analysis is to identify the range of possible results of the system. Figures 5, 6, and 7 present the uncertainty analyses of human health, ecosystems, and resources, respectively.

Normal distribution was used for all analyses. While the –*y*-axis in Fig. 7 presents the probability, the –*x*-axis presents the cost results. The occurrance

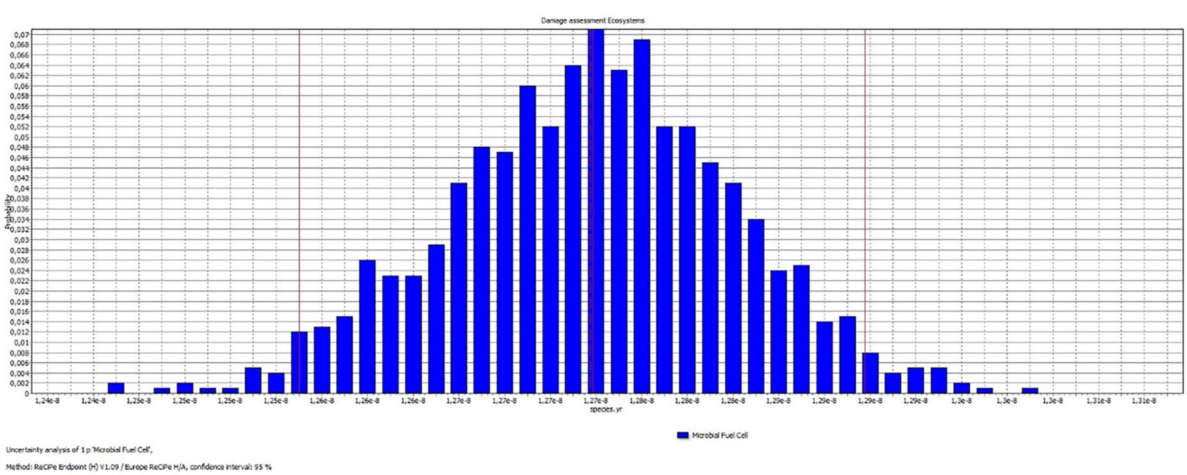

Fig. 6 Monte Carlo uncertainty analysis for ecosystem

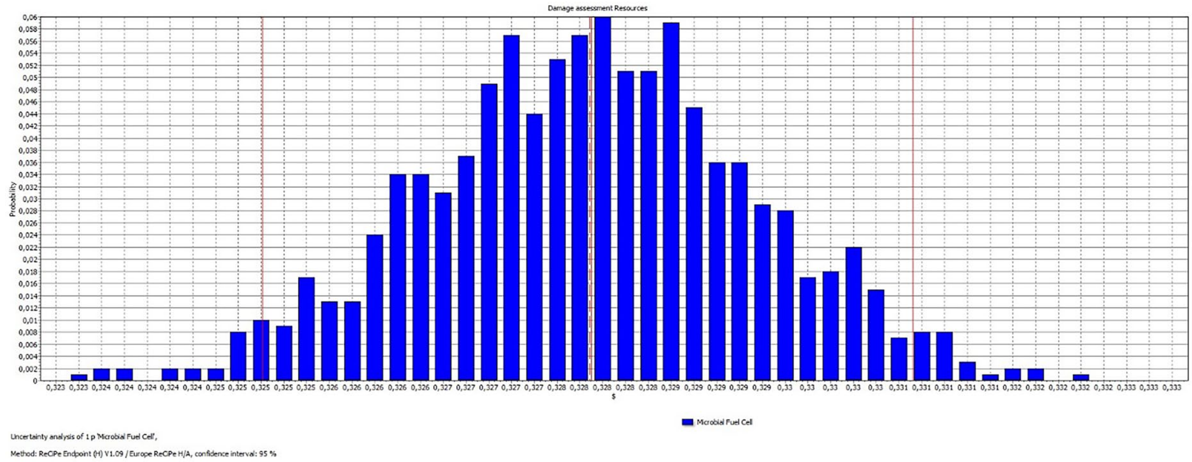

Fig. 7 Monte Carlo uncertainty analysis for resources

possibility of the values in the midst of the graph is much higher than the others. The confidence interval is 95%, which is the default value by the system. Calculating the likelihood of values in a confidence interval rather than relying to an exact value will provide better results for the reliability of the system. According to Figs. 5, 6, and 7, the median values of human health, ecosystem and resources are 0.00000423, 0.000000013, and 0.328, respectively.

Corbella et al. (2017) developed various scenarios on wastewater treatment systems and used 1 m^3 wastewater as the functional unit. Because the functional unit is very different from this study, comparing the results would not provide meaningful evaluations; however, it is worthy to mention this study. Zhang et al. (2019) also investigated on the LCA analysis of a microbial fuel cell. The authors considered all of the LCA phases, including the end of the life cycle of a cell, which is not considered in our study. According to the results, the greatest contribution to the global warming was from raw material extraction by 50.04%. On the other hand, it was also concluded that the end of life processes affect to the environment more than other processes.

Conclusion

In this study, it showed the improving of a MFC-biosensor for fast monitoring of the Pb^{+2} toxicity in the wastewater and MFC-biosensor that were analyzed via CFD simulations, and the optimum reactor design was obtained with orthogonal design. MFC-biosensor offers an innovative approach to reflect the real-time performance of the bioprocess with Pb^{+2} detection. Future research will be achieved to study the sensor reaction and improve a general kinetic model for toxic agents. The optimal performance for MFCs operation with Pb^{+2} was at HRT of 1 day according to highest current density achievement. But it should be noted that there is not a much difference between 1 and 2 days of HRT. On the hand, HRT 2 days were the most efficient design according to flow dynamics, which was achieved by CFD. The results showed that HRT play an important role on electricity generation performance in MFC-biosensor. According to the LCA results, the impacts of the system to the human health and the environment is very small. The production and operation of the system cause 0.000004228 loss of healthy years, extinction of 0.000000013 species in a year, and loss of $ 0.327778511 source availability. Besides, the amounts of emissions to air, water, and soil are at negligible level except CO_2 emissions. Although the units of other emissions to air and water are g, mg, and µg, the amount of CO_2 emission 1.26 kg, which is mainly caused by the production of stainless steel. Furthermore, electricity producing provide a reduction of emissions to the soil. The production and operation processes of the microbial fuel cell also caused a small amount of consumption of natural gas and crude oil.

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Data availability The data used to support the finding of the study are included within the article.

Declarations

Conflict of interest The authors declare no competing interests.

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