

Biohydrogen production from waste materials: benefits and challenges

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Received: 1 July 2019 / Revised: 28 September 2019 / Accepted: 29 October 2019 / Published online: 6 November 2019 © Islamic Azad University (IAU) 2019

Abstract

Escalating global energy demand has opened up a wide avenue for alternative energy research. One such alternative energy is biohydrogen (H_2) which is now projected as clean energy, since harnessed by biological means with high energy content; it finds the application on a broader scale. Recently, the employment of sustainable energy origins for generating biohydrogen has gained traction worldwide. Biohydrogen sourced from organic resources mainly of waste origins promises to provide sustainable energy in comparison with its other counterparts. The current work spotlights the various waste materials sourced for the generation of biohydrogen, bio-processing approaches, various microbes involved, conditions, factors, various relative advantages, and challenges. Diversities in biohydrogen processes such as utilizing different waste materials and biomass as raw material, probed akin to their chattels in the environment, bioreactor operative factors (temperature, pH, and partial pressure) are summarized. In this article, we have pursued to explicate the major hurdles confronted while procuring biohydrogen as a profit-making proposition by creating an appraisal of its improved role, also taking into account the diverse mechanism and procedures, while assessing its future perspectives.

Keywords H_2 yield \cdot Fermentation \cdot Organic waste \cdot Factors \cdot Biomass \cdot Energy

Introduction

In the current scenario, civilization mainly depends on energy. Nation's developments are sustained by their energy surplus, because it is the key indicator for its advancement economically. The energy dependence and reserve of country gains traction in the context of large investments needed to attain the growing energy demand. At present, only conventional fossil fuels are targeted as a major source to meet the global energy requirements. The scenario of growing deficiency of fossil resources and the incremental aggregation of greenhouse gases (GHGs) in the environment is no longer unavoidable as the situation already has surpassed the "critically high" threshold, and over dependence of fossil

Editorial responsibility: M. Abbaspour.

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fuel has made it as unsustainable (Singh and Rathore 2017). The emission of GHGs into the atmospheric environment owing to the increasing usage of fossil fuels attributes varied environmental threats like global warming, adverse and unpredictable climate shifts, biodiversity disruptions, ebbing of glaciers, and climb in sea level (Nikolaidis and Poullikkas 2017). Hence, the need for alternative green energy has become paramount. Sustainable energy origins are considered as an attractive surrogate to traditional fossil fuels and are predicted to be the central energy provider in the future that could augment the energy supply freedom with emission control and deliver a sustained profit for the farmers. Scientists are inspired by the attributes like renewable, sustainable, efficient, alternative green, and economically viable energy sources in order to overcome the burden sustained due to energy dependency, environmental protection, and viability focus on futuristic energy sources like biohydrogen (Gupta et al. 2013). Walsh et al. (2017) claim that amidst assorted available energy fount, hydrogen, biofuels, natural gas, and synthetic gas arise as leading, significant, ecologically prudent energy origins in the foreseeable time to come.

Hydrogen (H_2) finds broad spectrum in various purposes—being locomotive fuel and for power generation as



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it is a green energy bearer with huge energy load. Currently, fossil resources are supplying most of the hydrogen demand; these processes require high energy during hydrogen production (Argun et al. 2017). Hydrogen, as a fuel outshines other hydrocarbon fuels, because its energy efficiency high, it is recyclable, and it is considered as a green energy (Perera and Nirmalakhandan 2010). It has a high latent to be utilized in fuel cells for electricity generation due to its promising amount of energy content (140 kJ g^{-1}) (Oey et al. 2016). Various admirable attributes of hydrogen in contrast to conventional fuels eclipse its constraint in the generation mechanics (Staffell et al. 2019). Currently, hydrogen generation is limited as it is not easily available in nature and the production technologies are usually very expensive and unsustainable (Manoharan et al. 2019). Hydrogen as a fuel is either employed for direct combustion or in a fuel cell, by forming the by-product of water. Although a range of processes is applicable for H₂ production, all of them can be classified based on raw materials used into two leading divisions, specifically traditional and renewable technologies (Nikolaidis and Poullikkas 2017). In this current review, a pursuit has been contrived to appraise the current tendencies, the available technologies, processes and procedures in biohydrogen generation sourced from assorted organic waste ingredients, with a compilation of the merits and demerits of biohydrogen generation also briefly discussed.

Hydrogen production from biomass

All natural organic materials that are renewable, in extension to agronomical by-products, plant and trees, timber and wood debris, terrestrial and aquatic plants, grasses, animal residues (e.g., slurry or manure), or urban wastes (Kannah et al. 2019), etc., have been considered as biomass. Harnessing energy from biomass is an effective alternative due to zero net CO₂ effects than alternative to conventional feed-stocks (Das et al. 2008). This is because when biomass is converted into hydrogen energy it counterbalances the measure of CO₂ consumed amid flourishing cycle while biomass is formed. Biomass-derived fuels contributing CO₂ is considerably marginal than the CO₂ derived from fossil-origin fuels, which is accepted as characteristic biomass carbon balance.

A chief obstacle in discharge of biohydrogen as a suitable energy source is its dearth in its feature and the necessity for economically viable conversion methods (Chandrasekhar et al. 2015). Nevertheless, feedstock issues, such as cost, logistics and supply, etc., are main weakening issues which influence the overall economics of the biomass to hydrogen production techniques (Staffell et al. 2019). Biomass, which is suitable for biohydrogen production, is broadly divided into two categories, namely (1) bioenergy crops and (2) agricultural/wood-processing wastes. Based on



their origin, those can be further classified as plant- or animal-oriented biomass. In extension, amalgamated or pure microbial cultures were tapped for bioconversion of plant biomass to hydrogen, and the usual batch operational modes employed extensively have been recently substituted by continuous hydrogen production experiments (Salem et al. 2018). Figure 1 summarizes the different hydrogen production methods.

Hydrogen production from waste

Common anthropogenic activities are rejecting into the environment a wide range of materials in day to day practices. In the context existing energy scheme, global scientists progressively alter their focus from curbing pollution to resourcing waste for value added production like tapping green energy. Biological modes to treat wastewater are finding more traction, due to its versatile attributes like technical superiority, simplicity, economy, and ecofriendly. Waste materials used in the hydrogen production are segmented as agricultural waste, municipal waste, industrial waste, and other hazardous wastes. These are further compartmentalized as organic waste materials originating as or from food processing, crop residues, industry, animal manures, agricultural residue, domestic, and community wastes (Korres et al. 2013; Arizzi et al. 2016). Manipulating wastes as a probable source for H₂ generation has incited due interest for its sustainable nature and for opening new opportunity for the comprehensive use of everlasting renewable energy sources (Venkata Mohan et al. 2013; Zhang et al. 2007a, b; Venkata Mohan and Pandey 2013; Saidi et al. 2018).

Municipal solid wastes

Due to global population rise, municipal solid wastes (MSW) generated annually are rising and this escalation is disastrous. Cheap and abundant material availability is the key advantage of MSW over other wastes. In addition, it contains both macro- and micronutrients such as carbohydrates, lipids, proteins, minerals, and vitamins. These nutrient-rich MSW can be contemplated as a suitable source for hydrogen fermentation. Usually, direct fermentation of MSW proves to be less fruitful as various interfering agents hinder the process by reacting with the organic fractions (Lay et al. 1999). Since two-third of the organic component of MSW are bioprocessed for biohydrogen generation, many literature endorse MSW as the most potential source (Korres and Norsworthy 2017; Panigrahi and Dubey 2019).

Food waste

In general, food refuse can be designated as a credible source for generating energy, achieved through anaerobic

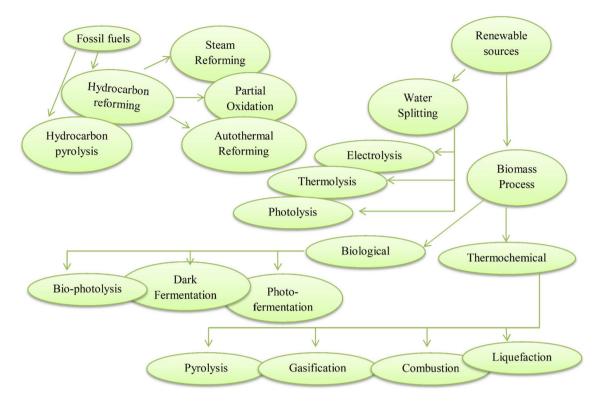


Fig. 1 Selective hydrogen production methods

degradation and also for its other beneficial attributes (Hwang et al. 2011). The physiochemical features of foodstuff rejects are key factors in developing a suitable process for biohydrogen production. Important parameters such as pre-operation processing of foodstuff rejects, ambient temperature, optimal pH and critical partial pressure determine the rate of biohydrogen generated and output (Dinesh et al. 2018). To harvest higher quantities of biohydrogen, after considering above factors, other aspects such as moisture load, volatile solid available, nutrients composition, and biodegradability are in addition paramount (Zhang et al. 2007). Biohydrogen generated from food rejects has been explored in large by various composite cultures from sources like anaerobic sludge, compost from various modes (Table 1).

Agricultural residual waste

Agriculture can easily be adjudged as the most profitable and viable process in the globe, and every year it leaves behind major volume of refuse, amounting to several billion tons, and this waste has carbohydrates in both simple and polymeric forms literally untapped (Ren et al. 2009). A significant portion of fruits and vegetables is lost or wasted during harvest, transport, and in the market, according to a Food and Agriculture Organization of the United Nations (FAO) analysis (FAO 2011, 2013). Several million tons of unmarketable vegetables could therefore be potentially sourced as biomass for dark fermentation, due to their carbohydrate content. Cellulose and hemicelluloses are the best degradable portions of the lignocellulosic matrix that can be used to produce biohydrogen by anaerobic flora (Menon and Rao 2012; Chatellard et al. 2017). Biohydrogen production from different agricultural residues by batch process is given in Table 2.

Animal generated waste

Disproportionate load of manure (and its slurry) produced from animal feeding operations were considered to be advisable resource for bioenergy production, but also found to be an incessant water polluter. Farmers can convert manure and slurry obtained at the culmination of the livestock management into biohydrogen by fermentation, which gives some financial along with positive environmental benefits (Sorathiya et al. 2014), but it has not been completely successful due to their chemical characteristics (Korres et al. 2013). Studies claim, in order to boost hydrogen yield, the animal waste can be helpful as a co-substrate due to its degradable nature and being a rich nutrient. The inclusion of an ambient pH along with microelements is needed for productive hydrogen



Substrate	Microorganism	Hydrogen yield	Fermentation mode	References
Distillery effluent	Coculture of <i>C. freundii</i> 01, <i>E. aerogens</i> E10, and <i>R. palustric</i> P2	2.7 mol H_2 mol ⁻¹ hexose	Batch	Vatsala et al. (2008)
Cattle wastewater	Mixed culture	12.4 mmol $H_2 g^{-1}$ COD	Batch	Tang et al. (2008)
Microcrystalline cellulose	C. acetobutylicum X9 þ and Ethanoligenens harbinense B49	$1.8 L H_2 L^{-1}POME$	Batch	Wang et al. (2008a, b)
Condensed molasses fermen- tation soluble	Coculture of <i>Clostirium</i> sporosphaeroides F52, and <i>C. pasteurianum</i> F40	1.8 mol H_2 mol ⁻¹ hexose	Batch	Hsiao et al. (2009)
Probiotic wastewater	Mixed culture	1.8 mol H ₂ mol ⁻¹ hexose	Batch	Sivaramakrishna et al. (2014)
Olive pulp water	Mixed culture	2.8 mol H_2 mol ⁻¹ hexose	Continuous	Koutrouli et al. (2009)
Cheese whey wastewater	Mixed culture	22.0 mmol $H_2 g^{-1} COD$	Continuous	Azbar et al. (2009)
Brewery wastewater	Mixed culture	6.1 mmol $H_2 g^{-1} COD$	Batch	Shi et al. (2010)
Vinasse wastewater	Mixed culture	24.9 mmol $H_2 g^{-1}$ COD	Batch	Fernandes et al. (2010)
Domestic sewage		$6.0 \text{ mmol H}_2 \text{ g}^{-1} \text{ COD}$		
Glycerin wastewater		$6.0 \text{ mmol H}_2 \text{ g}^{-1} \text{ COD}$		
Coffee drink manufacturing wastewater	Mixed culture	$0.20 \text{ mol } \text{H}_2 \text{ mol}^{-1} \text{ hexose}$	Continuous	Jung et al. (2010)
Purified terephthalic acid	Mixed culture	19.3 mmol H ₂ g ⁻¹ COD	Continuous	Zhu et al. (2010)
Cellulosic wastes	Anaerobic sewage sludge	3.6 mmol $H_2 g^{-1}$ cellulose	Batch	Lay et al. (2012)
Rice spent wash	Mixed culture	464 ⁱ	Batch	Roy et al. (2012)
Wheat straw	C. saccharolyticus	$3.6-4 \text{ mol H}_2 \text{ mol}^{-1}$	Batch	Willquist et al. (2011)
organic waste of composting plant	-	36 ml g V S ⁻¹	Batch	Boni et al. (2013)
Rice straw	Clostridium, Prevotella, Paludibacter, Ensifer, and Petrimonas	1.19 mol H_2 mol glucose ⁻¹	Continuous	El-Bery et al. (2013)
De-sugared molasses	Thermoanaerobacterium thermosaccharolyticum and Thermoanaerobacte- rium aciditolerans	132 ml $\rm H_2~G~VS^{-1}$	Continuous	Kongjan et al. (2013)
cornstalk	Anaerobic sludge	$12 L kg^{-1} TS^{-1}$	Batch	Liu et al. (2014)
Potato, pumpkin waste, other agro-industrial wastes	Anaerobic sludge	$46~\mathrm{ml}~\mathrm{H_2}~\mathrm{VS^{-1}}$	Batch	Ghimire et al. (2015)
Food waste	A. awamori and A. oryzae	85.6 ml g^{-1} food waste	Continuous	Han et al. (2015)
organic solid waste	Anaerobic granular sludge	$0.6 \text{ mmol H}_2/\text{gVS}$	Continuous	Castillo-Hernandez et al. (2015)
POME	Anaerobic sludge	$16 \text{ ml H}_2 \text{ gVS}^{-1}$	Batch	Suksong et al. (2015)
Whey waste	Clostridium sp.	$6.35 \pm 0.2 \text{ mol-H}_2 \text{ mol-}$ lactose ⁻¹	Batch	Patel et al. (2016)
Compost waste	-	$1.2 \pm 0.01 \text{ ml g}^{-1} \text{ VS}$	Batch	Arizzi et al. (2016)
Organic solid waste and distillery effluent	Anaerobic sludge	5.29 mol H_2 kg COD ⁻¹	Batch	Mishra et al. (2017)
Cheese whey and wheat straw hydrolysate	Anaerobic sludge	4554.5 H ₂ L ⁻¹	Batch	Lopez-Hidalgo et al. (2018)
Cottage cheese whey and fruit vegetable waste	Clostridium butyricum, Streptococcus henryi	$118.12 \pm 1.05 \text{ mmol } \text{L}^{-1}$	Continuous	Basak et al. (2018)
Vinasse	Oxalobacteraceae, Lacto- bacillaceae	$1.59 \pm 0.21 \text{ mol } \text{H}_2 \text{ mol}$ glucose ⁻¹	Batch	Sydney et al. (2018)
Sugarcane bagasse	T. thermosaccharolyticum MJ1	$6.2 \text{ L-H}_2 \text{L}^{-1}$	Continuous	Hu et al. (2018)
POME	Digested sludge	$2.45 \text{ mol-H}_2 \text{ mol-sugar}^{-1}$	Continues	Mahmod et al. (2019)

Table	e 1	Biohyc	lrogen	production	from	organic	wastes



Substrate	Microorganism	Hydrogen yield	Fermentation mode	References
Dairy industry wastewater	Biomass from fermenter	$2.56 \pm 0.62 \text{ mol H}_2 \text{ mol}$ carbohydrate ⁻¹	Continuous	da Silva et al. (2019)
POME	POME sludge	0.416 L H ₂ g ⁻¹ COD- removal	Batch	Mishra et al. (2019)
Beverage wastewater	Activated sludge	$72 \pm 31 \text{ mL L-d}^{-1}$	Continuous	Lay et al. (2019)
Complex food waste	Seed sludge	149 ml H ₂ g ⁻¹ Volatile solid added	Batch	Gadhe et al. (2014)
Food waste	Aspergillus awamori and Aspergillus oryzae	85.6 ml $H_2 g^{-1}$ substrate	Continuous	Han et al. (2015)
Waste bread	Aspergillus awamori and Aspergillus oryzae	7.4 L Ld ⁻¹	Continuous	Han et al. (2016)
Food waste	Anaerobic digestion sludge	174.6 ml H ₂ g ⁻¹ Volatile solid	Continuous	Cheng et al. (2016)
Food waste	C. butyricum	362 ml H ₂ g ⁻¹ Volatile solid	Batch	Kanchanasuta et al. (2017)
Food industry waste	Ruminococcaceae, Entero- bacteriaceae	$101.75 \pm 3.717 \text{ L H}_2 \text{ kg}^{-1}$	Continuous	Alexandropoulou et al. (2018)
Food waste	Mixed cultures	71.34 ml H ₂ g ⁻¹ Volatile solid	Batch	Rafieenia et al. (2019)
Waste wheat	Anaerobic mixed culture	$654.7 \text{ L H}_2 \text{ kg}^{-1}$	Continuous	Gorgec and Karapinar (2019)

 Table 1 (continued)

POME palm oil mill effluent

Table 2	Biohydrogen	production f	from different	agricultural	residues b	y batch process
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Substrate	Type of inoculum/pretreatment	Biohydrogen yield	References
Wheat straw	Seed sludge from H ₂ -producing CSTR	5.69 ml H ₂ g Volatile solid ⁻¹	Nasirian et al. (2011)
Wheat straw	Mesophilic anerobically digested sludge	10.52 ml H_2 g Volatile solid ⁻¹	Quéméneur et al. (2012)
Wheat stalks	Anaerobic digested activated sludge	23 ml H_2 g Volatile solid ⁻¹	Chu et al. (2011)
	Anaerobic digested dairy manure	37 ml H ₂ g Volatile solid ⁻¹	
Rice straw	Anaerobic sludge	24.8 ml H ₂ g Total dissolved solid ^{-1}	Chen et al. (2012)
Soybean straw	Cracked cereal acclimated in continuous stirred tank reactor (CSTR)	5.46 ml H ₂ g Volatile solid ⁻¹	Han et al. (2012)
Sunflower stalks	Anaerobic digested sludge	2.3 ml H ₂ g Volatile solid ⁻¹	Monlau et al. (2012)
Mixed vegetable and potatoes	Indigenous microflora	19 ml H_2 g Total dissolved solid ⁻¹	Marone et al. (2014)
Beet pulp	Seed sludge	90.1 ml H_2 g COD ⁻¹	Ozkan et al. (2011)
Fruit and vegetable waste	T. maritima	$3.46 \text{ mol mol}^{-1}$	Saidi et al. (2018)
Fallen leaves	Sewage sludge	$37.8 \text{ mL g}^{-1} \text{ VS}^{-1}$	Yang et al. (2019)
Agro-industrial waste	Consortium from Eisenia foetida lixiviated earthworm	232.72 ml H_2L^{-1}	Oceguera-Contreras et al. (2019)
Wheat straw hydrolysate	Escherichia coli WDHL	$269.2 \text{ cm}^3 \text{H}_2 \text{ g total reducing}$ sugars ⁻¹	Lopez-Hidalgo et al. (2017)
Sugar cane bagasse	Purple non-sulfur bacteria	$1.96 \text{ mol H}_2 \text{ mol sugar}^{-1}$	Mirza et al. (2019)
Rice straw	Anaerobically digested sludge	129 ml gCOD ⁻¹	Kannah et al. (2019)
Mixtures of agro-industrial wastes	Anaerobic granular sludge	4554.5 ml $H_2 L^{-1}$	Lopez-Hidalgo et al. (2018)
Various agricultural biomass residues	Anaerobic sludge	762.3 ml $H_2 L^{-1}$	Ren et al. (2019)
Mixed agriculture residue	Anaerobic sludge	472.75 ml $H_2 L^{-1}$	Li et al. (2018)
Corn stalk pith	Photosynthetic consortium	$2.6 \pm 0.3 \text{ mol } \text{H}_2 \text{ mol sugar}$ consumed ⁻¹	Jiang et al. (2016)



harvest. From a manufacture-economic perspective, the possible utilization of cheaper materials as auxiliary nutrients for biological conversion processes should be evaluated (Ahmad et al. 2019). Co-digestion procedure is generally used for increasing biogas production, since it provides balanced amounts of nutrients and a required buffering capacity, which also results in reducing the cost of nutritional supplements and pH control (Esposito et al. 2012). Recent studies demonstrated the increased hydrogen production with the addition of cattle manure as a carbohydrate-rich substrate. Manure is reported as a vital source for isolation of different efficient hydrogen-producing bacteria (Chatellard et al. 2017) and also reported as good biomass source for competent hydrogen fermentation (Ahmad et al. 2019). Wu et al. (2010) observed a high H₂ yield reaching 1.5 mol H₂ mol⁻¹ glucose at mesophilic temperature (37 °C). Table 3 includes reported results for hydrogen status with their main parameters dealing with co-fermentation of different animal wastes.

Wastewater

The conversion of organic wastes into hydrogen is an impressive strategy in both energy recovery and pollution control aspects. Due to deficiencies related to inhibition and microbial shift, very limited research has probed real wastewater as potential source for hydrogen generation (Hafez et al. 2009). Literature studies using wastewaters as substrate for hydrogen production via dark fermentation, including wastewaters of domestic use and from industries such as paper mills, starch and food processing, rice winery, palm oil mill, glycerol-based, chemical, cattle, dairy process, rice industry, winery, noodle industry, sugar processing, sugar beet and molasses manufacturing, etc., have been recorded

 Table 3
 Biohydrogen production from different manures and process conditions

Substrate/substrate pretreatment	Process condi- tions (g L ⁻¹ , °C, pH)	Type of process	Hydrogen production	References
Cow slurry/No	13.4, 37, 8.2	Batch	0.7 ml H ₂ gVS ⁻¹	Yokoyama et al. (2007)
Buffalo manure/No	8, 37, 6.7	Batch	$10.4 \text{ ml H}_2 \text{ gVS}^{-1}$	Concetti et al. (2013)
Buffalo slurry/sterilization	2.06%VS, 37, 6.5	Batch	$6.57 \text{ ml H}_2 \text{ gVS}^{-1}$	Marone et al. (2015)
Dairy manure/acid pretreatment	50.0, 36,7.0	Batch	$18.1 \text{ ml H}_2 \text{ gVS}^{-1}$	Xing et al. (2010)
Buffalo manure/sterilization	8, 37, 6.7	Batch	$37.7 \text{ ml H}_2 \text{ gVS}^{-1}$	Concetti et al. (2013)
Cow manure slurry/minced to 1-mm mesh size and passed through a sieve	6%VS, 60, 5.2	Semi- CSTR	$10.25 \text{ ml H}_2 \text{ gVS}^{-1}$	Wang et al. (2013)
Swine manure/shredding and filtration 1-mm mesh size	3.32%VS, 70, 6.7	Batch	$3.65 \text{ ml H}_2 \text{ gVS}^{-1}$	Kotsopoulos et al. (2009)
Buffalo manure/LPCW*/crude glyc- erol/(20/70/10)/sterilization	8, 37, 6.7	Batch	$170 \text{ ml } \text{H}_2 \text{ gVS}^{-1}$	Concetti et al. (2013)
Buffalo slurry/cheese whey/ (33VS/67VS)/sterilization	2.06%VS, 37, 6.5	Batch	117 ml H ₂ gVS ⁻¹	Marone et al. (2015)
Liquid swine manure/beet molasses/ (0.75/10 g/L sugar)/sieved and boiled for 30 min	12, 37, 5.4	Sequencing batch	1.57 mol $\rm H_2~mol^{-1}$ sugar	Wu et al. (2013)
Liquid cow manure/cheese whey/olive mill wastewater (5/40/55)/No	63.52, 37, 6	Batch	23.8 (0.64 mol $H_2 \text{ mol}^{-1}$ glucose)	Dareioti et al. (2014)
Liquid cow manure/cheese whey/olive mill wastewater/(5/40/55)/No	84.69, 37, 6	Continuous	$0.54 \text{ mol } \text{H}_2 \text{ mol}^{-1} \text{ glucose}$	Dareioti and Kornaros (2014)
Cattle manure/slaughterhouse risk material/(90 (wt dry matter)/10 (wt dry matter)) Heated at 90 °C for 3 h	40, 55, 7.1	Batch	33 ml H_2 gVS ⁻¹	Gilroyed et al. (2010)
Cow manure/milk waste/(30/70)/ Sieved and heat-treated/-	40, 55, 6.5	Batch	59.5 ml $H_2 gVS^{-1}$	Lateef et al. (2012)
Swine manure/fruit-vegetable waste/ [35(w/w)/65(w/w)]s Sieved/shredded in a blender	20, 55, 5.45	Semi-continuous	$126 \text{ ml } \mathrm{H_2} \mathrm{gVS^{-1}}$	Tenca et al. (2011)
Microalgae/Swine manure/ultrasonica- tion with enzyme pretreatment	5, 35, 7	Batch	116 ml $H_2 gVS^{-1}$	Kumar et al. (2018)
Buffalo manure/cheese whey/	4, 55, 4.8–5	Continuous	215.4 ml $H_2 \text{ gVS}^{-1}$	Ghimire et al. (2017)



(Balachandar et al. 2013). In relation to this, blending different wastewaters can serve as a compelling source and as a substrate in harnessing biohydrogen. For example, blending carbon-rich wastewater with nitrogen surplus wastewater may lead to improved hydrogen yield (Huang et al. 2010). Apart from this, a combination of wastewater and solid organic wastes, sewage sludge developed from the wastewater system can also be considered as an approach for biohydrogen production.

Ample accessibility and being cheap in comparison with other wastes are very strong attributes of wastewater or effluents. In spite of these attractive qualities, it has not intrigued researchers to serve as a source for inoculum considerations (Kotay and Das 2008). Hence, like MSW these also require pre-operation processing to make eligible this wastewater/ sludge suitable as a substrate for dark hydrogen fermentation, despite the rich methanogenic bacteria population present in it. Properly processed sewage sludge/wastewater usage can considerably bring down the expenditure of hydrogen generation and may also prove valuable and effective handling of these types of waste.

Fermentative hydrogen production

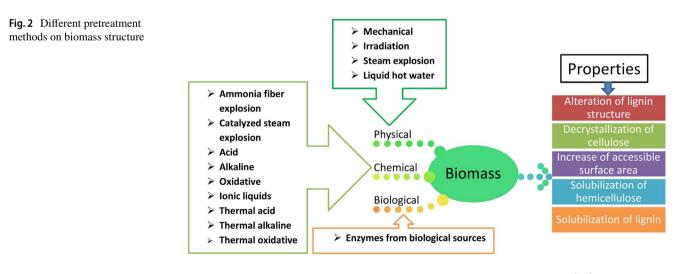
Usually, pretreatment of biomass can be assorted as physical and mechanical means, chemical route and via biological aide. Pretreatments could be employed in the context of biomass structure as a single process or a combination of various processes (Tu and Hallett 2019; Panigrahi and Dubey 2019). The impacts of pretreatment choice on the biomass structure are illustrated in Fig. 2. Reduction of size or disintegrating the biomass structure with the aid of a physical force is grouped as physical pretreatment. Usually, high-temperatures are accompanied with severe acidic or alkaline chemical pretreatment. Biological pretreatments, can be adept at using microorganism at ambient operating environment, but are less effective in their transformation rates and yield of monomers from the composite carbohydrates (Wang and Yin 2018). The intention of seeking pretreatment steps is usually to assist microbial approach the usable sugars within the biomass (Argun et al. 2017).

Hydrogen production by anaerobic bacteria mediated fermentation is renowned as more preferred biological routes, because of the ability of these organisms to produce the valuable biohydrogen energy from biomass and organic wastes (Sen et al. 2016). Different fermentation such as direct biophotolysis, dark, combined sequential dark-photo, and photofermentation were explored for H₂ production (Wang and Yin 2018).

In the direct biophotolysis process by involving solar energy, water molecules are split into hydrogen ions and oxygen. Hydrogenase enzymes convert these hydrogen ions into H_2 . Divergent cyanobacteria and variety of micro- and macro-algal species were explored for producing H_2 (Hallenbeck and Benemann 2002; Das and Veziroglu 2008; Holladay et al. 2009). Elimination of O_2 poses a challenge in this process, as it acts as an inhibitor of hydrogenase enzyme action and therefore hinders H_2 production (Miandad et al. 2017).

The indirect biophotolysis system comprises two stages. On the first stage, O_2 is discharged with CO_2 fixation and in the second stage H_2 is generated (Momirlan and Veziroglu 2005). The mechanism of direct biophotolysis can be borne in a single reactor achieving O_2 and H_2 generation in an fluctuating cycle or in separate reactors like open ponds and photo-bioreactors (Miandad et al. 2017).

In microbial electrolysis electrical current is spawned by the bio-electrochemical system that produces H_2 in action by reduction of protons are called bioelectro-hydrogenesis. The microbial electrolysis cell (MEC) constitutes four excerpts, which are, electronic separator, cathodic chambers, anodic chambers, and external electrical power source (Hamelers





et al. 2010; Miller et al. 2019). Domestic and industrial wastewater and agro-industrial residues containing cellulosic and starch biopolymers are utilized by this process to produce H_2 . Key factors such as microbial physiology and physico-chemical transport processes influence the performance of the MEC. Still the greatest threat is to retain the electrical potential in harmony at both the bioanode and biocathode chambers (Liu et al. 2005; Miandad et al. 2017).

The gram-positive bacteria were found encouraging, because under dark conditions they were reported for higher hydrogen yield and rate of biohydrogen accumulated. The endospore formation and fast growing nature of the microbes make them as a better choice for industrial applications. The volume of hydrogen harnessed from glucose by bacterial culture is influenced by metabolic pathway and finished products (Krupp and Widmann 2009: Gadhae et al. 2015).

In photofermentation operation, the existence of light is essential for the photoheterotrophic bacteria, to modify organic acids (e.g., lactic, butyric and acetic) to CO_2 and hydrogen covered by anaerobic conditions. Hence, at the time of acidogenic reaction, the formed organic acids are transformed into H₂ and CO₂ by these photoheterotrophic anaerobic microorganisms. The photofermenter system has to be constructed with appropriate dissemination of light in order to limit shading, higher surface area to volume ratio are mandatory in any externally lit up photo-bioreactor at commercial scale (Zhang et al. 2019). Overall chemical reactions involved in the above-mentioned biological H₂ generation compiled in Fig. 3.

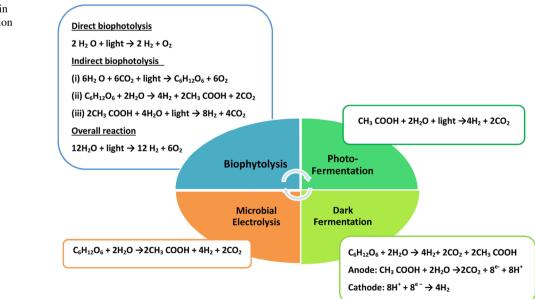
Sequential dark and photofermentation was a productive approach in biological hydrogen gas production. Dark fermentation and photofermentation can be connected because the refuse from dark fermentation was enough to source the organic acids needed for photofermentation which give rise to higher biohydrogen yield than the individual fermentation process (Zhang et al. 2019). Assorted microorganisms are competent in H_2 generation from any accessible renewable substrate covered under moderate environmental setting, which make such biological approach quite attractive compared with conventional process (Cai et al. 2019).

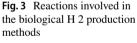
Factors affecting biohydrogen process

Biological hydrogen production mechanisms are not only environmentally friendly, but also inexhaustible (Benemann 1997; Greenbaum et al. 1983). In extension, the hydrogen output and the rate of hydrogen generation from various processes are shifting due to the outcome of some factors which are intricate in those procedures.

pH: pH is known to be one of the leading environmental components bearing upon the metabolic pathways and the hydrogen yield. During glycolysis, many facultative anaerobes are capable of generating hydrogen by the disruption of glucose to pyruvate. Metabolites generated midst breakdown of pyruvate, in turn, influence the hydrogen yield (Preethi et al. 2019). Metabolites that encompass supplementary hydrogen atoms such as ethanol and other alcohols are not exhibited in their corresponding acids. All the enzymes are dynamic only in a particular range of pH and have utmost activity at the optimal pH (Lay et al. 1997; Dinesh et al. 2018).

Carbon source: Carbon sources impact nitrogenase activity, which disturb hydrogen synthesis by cyanobacteria. Variations in electron donation competencies of nitrogenase are





influenced by varying concentrations of carbon sources and thus alter hydrogen production. During photosynthesis/fermentation, the starting load of glucose in the substrate was found to facilitate hydrogen yield (Gorgec and Karapinar 2019). The hydrogen generation decreased with increasing glucose concentration beyond 10 g L⁻¹ (Nagarajan et al. 2019; Dinesh et al. 2018; Preethi et al. 2019).

Temperature: Variable temperature range has been reported for harnessing biohydrogen: mesophilic (25–40 °C), thermophilic (40–65 °C), extreme thermophilic (65–80 °C), or hyperthermophilic (> 80 °C) (Preethi et al. 2019). Scrutiny of literature exposed that most of the investigations conducted at the level of laboratory scale have been carried out using mesophiles (Li and Fang 2007). The findings highlight the role of temperature as a significant effector on hydrogen generation (Balachandar et al. 2013).

Hydrogen Partial Pressure: Hydrogen partial pressure is one of the sensitive parameters to be considered while exploring generation of biohydrogen. Partial pressure of hydrogen in the system would get boosted if H_2 get aggregated in the headspace. As per Le Chatlier's principle, if hydrogen gets accumulated, the onward reaction will be largely hindered. Thus, the greater partial pressure of hydrogen negatively (Balachandar et al. 2013). Investigations have also highlighted that the partial pressure of hydrogen is a crucial aspect of extended hydrogen production (Nagarajan et al. 2019).

Volatile Fatty Acid (VFA): In fermentative hydrogen creation, metabolic end by-products were established for the decline in hydrogen yield. Ethanol, acetic acid, butyric acid, and propionic acid were found to form as the dominant end metabolites (Preethi et al. 2019). However, the medium ionic strength heightened with the increment of soluble end metabolites, which increased cellular lysis toward the stationary phase. To regulate the limiting issue of VFAs on hydrogen production, introducing soluble metabolites into the medium had an impact on substrate deterioration, the rate of hydrogen generation, and hydrogen yield (Kumar et al. 2019; Sydney et al. 2018; Balachandar et al. 2013).

Nutrients: It is crucial to add nitrogen, phosphate, and other inorganic trace minerals, amidst fermentation operation, to enable escalated hydrogen yield, while employing carbohydrate as a nutrient source for hydrogen production (Balachandar et al. 2013). Nitrogen is an indispensable component of amino acid synthesis and is required for optimal growth of the microorganism. In this scenario, Yokoi et al. (2002) demonstrated the suitable alternation of corn-steep liquor for sourcing nitrogen. Appreciable load of phosphate is also enticing for enhancing comprehensive achievement of the process and also for optimal hydrogen generation (Lin and Lay 2004). Additionally, the C/N ratio is crucial in sustaining the dark fermentation and influencing the hydrogen fecundity and specific hydrogen production rate (Lin and Lay 2004). However, maximal VFA retention in the system is not advisable as it switch the cellular reductants away from hydrogen transformation (Balachandar et al. 2013).

Gaseous Environment: Oxygen-susceptible attribute of the enzymes impact on hydrogen generation, it is mandatory to sustain an anaerobic atmosphere in the system. Impact of gaseous environment on biohydrogen production has been reported by various authors (Nagarajan et al. 2019). Generation of hydrogen was found to be naturally influenced by the existence of varying concentrations of inert or anoxic gases like argon, CO₂, and CH₄. Yoon et al. (2002) described increased hydrogen production by *Anabaena variabilis* when CO₂ was introduced repeatedly during the growth cycle, exposure to CO₂ heightened the levels of reductants impacting enhanced hydrogen yield during the process.

Metal Ions: Augmenting specific metal ions in the media are also paramount to any fermentation process. These metal ions are intricate in the cellular transport mechanisms and also participate as enzyme cofactors. According to Nicolet et al. (2010), hydrogenase is a key enzyme for hydrogen generation and it contains a bimetallic Fe–Fe center surrounded by FeS protein clusters. Many researchers consider the effect of supplementation of iron for biohydrogen production, during the glycolysis process. Voet et al. 1999 showed the role of magnesium ion as a critical cofactor for enzymes like hexokinase, phosphofructokinase, and phosphoglycerate kinase. In yet another article, Lin and Lay (2004) proved the effect of assorted trace elements such as Mg, Na, Zn, Fe, K, I, Co, Mn, Ni, Cu, Mo, and Ca for hydrogen production.

Hydraulic Retention Time (HRT): The volume and flow rate in a reactor, average duration of fermentation are important factors while considering, design, energy requirements, cost in operation, etc. Optimal HRT is paramount in the hydrogen generation process. Sourcing microbial cultures screened for their ability to sustain the mechanical disruptions created by the continuous volumetric flow is considered the paramount parameter in the overall process (Silva-Illanes et al. 2017; Lu et al. (2019).

Economics of biohydrogen production

Many countries have initiated the promotion of numerous ways for biohydrogen generation as it showed promise as a surrogate source of energy. Despite ample findings through scientific investigations and notable momentum in improving the rate of biohydrogen production, there are only limited research information available on its economics while considering avenues for commercialization (Kaushik and Sharma 2017). Construction and development of the bioreactor and the assembly systems designated for harnessing biohydrogen production methods were assessed by



important considerations involving various facets, including energy requirements, hydrogen yield, and production dexterity. Some of the cost estimates highlighted by different biohydrogen scientific explorers reported a provisional cost scenario which would aid in appraising the prospects of biohydrogen role as fuel for varied utilities in the future (Sekoai and Daramola 2015). Cost, performance, distribution and storage issues, environmental profits, national plan and policy and rules and legislation are the predominant concerns in introducing hydrogen as a fuel and also impact consumer choices, by stimulating the use of hydrogen (Nagarajan et al. 2019).

Challenges of waste-to-hydrogen energy production

In the recent years, focus on biological mode of hydrogen generation has extensively heightened among researchers. Yet, only a few studies have addressed the economic feasibility of commercial biohydrogen production. Dutta et al. (2005) showcased the lower price of photobiologically harnessed hydrogen much lower (\$25 m⁻³) in contrast to photovoltaic processed ($\$170 \text{ m}^{-3}$). Lee (2016) forecasted the cost of energy for biohydrogen will be sustained at 2.5\$/Kg and would compete well with fossil fuel cost in the future. Experimental studies favored dark fermentation as a cheap method; contrary to that photofermentation was a more efficient method, but it was found to be relatively pricier. The function of the indirect photolysis approach of hydrogen production was anticipated to be around 1220\$ per GJ/year, while the capital cost was predicted to be 2.4\$/gigajoule/year (Menetrez 2012; Ghosh et al. 2017).

Forecasts with respect to the deficiency of fossil fuel reserves in the current century impose energy scientists to focus on alternative renewable energy sources. The advantage found in current biological processes explored for generating biohydrogen is the high efficiency of conversion of various biobased waste materials into hydrogen energy. This finding has encouraged the H₂ production processes via these routes. However, to shift the economy from fossil fuel dependence to H₂ energy-based, efforts are needed to rectify the demerits of H₂ production pathway toward optimizing the production processes. As per Momirlan and Veziroglu (2002), the confronting issues in biohydrogen generation and usage are its higher processing cost, transport, stockpiling, distribution and delivery, lower conversion rates, and rudimentary stage in consumer utilization etc. The challenges in H₂ production and selective biological production methods are depicted in Fig. 4 and Table 4, respectively.

Threats in hydrogen supply avenues for divergent transport systems bank on, to a large degree, the form of storage facility available on board. While considering conditioning of hydrogen, both compression and liquefaction are



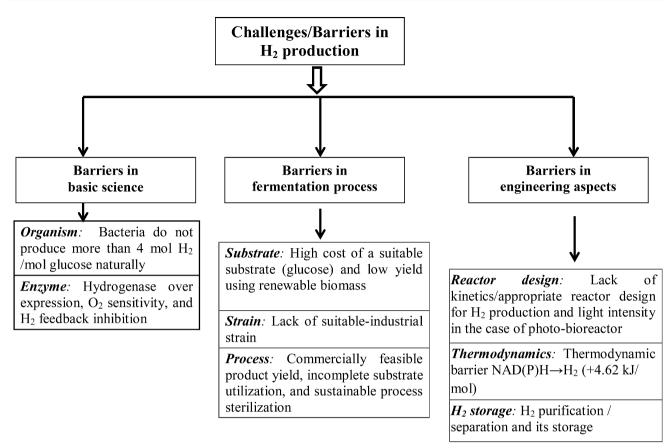
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considered, as they are also viable on the context of commercial feasibility, but committed for ample advancements and improvements in this avenue, but these factors poise as emerging threat against the demand of H_2 as an attractive transportation fuel. Important demanding issues in the introduction of hydrogen during transport are depicted in Fig. 5.

Importance of biohydrogen generation in under developing countries

Energy intensity is measured simply as the ratio of gross domestic products (GDP) has fallen faster in non-Organization for Economic Co-operation and Development (OECD) countries. In the last decade, the OECD countries appraised for 52% of global energy consumption. This average per capita energy consumption in OECD countries is four times higher than non-OECD countries and seven times higher than Africa (IEA Energy statistics 2007; Ahuja and Tatsutani 2009). At least one-fourth of the global population is unable to enjoy the advantages of modern forms of energy. The average energy consumption per citizen in OECD countries is measured as 8365 kWh, which is significantly more compared to Asia (646 kWh) and African countries (563 kWh). Forty percentage of people in developing countries are inaccessible to electricity although 40-60 billion dollars are annually spent on harnessing and consumption of electricity in these countries. Providing basic electricity to these people at an average consumption level (50 kWh/ person) would vastly impact the end-user demand. In low economic countries, limiting the demand for imported fuels and diversifying the domestic energy resource will invoke potential benefits. Biohydrogen-based energy can be a promising source of renewable energy technology to provide electricity at a minimal cost in low economic countries, wherever an extension of modern conversion technologies and process are implemented in a right manner (but can vary strongly with the impact of local conditions) (Ahuja and Tatsutani 2009).

However, biohydrogen generation in countries where inadequate economics prevail will impact societal norms like security on primary supply concerning the contribution for energy sufficiency, per capita GDP contribution, societal lifetime cost, etc. (Ren et al. 2013; Sun et al. 2010). Stanislaus et al. (2017) have reported that biohydrogen from digested sludge shows a positive energy balance. This indicated that biohydrogen can be a sustainable approach to reduce the negative impact of global warming with a low cumulative non-renewable energy demand. Singh et al. (2016) and Sekoai and Daramola (2015) suggested that hydrogen is the safest fuel due to its natures like non-toxicity and other positive attributes. Therefore, biohydrogen production is much needed in developing countries, because it is whispered that developing



(Bhutto et al. 2011; Kapdan and Kargi 2006; Das and Veziroglu 2001; Kotay and Das 2008; Hong et al. 2013)

Fig. 4 Challenges in H₂ production

Table 4	Major	challenges	in selective	biological	production
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Major challenges in		
Biophotolysis	Photofermentation	Dark fermentation
High cost	High cost	Inefficient and costly pretreatment methods
Large surface area requirement	Needs a high intensity light	Incomplete substrate conversion
Formation of explosive gas mixer	Low photosynthetic conversion efficiency	Difficult fermentive substrate utilization
Expensive photobioreactors	Expensive bioreactors	Low chemical oxygen demand (COD) removal
Low H ₂ productivity	Complex photobioreactor design	Difficult fermentive substrate utilization
Less than 10% solar energy utilization	Low productivity of nitrogenases	Incomplete substrate conversion
Oxygen intolerance H ₂ producing enzymes	Problematic practical applications	-
Problems in hydrogen recovery from reactor	Low solar energy utilization	
Low photosynthetic conversion efficiencies	Oxygen intolerant photobiological enzymes	



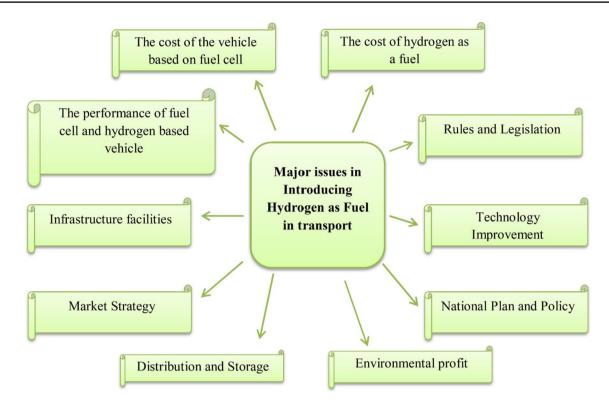


Fig. 5 Major issues in introducing hydrogen as fuel

countries are accounted for huge sharing of overall gas emissions and contribute to the negative environmental impacts (Ahuja and Tatsutani 2009). Also, biohydrogen will reduce the lifetime cost-competitive with gasoline vehicles in terms of vehicle retail cost, the externality cost of oil use, non-cost social transfers, etc. (Ogden et al. 2004; Rathore et al. 2019). This cost reduction will be a big boon impacting directly on the allocation of annual investment of countries, especially for low economic countries.

Future constraints of H₂ production

The world is witnessing a momentum in the development of technologies toward hydrogen energy generation. Harnessing hydrogen, dissemination, and utility have become important aspects of research, planning, and policy making. The carbon footprint impact of hydrogen from fossil fuel as well as other sources is more promising in comparison with conventional fuel processing (McLellan et al. 2005; Burmistrz et al. 2014). According to Derwent et al. (2006), substituting the fossil fuel dependence with biohydrogen would only have a climate impact on 0.6% of the current system. Numerous technologies can be used to produce hydrogen using primary energy sources (Balat and Kırtay 2010). Highthroughput investigations impart a vital role in biohydrogen fermentation operations, in order to attain reliable data for scale-up studies. Novel reactor designs with high levels of parallelization combined with online computer systems are required to evaluate the acute process setting during the procedure. Aid of mathematical and statistical tools in biohydrogen fermentation mechanisms is also crucial to assist analysis on the synergistic effects of various factors on the overall yield (Sekoai and Daramola 2015).

Application of biological tools in hydrogen production is the prevailing threat for biotechnology emphasizing on the present and unknown future environmental concerns. The potential scope of biological mode of hydrogen generation is not only resolved by scientific overtures (e.g., the genetic alteration of microorganisms for competency enhancement, designing of bioreactor) but also by economics, societal acceptance and the progress in systems for hydrogen energy (Singh et al. 2017). Several researches have to be investigated on the context of environmentally sustainable energy forms



substituting traditional fuels sourced through biomass and emerging organic wastes. Complete technology demonstrations are pivotal for hydrogen production from biomass to overcome major challenges to make economically competitive (Balat and Kırtay 2010). State-of-art blueprint such as boosting operation conditions like temperature, pH, OLR and HRT, bioreactor alterations, substrate choice, strain selection and nutrient enrichments, microbial immobilization and the metabolic construction of biohydrogen pathways, need to be channelized on enhancing biohydrogen procurement (Arimi et al. 2015; Soydemir et al. 2016). Development in fuel cell encourages rapid usage of hydrogen for domestic, thermal, industrial, and transport energy requirements. Nascent approaches are expected to emerge for hydrogen transformation, reduced rate, and cost when harnessed in industrial scale (Preethi et al. 2019).

Conclusion

Hydrogen in its free form is hardly unavailable in nature while comparing its counterparts; hence, the need for exploring new channels of the worthwhile generation of hydrogen. This review work comparatively evaluates and assesses preferred processes involving hydrogen harnessing methods against selected organic waste. Utilization of solid wastes like food waste, agricultural waste, animal waste, municipal waste, sewage waste, industrial waste, and wastewaters was found as attractive as well as feasible for biohydrogen production. Existing biohydrogen production processes are required to be modified for better fermentation, for unlocking new openings in biohydrogen production from renewable biomass. Large working reactor volumes, suitable tested environments, advanced technology, different storage, and transportation facilities are required to overcome the drawbacks like low yields and rate of hydrogen formation, while converting organic waste to biohydrogen. Developments in the field of biotechnology involving metagenomics approaches and genetic modifications are the recent technological advancements assist to make microbial assisted generation of hydrogen commercially viable, practical, and economically feasible in the near future. Although biohydrogen promises a lot as a potential fuel, further research and development of available current methodologies are the need of the hour, for improving the yield of biohydrogen and to validate its potential impacts, so as to consider hydrogen energy as a future sustainable energy source.

Funding The authors received no specific funding from any agency or organization toward making of this manuscript.

Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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