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Review: Nanoparticles can change (bio)hydrogen competitiveness

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ABSTRACT

Hydrogen has a negligible share on the global fuel market, yet it attracts a lot of investors. The main obstacle to the development of the hydrogen economy is its low cost-competitiveness. In order to meet energy demand and mitigate environmental damage, it is advisable to replace the existing fossil fuels with technologies that are more environmentally friendly and cost-competitive at the same time. Nowadays, some 97 % of hydrogen production comes from steam reforming of natural gas via energy that is obtained from fossil fuels. The production costs for 1 kg of hydrogen produced in this way, are between 2 and 4 €, while approximately 10 kg of CO_2 is emitted. The production cost of hydrogen produced by electrolysis from water is about 7 €, 80 % of which is electricity cost. The production of (bio)hydrogen (via photobiological and dark fermentation techniques) from biowaste using renewable energy sources has recently come to the fore. This review discusses use of various types of nanoparticles (organic and inorganic) in (bio)hydrogen production. A diversity of organisms, in pure as well as mixed forms, could perhaps produce (bio)hydrogen using pure (preferably simple form) carbohydrates and biowaste as a feedstock in the existence of various forms of nanoparticles. Furthermore, the (bio)hydrogen production potential (and cost), have indeed been reported to change considerably depending on what type of nanoparticles used as well as their dosage.

Keywords: Bioeconomy, (bio)hydrogen, waste management, nanoparticles, biowaste

1. Introduction

Rising population and industrialization are driving up global energy demand. Furthermore, due to the finite supply of fossil fuels, as well as the associated high price volatility and negative environmental impact, the world's attention is now focused on the search for alternative energy sources [**39**]. Numerous problems associated with fossil fuels can been solved by proper utilization of the renewable energy [**84**]. Among them, biofuels are regarded as a promising, sustainable, and viable renewable energy option in this context because they emit less pollution, easy to manufacture, extremely efficient, cost-effective, and eco-friendly [**37**]. Liquid and gaseous forms of biofuels include biodiesel, bioethanol, biogas, biobutanol, bioethane, biooil, biomethane, biohydrogen, and -bio-butane [**85**]. Biohydrogen has received substantial attention among some of the established biofuel production, since it offers numerous advantages [**71**]. Which would include the potential to deliver pollution-free byproducts as water vapours, with a highly energetic substance (120 kJ g^{-1}), the competence to use a huge variety of raw material as well as bacteria exist in multiple natural environments, and the ability to produce on a massive scale at sufficient temperature and pressure [**71,87**]. Biohydrogen can be fabricated through a wide range of biomedical applications including biophotolysis (directly and indirectly), microbiological electrolysis, and fermentation methods which including dark and photo-fermentation [**54**]. Various organic substrates and microbes are being used in the various biological routes for biogas generation [**24**]. Because of its elevated cellulose content, renewability, and wide availability, biomass is by far the most adaptable organic raw material and that can be used for biologically important biohydrogen production [**53**]. Regardless of the fact that biomass-to-biohydrogen production is the most environmentally friendly compared to biomass-to-biofuels technology [**86**]. A variety of methods have been proposed to enhance the enzyme activity in fermentation process, effectiveness, yield, as well as percentage of biohydrogen fabrication, but they were far from being sustainable from practical implementation in terms of economics [**71**]. Biophotolysis, dark-fermentation, microbial electrolysis, and photo-fermentation are all methods for producing biohydrogen production. Among such process steps, biohydrogen manufacturing through evolutionarily diversified dark fermentation as well as photofermentative microbes has been widely reported. The rapid advancements in the field of nanotechnology have been increased their possible applications in enhancing biochemical mechanisms [**57**]. It has been assisted significantly by the accessibility of nanoscale materials (NPs) with appropriate physiochemical properties [**14**]. Nanomaterials, unique properties, can play a significant role in determining the efficiency of biomass (microbes based or plant based) to biohydrogen production line. Characteristics such as elevated electroconductivity, high surface area, as well as a high surface to volume ratio can catalyse each phase of cellulose based biohydrogen manufacturing technologies [**18**]. Some of the notable nanoparticles, such as Fe, Ni, Cu, Au, Ag, and Ti have been shown to enhance the biohydrogen fabrication through a wide range of biological pathways, including bio-photolysis, photo and dark-fermentation [**6**]. Nanomaterials, particularly Fe and Ni, serve an important by acting as cofactors upon that active sites of hydrogenase as well as nitrogenase enzymatic reactions, massively improving the biohydrogen product yield [**76**]. Furthermore, nanoparticles behave as O_2 scavengers, attempts to remove unwanted O_2 existence during fermentation and thus lowering the redox potential. This creates an appropriate anaerobic condition for such action of the hydrogenase enzyme, resulting in increased biohydrogen yield [**10**]. Furthermore, using nanomaterials in the pre-treatment phase of biomass (lignocellu-lose enriched) might very well enhance lignin removal, increasing carbohydrate yield as well as speeding up the processing time [**3**]. In addition to that, nanoparticles can also improvise the production, thermal rates and pH consistency of cellulases for effective hydrolysis, i. e. the cellulose to carbohydrate transesterification reaction. The advances made in this area look into how using nanoparticles in a variety of biological routes for biohydrogen fabrication can aid in the integration of innovative and one-of-a-kind strategies for biohydrogen fabrication that are sustainable [**29**]. In this article, the ability of various types of nanoparticles (plant and microbes mediated) and their proportions upon that biohydrogen production through individual or microbial blend consuming carbohydrates as well as biowastes as nutrient were reviewed [**92**].

2. Biohydrogen production

Numerous strategies is being used to produce hydrogen via the biological pathway. Indirect and direct bio-photolysis, dark fermentation, photo-fermentation, and hybrid systems [encompassing dark fermentative as well as photosynthetic approaches] are examples (**Fig. 1**) of these [**32**]. The effectiveness or generate of biohydrogen fabrication during dark- as well as photo-fermentative fermentation is determined by the type of microbes as well as the active ingredients generated as final byproducts [74].

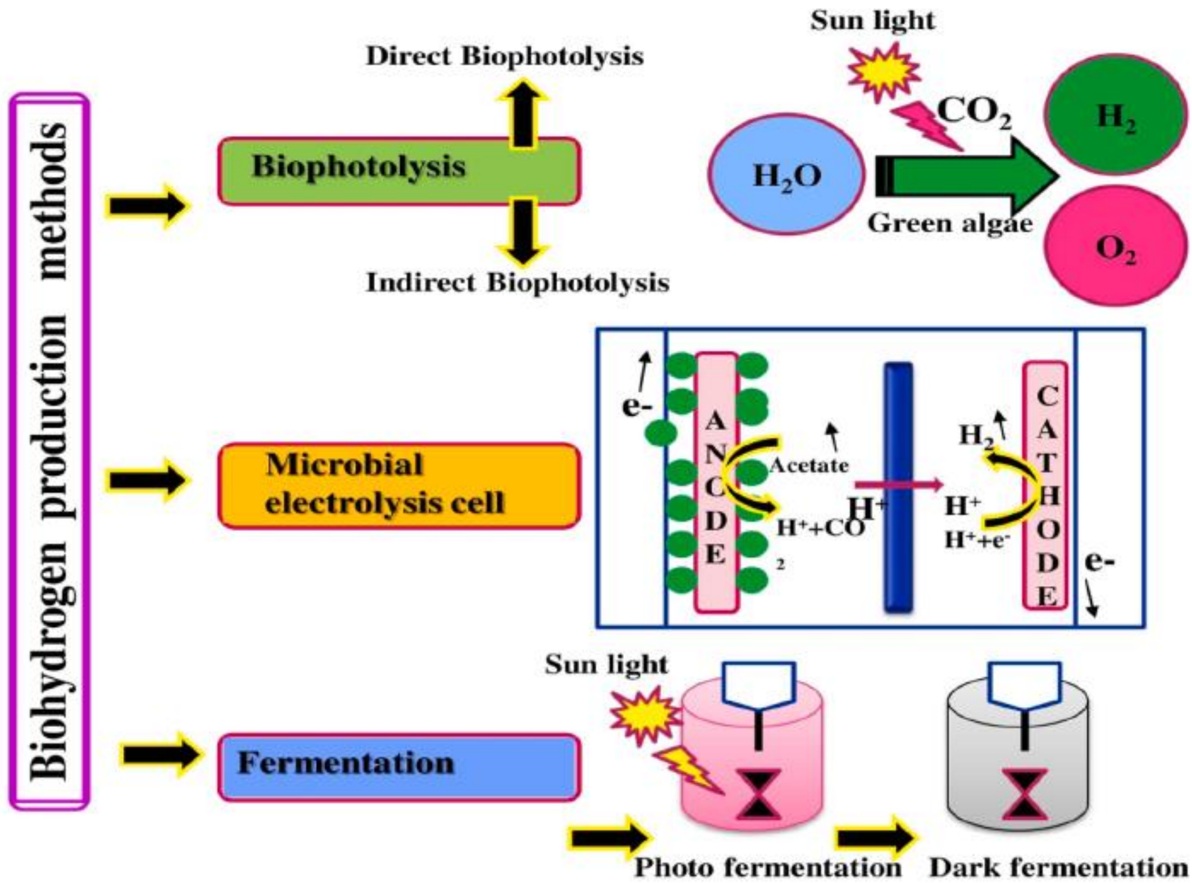

Fig. 1. Preferable methods used for biohydrogen production.

Pure carbohydrates are broadly used as nutrient for biohydrogen production through various microorganisms, including Citrobacter, Bacillus, Enterobacter, Caldicellulosimptor, Rhodobacter, Clostridium, Escherichia, Rhodopsedomonas, Thrmotoga, and Klebsiella under mesophilic as well as thermophilic circumstances [**15,67**]. Such organisms produce biohydrogen at rates ranging from 0.6 to 3.98 mol mol^{-1} of saccharides (e.g. hexose). The comprehensive utilization of various form sugars results in increased yields of 4 and 8 mol through the use of dark and photo-fermentative methods, respectively, which resulting in 12 mol biohydrogen mol^{-1} of saccharides [**4**]. Only one-third of the substrate was used for biohydrogen production via the dark-fermentative system. Hence, dark-fermentative system's reduced energy conversion potential leads to practical constraints at the mass production [**31**]. Furthermore, the increased cost of nutrient proves to become a massive obstacle. Even with the perfect combination of biohydrogen manufacturers from dark as well as photo-fermentative methodologies, highest biohydrogen production comparable to stoichiometric output of 12 mol biohydrogen mol^{-1} of saccharides (eg. hexose) is not possible [**43**]. The synthesized enzyme mediated sequence framework demonstrated the effectiveness of generating biohydrogen close to the highest theoretical yield while using sucrose as a nutrient [**71**]. Hence, the high expense of enzymes applied during cascaded systems, combined with their poor constancy, is a serious obstacle to commercial quantity of fabrication. Because their chemically complex existence, there seems to be a top consideration for the development of improved biohydrogen fabrication systems that use bio-waste as a feed [66]. Numerous primary treatment methodologies, such as physical, chemical, and microbial, are being used to improve biomass hydrolysis and biohydrogen fabrication. Amongst which, microbiological pretreatment of organic matter (biomass) appears to be the most cost-effective strategy for biohydrogen fabrication, as it can continue operating in unsterile conditions [**59**]. The enzyme catalyzes for producing biohydrogen through microbes emerges either through the disposal of surplus reduction counter parts or as a byproduct of nitrogen fixation [**93**]. Nitrogenase as well as hydrogenase catalysts in dark- as well as photo-fermentative microbes catalyse these reactions (**Fig. 2**) [**75**]. Hydrogenase enzymes have been divided into 3 groups according to the type of metal atoms existing in their catalytic site: as [NiFe]-, [FeFe]-, as well as [Fe]-hydrogenases.

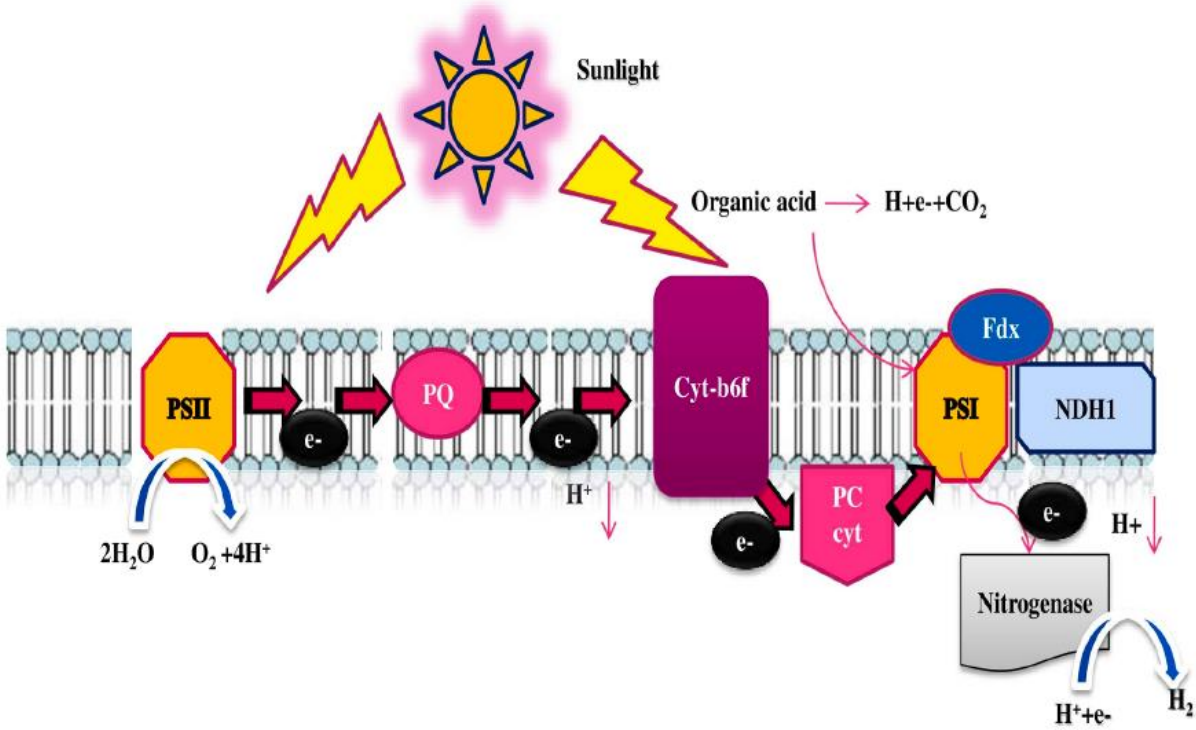

Fig. 2. Influence of nanoparticle on nitrogenase enzyme for the enhance biohydrogen synthesis under photofermentation.

Because of their effectiveness in improving hydrogenase action, the existence of Fe and Ni metal ions has a significant impact on either pure or mixed culture on biohydrogen production [**7**]. Recently, the use of metallic nanoparticles has been proposed to overcomes the constraint of limited biohydrogen fabrication under dark-fermentative processes through: (1) improved intracellular electron transport, (2) helping the H2 actually creating [NiFe]- and [FeFe]-enzymes, as well as (3) their anti-microbial characteristics to enhance preferential H2 producers in the microbial consortium [**42**]. The bioavailability of metallic particles, on either hand, is a main consideration for H_2 producers. Moreover, biosynthesized nanoparticles emerged to become a more promising method than physical as well as chemically synthesized nanoparticles [**19**].

3. Nanoparticles' impact on biohydrogen fabrication

The use of nanoparticles for various applications including biofuel production, protein immobilisation, and biosensors has grown considerably. Nanoparticles can also have a significant impact on microbial energy metabolism for biohydrogen production under aerobic conditions by transferring electrons efficiently [**69**]. Thus, an optimistic influence of multiple NPs (Ag, Au, Cu, Fe, activated carbon, Ni, Pd, SiO_2, Ti, carbon nano-tubes, and composite) on biohydrogen fabrication has been reported [**26,56**].

Concisely, such nanoparticles may be stimulating biohydrogen fabrication due to their surface as well as quantum size effects [**63**]. As a result of the surface influences, the relatively small dimensions of the nanoparticles, the greater the specific surface morphology and thus the greater the potential to adsorb charged particles [**60**]. The size of nanoparticle is proportional to velocity of e-(electron) transport among nanoparticles as well as enzymatic substances such as hydrogenase, which appears to catalyze the transformation of hydrogen to proton as well as vice versa, whether as electron drains or to generate reducing energy from oxidation, as $H_2 \leftrightarrow 2H^+ + 2e^-$ [**74**].

4. Various form of nanoparticles involved in biohydrogen production under the stimulation of microbes

4.1. Organic nanoparticles influence on biohydrogen production using microbes

The **Table 1** shows the effects of individual organic nanoparticles as well as nanoparticles blends on biohydrogen fabrication yield in various organisms. Carbon nanotubes are a unique tubular carbon-based material. Because of its effective role in reducing oxidation reaction and electron transport possibilities, it has been recognized application forms in biosensors as well as microbial biofuel cells [**52,60**]. Anaerobic conditions without carbon nanotubes drained out of the reactor after 2 weeks of operating condition under similar circumstances. In comparison to activated carbon, the carbon nanotubes based up flow anaerobic sludge blanket (UASB) reactor produced 1.7 times more biohydrogen yield. Correspondingly, activated carbons in granular form as well as powder forms demonstrated a considerable improvement in biohydrogen production, with 94 % as well as 44 % respectively, utilizing acidogenic culture using starch effluent as nutrient [**44**]. Activated carbon at dosages of 33 and 33.3 mg L^{-1} was found to be very efficient in increasing biohydrogen production yield using sucrose by 62.5 % and 730 % under batch as well as UASB modes, respectively [**77**]. The reduced biohydrogen production yield by microbe to expected calculations of 4 mol mol^{-1} of hexose as well as the increased feed expenses are the major constraints for huge quantity production under dark-fermentative circumstances [**78**]. Hence, numerous methodologies have been implemented to tackle these issues, including the parameter optimization, the screening of efficient biohydrogen producers, through the use of minimal nutrient, including organic wastes material [**61**]. The hydrogenases are important enzymes in the biohydrogen production, as well as their activity was considerably influenced by the metal ions (Fe^{2+} and Ni^{2+}). Recent time, the nanoparticles aspects of such metals, among others such as activated carbon, CNTs, Ag, Pd, and so on caused a 6.7 fold rise in biohydrogen yield [**21**]. In this case, the increase in biohydrogen production is related to the dosage and attributes of nanoparticles. CuNPs, on the other hand, had a detrimental impact on biohydrogen fabrication at minimal (2.5 mg L^{-1}) doses [**46**]. The differential effects of nanoparticles or their blends on biohydrogen production were demonstrated employing pure as well as mixed cultures and using both sugars as well as bio-wastes [**49**]. Such nanoparticles primarily increase biohydrogen yield by having a strong positive influence on microbial growth, nutrient degradation potential, as well as the transitional metabolite characteristics [**28**]. Predominantly, in the existence of nanoparticles, biohydrogen producers shift transitional metabolites toward an elevated acetate-to-butyrate ratio and inhibit ethanol as well as propionate fabrication [**34**]. Using anaerobic sludge as a mixed microbial population, a combination of Fe and Ni nanoparticles was found to be good for significantly increasing the -biohydrogen yield for up to 200 % [**25**]. Correspondingly, unidentified mixed consortia demonstrated a highest increase in biohydrogen yield of up to 666 %when SiO_2 nanoparticles were used [**44**].

Table 1 Organic nanoparticles and mixture of inorganic nanoparticles influence on biohydrogen production through various biological process and microbes.

Organisms/Produced by	Nanoparticles	Dosage (mg L^{-1})	Nutrient	Biohydrogen yield (%)	Process conditions			Reference
					pH	Mode	T (C)	
Anaerobic Sludge (AS)	Activated carbon (powder form)	33	Sucrose	62.5–73.0	5.5	Batch/ UASB	37	[77]
Anaerobic Sludge (AS)	CNT	100	Glucose	NA	6.5	UASB	25	[38]
Acidogenic consortium	Activated carbon (powder form)	5000	Starch effluent	44.0	5.5	Continuous	28	[60]
AS	Activated carbon (Granular form)	100	Glucose	NA	6.5	UASB	25	[38]
Acidogenic consortium	Activated carbon (Granular form)	10,000	Starch effluent	94.5	5.5	Continuous	28	[60]
AS	Fe2O3 NPs + NiO NPs	200 + 5.0	Molasses effluent	62	5.5	Batch	37	[13]
AS (AS)	Ni + Fe	37.5 each	Starch	200	7.0	Batch	37	[51]

Throughout many cases, nanoparticles synthesized using physical as well as chemical methods have been used in the biohydrogen production. Because biologically synthesized NPs are much more biocompatible than those synthesized using physical as well as chemical methods, more studies in this field is required for their potential application in biohydrogen production [**62**]. The correlation of anaerobic digester microbiomes for biohydrogen fabrication in the existence of nanoparticles proposed that microbial framework had been significantly altered [**79**]. Hence, such method can be modified to enhance the enhancement of precise biohydrogen producers during in the fermentative system. Ideally, nanoparticles seemed to be more efficient during biomass pre-treatment as well as immobilisation of whole microbial cells used for biohydrogen production, suggesting their wide range of applications [**68,22**]. Additionally, assimilation of dark-fermentative biohydrogen fabrication with other processes such as photo-fermentative, methane production, and polyhydroxyalkanoates (PHA) has been proposed as being more successful methods in multistage systems to enhance method economic system [**33,64**]. Thus, more interdisciplinary processes are required to assess the function of nanoparticles. These methods can also be used effectively to enhance multistage systems employing methane and simulated biofuels to generate further bioenergy including methanol and significance bio products utilising methanogenic organisms for long-term development [**62**].

4.2. Effect of nanoparticle blends on biohydrogen fabrication

Influence of individual metal nanoparticles for hydrogenase activity on microbes and biohydrogen production have been reported widely [**12,73**]. Thus, the influence of nanoparticles blends for biohydrogen fabrication receiving more attention among researchers (**Table 1**). Accordingly, the nanoparticles mixer such as Fe and Ni nanoparticles with various concentrations (0-50 mg L^{-1}) showed considerable improvement in the biohydrogen production using the starch as nutrient [**25**]. At 37.5 and 37.5 mg L^{-1} dosage of Fe and Ni respectively, showed increased biohydrogen production 150 l kg was observed. Correspondingly, the combined effect of NiO as well as Fe_2O_3 nanoparticles on biohydrogen production through anaerobic sludge with molasses as well as dairy effluent was investigated [**12**]. The highest biohydrogen production was reported from dairy industrial effluents as 17 mol kg^{-1} of COD in the existence of Fe_2O_3 (50 mg L^{-1}) as well as NiO (10 mg L^{-1}), correspondingly. The blend of nanoparticles resulted in a 27 % increase in biohydrogen yield when compared to controls [**12**]. Surprisingly, the optimal addition of such nanoparticles reduced (3.6 to 2.8 h) the lag phase of biohydrogen production. The size as well as surface area of nanoparticles considerably involved in the biohydrogen production through enhancing the activity of various enzymes such as ferredoxin oxidoreductase, hydrogenase, and ferredoxin [69]. Extent biohydrogen fabrication, on the other hand, had been demonstrated to be around 9 mol kg^{-1} of COD employing 200 mg L^{-1} of Fe_2O_3 as well as 5 mg L^{-1} of NiO nanoparticles from complex distillery effluent [**13**]. In this case, a 62 % increase in biohydrogen yield was obtained when compared to control. Surprisingly, the rate of biohydrogen production was increased by 221 % in the existence of nanoparticles mixtures.

4.3. Inorganic nanoparticles influence on biohydrogen production using organism

The physical, chemical, and biological processes have all been used to produce multiple types of nanoparticles for use in biohydrogen production [**30**]. Among such conceptual point of view, the eco-friendly nanoparticles synthesis via biological approach utilizing microbes and plant extract were proposed as an appropriate alternate strategy to the extreme conditions used in physical as well as chemically synthesized nanoparticles for biohydrogen production processes [**70**] (**Table 2**).

4.3.1. CuNPs

The nature and concentration of nanoparticles determine their influence either positive or negative on biohydrogen production through the presence of microbes [**69**]. According to this the CuNPs inhibited biohydrogen fabrication by $C.acetobutyricum$ as well as $E.cloacae$ strains across wide range of concentrations of 2 to 12 mg L^{-1} which use glucose (pure form) as nutrient.

Table 2 Inorganic nanoparticles influence on biohydrogen production through various biological process and microbes.

Organisms/Produced by	Nanoparticles	Dosage (mg L^{-1})	Nutrient	Biohydrogen yield (%)	Process conditions			Reference
					Optimal pH	Operating mode	Optimal T (C)	
C. acetobutylicum	CuNPs	2.5	Glucose	3.5	7.0	Batch	37	[46]
Bacterial consortium	AgNPs	20.0	Glucose	67.6	8.5	Batch	35	[81]
AS	AuNPs	1 mM	Acetate	NA	7.2	Biofilm reactor (electrochemically active)	35	[27]
Enterobacter cloacae, AS, *Clostridium butyricum*, *Rhodobacter sphaeroides*, *Escherichia coli*	FeNPs	5.0–323	Glucose, water hyacinth, Malate	19.4–100	5.5–7.0	Batch	32–37	[41,8]
C. acetobutylicum, *E. cloacae*, *Clostridium pasteurianum*, *E. aerogenes*, AS, *microbial consorrtium*	Fe_2O_3NPs	25–800	Glucose, sucrose, Cassava starch, dairy, molasses, and starch effluent,	4.8–63.1	5.5–7.0	Batch	35–60	[21,11,60]
AS	NiNPs	2.5–60	Glucose, sugarcane bagasse effluent	0.90–23.0	5.5–7.0	Batch	37–55	[47,73,9]
AS	NiONPs	4.8–23.5	Glucose, dairy and molasses effluents	5.0–200	5.5–7.0	Batch	37–60	[12,11]
E. cloacae and bacterial consortium	PbNPs	5.0	Glucose	1.48–2.48	7.0	Batch	37	[45]
Chlamydomonas reinhardtii, *C. butyricum*, *Acidogenic consortium*	SiO2NPs	5.1–120	Glucose and effluents	4.3–666	4.2–7.6	Batch and continuous	28–30	[38,12]
Rhodopseudomonas palustris, *C. pasteurianum*, *R. sphaeroides*	TiONPs	50–100	Glucose, Malate, and waste sludge	5.0–69.9	7.0–8.0	Batch	30–35	[83,21]

When compared to the control, the lower amount of (2 mg L^{-1}) this CuNPs resulted drop in biohydrogen yield as 1.74 from in a 3.5 % of biohydrogen [**46**]. At 12 mg L^{-1} concentrations, there was a considerable decrease in Hydrogen yield of 56.9 to 72.2 %, respectively. Lower hydrogen yields were linked to CuNPs' inhibition activity on $E. cloacae$ as well as $C. acetobutylicum$ and that were close to the extent inhibitory doses of 13 & 15 mg L^{-1} correspondingly. Furthermore, reduced acetate as well as butyrate proportion with elevated CuNPs concentrations rationalized the reduction in yield [**35**]. In general, higher biohydrogen yield has been correlated with higher acetate-butyrate ratio, while lower yield was correlated with elevated propionic acid as well as alcohol production [**48**].Ultimately, CuNPs had a greater inhibition effect on biohydrogen fabrication yield by both $C. acetobutylicum$ as well as $E. cloacae$ than Cu^{2+} ions, and that may be related to CuNPs' increased antimicrobial activity [**46**]. $C. butyricum$, on the other hand, noticed the CuNPs encapsulate SiO_2 composite significant benefit in increasing H_2 production from 0.92 1.01 mol moP1 hexose [**60**]. This finding implies that lowering the dosage of CuNPs (2.5 mg L^{-1}) or encapsulating them in a absorbent medium could be utilized to increase biohydrogen production in addition to regulating their antibacterial activities [**46**].

4.3.2. AgNPs and AuNPs

Because of their distinctive attributes, silver nanoparticles have a broad array of applications, such as protein immobilization, electronic parts, nourishment, as well as healthcare industries [**55**]. Despite their recognized antibacterial properties, Zho et al. reported the efficient use of Ag nanoparticles for biohydrogen fabrication from glucose via a mixed culture influenced by $C. butyricum$ [**81**]. In this case, higher concentration of Ag nanoparticles resulted in increased biohydrogen fabrication, which remained constant until the concentration reached 200 nM. Zhang and Shen demonstrated the importance of Ag nanoparticles in enhancing fermentation process for biohydrogen fabrication [**80**]. At 5 nM of Au nanoparticles, anaerobic bacterial culture produced 62 % more yield (2.3 mol biohydrogen mol^{-1} of hexose) compared to control (1.4 mol biohydrogen) (**Table 2**). Surprisingly, the increase in biohydrogen production was proportional to the concentration of AuNPs. AuNPs further changed the dosage of metabolites during biohydrogen fabrication process [**81**]. The increased acetate-to-butyrate proportion as well as minimal ethanol fabrication in the existence of AuNPs are related to an increase in biohydrogen fabrication. This investigation suggests that using an anaerobic culture influenced by $C. butyricum$ as such an inoculum as well as AuNPs as such an inoculum offered a suitable methodology for effective biohydrogen fabrication using sucrose [**60**]. Accordingly, in the existence of AuNPs, the suitability of biohydrogen fabrication with solo chamber reactor employing electrochemically-active-biofilm (EAB) as well as acetate as a material through activated sludge was evidenced [**49**]. EAB established on metal alloy mesh as well as carbon paper along with activated sludge obtained 44 % H2 of maximum production from acetate as nutrient at 1 mM dosage of AuNPs. Remarkably, in the apparent lack of AuNPs, EAB did not yield such a biohydrogen [**27,49**].

4.3.3. PdNPs

The PdNPs also possess considerable level of influence on organisms involved in the biohydrogen production (**Table 2**). Accordingly, Mohanraj et al. investigated the impacts of green synthesized palladium NPs (PdNPs) derived from $C. sattvum$ leaves extract on biohydrogen fabrication by $E. cloacae$ in a mixed population fed with glucose [**45**]. At 5.0 mg L^{-1} PdNPs, there had been a 0.6 to 6.4 % increase in yield, with high productivity around 1.5 to 2.48 mol biohydrogen mol^{-1} of glucose, respectively, when compared to their corresponding controls. In this case, the occurrence of PdNPs lowered lag phase for biohydrogen fabrication as well. Pd^{2+} substance, on the other hand, had a negative impact on yields and the lag phase of biohydrogen fabrication under similar circumstances [**23**]. Ultimately, mixed culture produced more biohydrogen than $E.cloacae$. Surprisingly, PdNPs addition up to 20.0 mg L^{-1} had no effect on the biological properties of $E.cloacae$ as well as mixed culture. This investigation results suggests that the Pd^{+2} ions inhibited biohydrogen fabrication more than PdNPs, resulting in a substantial decrease in glucose reducing efficiency under identical circumstances. Furthermore, increased propionate synthesis as transitional metabolites compounds revealed its detrimental impact on biohydrogen fabrication [**45**]. The increased hydrogenase potential in the existence of PdNPs could be actively related to the increased H2 fabrication over Pd^{2+} compound [**5**].

4.3.4. FeNPs

Numerous bacterial isolates have been tested for biohydrogen production in the presence of FeNPs, using carbohydrates and organic wastes as nutrients [**8**]. Taherdanak et al. investigated the effects of FeNPs vs Fe^{2+} substance at concentrations ranging from 0 to 50 mg/l on anaerobic activated sludge-based microbial fermentation on biohydrogen production from glucose [**73**]. Alike Fe^{2+} substance and FeNPs increased biohydrogen yield by up to 37 % at dosage of 25 mg L^{-1}, respectively, than control. Elevated biohydrogen fabrication was associated with a substantial change in the transitional metabolites more towards an elevated acetate to butyrate proportion as well as a decrease in the dosages of ethanol as well as propionate [**17**]. Surprisingly, FeNPs lowered propionate fabrication by 75 % compared to 35 % in the existence of Fe^{2+} substance [**73**]. Nevertheless, Nath et al. demonstrated the effect of plant mediated FeNPs employing $S. cumini\ extracts$ (leaf and bark) and Fe^{2+} at elevated dosage of 200 mg L^{-1} on biohydrogen fabrication using $E.\ cloacae$ [**51**]. In the existence of FeNPs rather than Fe^{2+}, a comparable significant impact on biohydrogen yield was observed. FeNPs (100 mg L^{-1}) outperformed the control (0.95 mol biohydrogen mol^{-1} of glucose) by 100 % (1.9 mol biohydrogen mol^{-1} of hexose). In the existence of 25 mg L^{-1} Fe^{2+}, however, an extent biohydrogen production around 1.5 mol mol^{-1} of glucose had obtained. Surprisingly, FeNPs increased the $E.cloacae$ growth. Hence, such findings, FeNPs appear to improve the metabolic activities of $E.\ cloacae$ for biohydrogen production [**51**]. In contrast, FeNPs at elevated dosage of 400 mg L^{-1} increased biohydrogen production (1.23 mol) through such a blend of microbial mixture using the glucose as nutrient by 38 % [**79**]. Surprisingly, at elevated dosage (500 mg L^{-1}), neither deleterious impacts of nanoparticles on biohydrogen fabrication (**Table 2**) had been -reported [**40**]. Dolly et al. proved photo-fermentative biohydrogen production using malate with the co-culture of $E.coli$ as well as $R.sphaeroides$ in the existence of bulk- as well as nanoparticles forms of Fe at various doses [8]. At such an optimum dosage of (312 mg L^{-1}), Fe nanoparticles was discovered to be 19 % more potential than the bulk form in biohydrogen production.

4.3.5. Fe_2O_3NPs

The $E.aerogenes$, $E.cloacae$, and $C.acetobutylicum$ fed with glucose considerably increased biohydrogen yield by around 17-33 %at dosages of 175 to 200 mg L^{-1} of Fe_2O_3 nanoparticles [**60**]. Surprisingly, Mohanraj et al. proposed that perhaps the sugar content used as nutrient and showed significant influence on biohydrogen production by $E.cloacae$ in the availability of 200 mg L^{-1} of Fe_2O_3 nanoparticles [**46**]. Glucose was discovered being a superior source of nutrition, yielding 21 % more biohydrogen than sucrose (4 %) (**Table 2**). Mixed culture, on either side, conducted well, increasing sucrose production by 33 % at the similar dosage of Fe_2O_3 nanoparticles [**20**]. Accordingly, Gadhe et al. proposed that industrial effluents from various sources necessitated various concentrations of Fe_2O_3 nanoparticles for optimal biohydrogen production through anaerobic sludge [**12**]. Nasr et al.

demonstrated that immobilization of anaerobic (activated) sludge with Fe_2O_3 NPs improves sugar yield up to 57.8 %. This was proposed that combining the dark- and photo-fermentation processes could result in increased biohydrogen yields [**50**]. $E. aerogenes$, on the other hand, demonstrated a 63.1 % raise in biohydrogen yield used cassava starch at a dosage of 200 mg L^{-1} of Fe_2O_3 nanoparticles, and this was well almost four fold greater than that documented on $E. aerogenes$ and that used pure glucose as nutrient [**36**]. A preliminary morphological examination of $E. aerogenes$ revealed a highest cell accumulation at increased (200 mg L^{-1}) dosage ofFe_2O_3 nanoparticles. Such a phenomenon was most probably connected to the bacterial nanowire emergence in reactions to nanoparticles, since it contributed in enhancing transfer of electrons between many cells during fermentation [**60**]. Cellular internalization of Fe_2O_3 nanoparticles as black patches in the $E. aerogenes$ cytosol affirmed [**36**]. Under mesophilic temperature, Zao et al. revealed a 26.4 % raised the yield as 1.53 mol of biohydrogen/mol glucose through anaerobic sludge at increased concentrations (400 mg L^{-1}) of Fe_3O_4 nanoparticles [**82**]. Malt effluent, on the other hand, has been identified as such an excellent nutrient for increasing biohydrogen fabrication in the presence of mixed culture up to 83.3 % with reduced dosage (50 mg L^{-1}) of Fe_3O_4 nanoparticles [**41**]. Another study reported that the addition of 200 mg L^{-1} ofFe_3O_4 nanoparticles with bagasse (sugarcane) inoculated with anaerobic sludge considerably increased the biohydrogen yield up to 69.6 % [**65**]. They suggested that the bacterial community structure as well as hydrogenase transcriptomic model might be interacted with Fe_3O_4 nanoparticles resulted increased quantity of biohydrogen yielding communities as well as hydrogenase gene function when compared to the control and the presence of Fe^{2+} substances. Furthermore, increasing the concentration of Fe_3O_4 nanoparticles to 400 mg L^{-1} used to have a substantial impact on biohydrogen production [**65**]. In such case, elevated nanoparticle concentration may lead to toxicity and also the establishment of free radicals, which inhibits microorganism growth and reduce the biohydrogen production [**3**].

4.3.6. NiNPs and NiONPs

Nickle ions have long been known to improve biohydrogen yield by enhancing the catalytic activities of hydrogenases [**69**]. Taherdanak et al. investigated the effect of Ni^{2+} ions as well as Ni nanoparticles upon that fabrication of biohydrogen from anaerobic sludge nourished with glucose [**73**]. Ni nanoparticles at a concentration of 2.5 mg L^{-1} yielded 0.9 % of increased biohydrogen production than the test control, which was substantially reduced as 99.0 l kg^{-1} against at higher (50 mg L^{-1}) dosage of Ni nanoparticles [**73**], The Ni^{2+} ions increased the biohydrogen yield by around 55 % at a dosage of 25 mg L^{-1}. Finally, the application of Ni nanoparticles seemed to have a negative effect on biohydrogen yield via anaerobic sludge [**73**]. Mullai et al. discovered a 22.7 % increase in biohydrogen fabrication of 2.54 mol moP1 of sugars via anaerobic sludge in the presence 5.7 mg L^{-1} of Ni nanoparticles [**47**]. Similarly, using NiO as a nanoparticle, anaerobic sludge demonstrated a differential increase in biohydrogen fabrication with effluents of molasses and dairy [**12**]. Molasses outperformed dairy wastewater by 23.5 % at NiONPs dosage of 5 mg L^{-1}, respectively (16 %) (**Table 2**). Glucose, on the other hand, resulted in a reduction progress in production of biohydrogen as 4.8 % (1.30 mol biohydrogen mol^{-1} of hexose) at considerably greater (200 mg L^{-1}) concentrations of NiONPs [**11**]. In this case, variations in biohydrogen yields could be related to variations in feed composition.

4.3.7. TiO_2 NPs

According to Jafari and Zilouei, the TiO_2 treated biomass produced approximately 101 l kg^{-1} of biohydrogen than the control (44.2 l kg^{-1}) [**22**]. Correspondingly, packaging of $C. reinhardtii$

organisms with TiO_2 casings resulted in approximately twofold increased biohydrogen production as 100 mL L^{-1}) than free living organisms [**72**]. Similarly, Zhao and Chen investigated about influence of TiO_2 nanoparticles on the growth as well as activities of biohydrogen producing enzymes in the photo-fermentative system of $R. palustris$ on dark-fermentative treated wastewater [**83**]. The biohydrogen production and nitrogenase activities were significantly increased in the existence of TiO_2 (100 mg L^{-1}) compared to the control (**Table 2**). However, when TiO_2 was present, the uptake hydrogenase activity decreased significantly. Hence, the minimal utilization of yielded biohydrogen via uptake hydrogenase, a constant increase in yield was achieved, as well as a raise in $R. palustris$ biomass. In the existence of TiO_2NPs, the biohydrogen fabrication increased by 46 %, with a production of 1 mol kg^{-1}. Ultimately, the 2 different biohydrogen yield -with a production attributes of around 1.90 mol kg^{-1} indicated that such an integrated model of dark- followed by photo-fermentative is sufficient to reach increased biohydrogen recovery efficiency from effluents [**83**]. Correspondingly, as per photo-fermentative environments, $R. sphaeroides$ enhanced average biohydrogen yield rate as well as duration by 1.7 and 1.9-fold, correspondingly, than control TiO_2 NPs [**58**].

4.3.8. SiO_2 NPs

Silica has long been known to be a much more biomaterial support for both polypeptides and microbes [**1**]. Venkta Mohan et al. revealed about efficient exploitation of mesoporous SiO_2 nanoparticle in biohydrogen fabrication using blended consortia from widespread chemical effluent [**44**]. Overall, increased feed packing had a negative impact on biohydrogen production by blended consortia owing to reduced degradation or ineffective feed utilization. Surprisingly, SBA-15 nanoparticles (120 mg L^{-1}) considerably increased the biohydrogen fabrication by up to 666 % (**Table 2**) compared to the control at increased feed packing (2.55 kg COD day-:), possibly due to selfimmobilization of substances just on nanomaterials during in the fermentations [**44**]. Such an increased biohydrogen yield was connected with a 37 % efficient feed breakdown when compared to the control (23 %). Furthermore, maximum and minimum levels of acetate as well as propionate as bioavailable compounds revealed the increase in biohydrogen production in the existence of SBA-15 than control. Similarly, Beckers et al. demonstrated that amorphous SiO_2 nanoparticles (5.1 mg L^{-1}) had no effect on the biohydrogen yield and biochemical transitional profiles of $C. butyricum$ when fed glucose [**2**]. Only a 4.3 % increase in biohydrogen production (0.96 mol mol^{-1} of glucose) had been noted. The $C. reinhardtii$ had been yielded 45 % of biohydrogen in the existence of SiO_2 nanoparticles (60 mg L^{-1}) when compared to a control under photo-fermentative circumstances [**16**]. Surprisingly, the existence of SiO_2 nanoparticles stimulated the development of C. reinhardtii, with 23 % raise in photosynthetic pigment content during biohydrogen fabrication correlated to better distribution of light [**16**].

5. Techno-economic analysis of biohydrogen production

Economic analysis predicts the commercial interest of any products in the large scale [**94**]. This is usually performed to analyze the feasibility of any projects based on costing analysis, commercial load and climate data [**95**]. Based on the results procured from these analyses the decision making for any projects is simple [**96**]. Based on literature the techno-economic evaluation of each process calculated based on the assumptions such as, depreciation, income tax rate cost, ideal breakeven, inflation, baseline discount rate, selling price, and consumable cost were considered [**88,89**]. There are many tools to predict economic analysis, recently Aspen received huge attention for the calculation of the capital cost, operating cost, cumulative cash flow and profit cost [**97**]. Added to above, the sensitivity analysis was also performed based on the raw material cost, discount rates and minimum selling price of biohydrogen [**90**]. Minimum selling price of the any process can be calculated from the biohydrogen production cost. For instance, if the biohydrogen was generated from thermochemical process, the process cost is considered to be the minimum cost after adding few margins. The accumulated cost must be competitive with the cheapest proce in the market based on the current price. While calculating the raw materials there must be some tolerance in the calculation since the price of the raw materials always fluctuate based on the global market. In general, during calculation they will be keep the tolerance to 20 %. Based on some notable studies, gasification and anerobic digestion are the two techniques which can be used to produce the biohydrogen in cheap cost [**91**]. Furthermore, the selling price of the hydrogen were sensitive to the CO_2 production and nanoparticles usage [**98**]. Further, there also some effects on the costing owing to inflation. Inflation affects the feed cost prices, equipment cost and labor cost.

6. Conclusion and future perspectives

There is wide consensus across many publications that the use of nanoparticles is capable to significantly accelerate (bio)hydrogen production kinetics as well as yield, which represents a significant potential to reduce production costs. Although, there are still many challenges, nanoscience maintains a promising direction that make (bio)hydrogen a reliable alternative to fossil fuels. The prerequisite for this is the complex refining of biowaste into a wide range of high value-added products and the recovery of waste heat.

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