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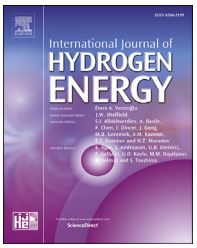

Effect of cell immobilization, hematite nanoparticles and formation of hydrogen-producing granules on biohydrogen production from sucrose wastewater

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ABSTRACT

This study investigated the effect of granules formation, hematite nanoparticles and biofilm carriers on biohydrogen production from sucrose wastewater in continuous stirring tank reactors operated at 12 h HRT, pH of 5.5 and 35 °C. Granular-based bioreactor was subjected to acid incubation period for 24 h by shifting the pH from 5.5 to 3. Before application of the acid incubation, hydrogen-producing granules (HPGs) diameter and hydrogen production rate (HPR) of 0.5 mm and 4.3 L/L.d, respectively were measured at 10 g-sucrose/L. Application of acid incubation enhanced the granulation process, where the particle size increased to 2.8 mm and higher HPR of 7.8 L/L.d was obtained. Higher sucrose concentration (15–30 g⁄L) enhanced HPGs diameter and increased the HPR. At 10 g-sucrose/L, addition of hematite nanoparticles increased the HPR to 5.9 L/L.d higher than 3.87 L/L.d measured in control reactor. Biofilm-based reactor showed HPR of 2.48 L/L.d lower than the control reactor.

Introduction

Nowadays, the increase in global population as well as the depletion of fossil fuel supply has become crucial due to high energy demands, this gap has to be filled by finding renewable energy sources. Biofuel production (biohydrogen, biodiesel, bioethanol, biomethanol) can be considered one of the alternative energy sources for the future to balance the global energy shortage [1–4]. Among various biofuel candidates, hydrogen has attracted more research attention as a dream fuel for the future because it is clean energy source (water is the only combustion byproduct) with high energy content [1], as well as hydrogen can used for production of electrical

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energy using fuel cell e.g. polymer electrolyte fuel cell (PEFC) [5]. Conventional hydrogen production methods such as steam reforming of methane, non-catalytic partial oxidation of fossil fuels and auto-thermal reforming are highly energy intensive processes. Biological methods show distinct advantages for hydrogen production such as operation under ambient environmental conditions and specific substrate conversions [6]. Hydrogen production via dark fermentation has gained a considerable attention over other biological processes because of its advantages such as low energy requirements as well as production of clean energy source from negative-value organic wastes [7,8]. Hydrogen production from wastewater can achieve the dual environmental benefits of waste reduction along with production of high value energy fuel. Moreover, wastewaters can be considered as ideal substrates for biohydrogen production since they are rich in carbon and nitrogen [9,10]. However, biohydrogen production using dark fermentation process has some limitations. The main drawbacks of dark fermentation process are low hydrogen yield and slow rate [11] since only one-third of the substrate can be converted to hydrogen gas, while the remaining two-thirds of the substrate are consumed to form other fermentation products such as volatile fatty acids (VFAs), alcohols, acetone, etc [12]. In order to obtain high efficient hydrogen production process, it is necessary to maintain high biomass concentration in the bioreactor while operating at low hydraulic retention time (HRT) [13–15]. However, one of the main disadvantages of continuous stirring tank reactor (CSTR) is the low biomass concentration due to the washout of the bacterial species [15]. Production of granular sludge represents an efficient solution to avoid the problems displayed by CSTR bioreactor. Formation of HPGs can keep high biomass concentration even the bioreactor has been operated at low HRT [16]. Several operating conditions must be controlled in order to obtain stable granulation process. These parameters include pH, temperature, substrate characteristics, organic loading rate (OLR), microbial composition, nutrient feeding, metal ions, etc. [17]. Among these parameters, OLR is a key factor for the formation and stabilization of the granules [18]. Increasing OLR higher than the optimal value leads to increasing VFAs concentration and decreasing pH value [19], while low OLR results in accelerating the mass transfer reaction which can destabilize or even destruct of the granules due to lack of substrate [20]. Ghangrekar et al. [21] reported that OLR in the range of 2.0–4.5 kg-COD/m^3.d can be considered as an optimum value for good granules formation. Based on previous studies, it was observed that the optimum range of OLR for granules formation can be varied depending on the operating conditions [17].

Recently, several researchers observed that the addition of nanoparticles (NPs) improved the biohydrogen production from waste substrates [22–27]. The authors reported that NPs could promote the bioactivity of hydrogen-producing species, increase the bacterial growth and affect the distribution of the soluble metabolic products as well as the NPs can act as electron sinks due to their tendency for electrons, this behavior can enhance the reduction of protons to hydrogen [22,27]. Mu et al. [28] reported that the interactions between the NPs and hydrogen-producing species can enhance the biohydrogen production. To date, few studies have investigated the effect of NPs on biohydrogen production and most studies were conducted as batch tests. For this purpose, more work should be done to study the effect of NPs on continuous biohydrogen production. Therefore, the main objectives of this study are: first to enhance the biohydrogen production from sucrose wastewater by formation of HPGs, where different sucrose concentrations of 10–30 g/L with increments of 5 g/L were examined, second investigating the effect of immobilized hematite NPs and biofilm carriers versus suspended-based reactor on biohydrogen production, third analyzing the performance of the different bioreactors for chemical oxygen demand (COD) reduction and measurement of sucrose conversion percentages.

Materials and methods

Inoculum

The sludge was collected from wastewater treatment plant (KASSLERFELD) in Duisburg and sieved using a mesh (2 mm) to remove waste materials. Before inoculation, the sludge was heated at 100 °C for 2 h to inhibit the hydrogen-consumers e.g. methanogens and other non H_2-producing microorganisms and to enrich the hydrogen-producing microorganisms such as clostridium species.

Operation of bioreactors

Effect of hydrogen-producing granules

Hydrogen was produced in 8-L CSTR reactor with 6-L active volume using artificial sucrose wastewater prepared daily by diluting sucrose in water. The reactor was operated at 35 °C, 5.5 pH and 12 h HRT for 120 days. Different sucrose concentrations of 10, 15, 20, 25 and 30 g/L were studied. The bioreactor was started up with 10 g-sucrose/L and mixing rate of 300 rpm. These operating conditions were kept until steady-state conditions (stable hydrogen production) were attained. After reaching steady-state conditions, CSTR bioreactor was subjected to acid incubation by decreasing pH value from 5.5 to 3 for 24 h. Feeding (substrate and nutrients) was stopped during the incubation time. After that, the CSTR was operated at the normal conditions as pH of 5.5 and the stirring rate was decreased to 150 rpm. The sucrose concentration was increased gradually to 15, 20, 25 and 30 g/L.

Effect of hematite NPs and biofilm carriers

Three parallel CSTR bioreactors of working volume of 4.5-L were operated at 10 g-sucrose/L, 12 h HRT, 5.5 pH, 35 °C and continuously stirred at 150 rpm. The bioreactors were operated at three different reaction modes of suspended-based reactor (control), biofilm-based reactor and immobilized hematite NPs-based reactor.

A nutritional solution was used throughout this study contained (per L): 26 g NH_4Cl, 2.5 g K_2HPO_4, 2.5 g KH_2PO_4, 3.2 g $MgCl_2 \cdot 6H_2O$, 983 mg $CaCl_2 \cdot 6H_2O$, 860 mg $FeSO_4 \cdot 6H_2O$, 150 mg $CoCl_2 \cdot 6H_2O$, 150 mg $MnCl_2 \cdot 4H_2O$, 140 mg $(NH_4)_2MoO_4 \cdot 4H_2O$, 120 mg $Na_2B_4O_7 \cdot 10H_2O$, 267.2 mg $NiCl_2$, 230 mg $ZnCl_2$ and 100 mg $CuCl_2 \cdot 2H_2O$. The pH was monitored and controlled (iks Aquastar version 2.XX, iks Computer System GmbH,

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