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Rapid formation of hydrogen-producing granules in an up-flow anaerobic sludge blanket reactor coupled with high-rate recirculation

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abstract

Application of an up-flow anaerobic sludge blanket (UASB) reactor to dark fermentative H_2 production greatly improves H_2 productivity due to the maintenance of high biomass concentration. However, a long start-up HRT and start-up period are required to develop the H_2 -producing granules (HPGs) and to avoid washing out the suspended sludge at the start of the process. In the present work, a novel strategy to rapidly form HPGs was developed in UASB reactor. To induce highly active mass transfer in the UASB reactor, a high recirculation rate (15 times the influent) was adopted over 10 days, then recirculation was stopped. As the operation progressed, self-flocculation took place and HPGs developed after 90 h of operation. A stable production of H_2 was observed after 20 days of operation. The thickness of the HPGs layer in the sole UASB reactor increased progressively, and consequently the average HPG diameter and concentration were 1.86 mm (0.1–3.9 mm) and 52 g/L, respectively, after 60 days of operation. These findings seem to suggest that high-rate recirculation plays a crucial role in accelerating the formation of HPGs in such UASB reactors through high up-flow velocity, providing active mass transfer.

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1. Introduction

Since the development of industrial and postindustrial economies, energy requirements and environmental constraints are constantly driving the exploration of new and innovative methods for bioenergy generation. Hydrogen $(H₂)$ has gained importance in recent years because it has highest energy content per unit weight of any known fuel, which is 2.75 times greater than the best hydrocarbon fuels. In addition, the most important fact is that H_2 offers tremendous potential as a sustainable energy source as its combustion produces only water and energy [\[1\]](#page--1-0). Currently, most H_2 is made by energyintensive physicochemical processes; however, a lot of attention is being paid to promising biological approaches using biomass or wastewater. Biological approaches could be categorized into two basic concepts: photo- and dark fermentation. Notably, dark fermentation, often referred to as dark fermentative H_2 production (DFHP), has been receiving

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increased attention because it can play a dual role in solutions to both environment and energy issues.

Up to date, even though conventional suspended bioreactors have been widely employed, they have not been sufficiently competitive with immobilized bioreactors. This is primarily due to the limitation of having to maintain a high concentration of biomass in the reactor, which easily leads to operation failure [\[2\].](#page--1-0) To solve this problem, an up-flow anaerobic sludge blanket (UASB) reactor with ability to form H_2 -producing granules (HPGs), has been used to improve H_2 productivity. However, long start-up periods, generally a few months, are required for the formation of HPGs $[3-5]$ $[3-5]$ $[3-5]$.

A simple, novel approach to reduce the start-up period in an UASB reactor was recently suggested by our research group $[6-8]$ $[6-8]$. Processing with completely-stirred tank reactor (CSTR) prior to use of the UASB reactor demonstrated that initiation of self-flocculation of H_2 -producing bacteria within a few days, much faster than for directly operated UASB reactor. Provision of high shear forces by the cylindrical shapes and mechanical turbines used for mixing in the CSTR, resulted in improvement of the conditions favoring formation of HPGs. This is a good, fast approach to successful formation of HPGs, but the utilization of two reactors is nevertheless inconvenient procedures. Chang and Lin[\[3\]](#page--1-0) reported that cell flocculation can be enhanced under low HRT condition during operation of UASB reactor, providing high up-flow velocity. Therefore, in this study, to enhance the mass transfer in UASB reactor, high-rate recirculation was

adopted by coupling with a settling tank. Compared to previous research, this system was more effective in terms of developing HPGs and H_2 productivity. The increasing thickness of the HPGs layers and HPGs size distributions were monitored in the UASB reactor as well as microbial community. To the best knowledge, this was the first attempt to increase HPGs formation using high-rate recirculation.

2. Materials and methods

2.1. Seed sludge and substrate

The seed sludge was taken from an anaerobic digester at a local wastewater treatment plant. pH, alkalinity, and volatile suspended solids (VSS) concentration of the sludge were 7.5, 4.70 g $CaCO₃/L$, and 2.8 g/L, respectively. It was pretreated at 90 $^{\circ}$ C for 20 min to inactivate H_2 -consumers such as methanogens. The tested substrate was glucose at a concentration of 10 g chemical oxygen demand (COD)/L. Subsequently, in order to provide buffer capacity, NaHCO₃ was externally added to the substrate at concentration of 5 g/L. Concentrations of NH₄Cl, KH₂PO₄, and FeCl₂∙4H₂O were added for COD:N:P:Fe ratio of 100:5:1:0.33. Feed also contained the following nutrients (in mg/L): MgCl₂⋅6H₂O 100; CaCl₂⋅2H₂O 75; Na₂MoO₄⋅4H₂O 0.01; H₃BO₃ 0.05; MnCl₂∙4H₂O 0.5; ZnCl₂ 0.05; CuCl₂ 0.03; NiCl₂∙6H₂O 0.05; CoCl₂∙2H₂O 0.5; Na₂SeO₃ 0.05 [\[9\]](#page--1-0).

Fig. $1 -$ Schematic diagram of the UASB reactor coupled with high-rate recirculation.

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