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## Chemical Engineering Journal



journal homepage: www.elsevier.com/locate/cej

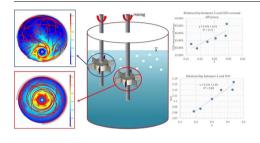
### Enhancing anaerobic fermentation performance through eccentrically stirred mixing: Experimental and modeling methodology



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#### G R A P H I C A L A B S T R A C T



#### A R T I C L E I N F O

Keywords: Anaerobic hydrogen production (AHP) Eccentrically stirred mixing Mixing eccentricity Mixing height Lyapunov exponent Navier-Stokes equations

#### ABSTRACT

Anaerobic hydrogen production (AHP) is a biochemical process capable of producing hydrogen and fatty acids from organic substrates in acidogenic phase. Currently, almost all of anaerobic systems use centrally stirred mixing where the flow is essentially rotated uniformly in the radial direction. Given the high energy requirement of mixing and the urgent needs for enhancing the contact between microbial particles and substrates, a new type of mixing strategy should be developed. This study focused on the enhancement of biogas production and organic removal efficiency in AHP systems through a novel chaotic mixing regime. The stirring impeller was operated eccentrically with the stirring center eccentrically located 2 and 4 cm off the center, respectively and at different heights (5, 10.5 and 16 cm). The eccentrically stirred mixing was expected to enhance the mixing efficiency and accelerate the contact between biomass and organic substrates in the AHP system. A numerical finite element model was developed to simulate the flow patterns and the biomass distribution, through which the Lyapunov exponent  $\lambda$  values were calculated in order to assess the strength of eccentrically stirred mixing at different mixing eccentricities. The model simulations showed that at the height of 10.5 and 16 cm, the Lyapunov exponent  $\lambda$  increased as the mixing eccentricity increased. This corresponded well with the experimental results, which showed that the organic removal efficiency and biogas production increased with the mixing eccentricity. Compared with centrally stirred mixing, eccentrically stirred mixing enhanced the interaction of flow and biomass particles and ultimately enhanced the biogas production and organic removal in AHP systems.

#### 1. Introduction

Anaerobic hydrogen production (AHP) is a biochemical process

capable of producing hydrogen and organic acids (e.g., acetic acid, butyric acid) from organic substrates (e.g., glucose and wastewater) under acidogenic phase. Compared with methane production from

https://doi.org/10.1016/j.cej.2017.11.088

Received 23 September 2017; Received in revised form 14 November 2017; Accepted 17 November 2017 Available online 21 November 2017 1385-8947/ Published by Elsevier B.V.

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anaerobic digestion (AD), AHP offers unique advantages including high reaction rate, high operational stability, and clean energy production with hydrogen gas as the major biogas component [1]. Previous researchers have found that several engineering parameters (e.g. pH [2,3], temperature [4,5] and substrate content [5,6]) directly affected hydrogen production (e.g. high hydrogen yield and suppression of the methanogens at pH of 4.8–5.5 and temperature of 60 °C [2]). In fact, the mixing of anaerobic systems is critical for reaction rate and energy consumption [7]. Normally, sufficient mixing ensures organic substrates efficiently distributed in anaerobic systems to react with biomass particles, leading to fast heat transfer, release of trapped bubbles, and avoid biomass precipitation [8]. Slow mixing (e.g. under 160 rpm) has been used to facilitate sludge granulation, although complete-mixing AD systems can cause a washout of granular sludge [9,10]. Traditional aerobic/anaerobic systems have historically used centrally stirred mixing (henceforth called centric mixing) where the flow is essentially rotated uniformly. Although this mixing pattern is well adopted and easily designed and operated, it might not optimize the interaction between substrates and biomass particles, especially in AHP systems where biogas production can complicate the flow field. As one of the major energy consumption factors in anaerobic systems, it is critical to better understand the mixing strategy and develop new energy-saving and efficient mixing patterns.

Mixing in fluids is accomplished slowly by molecular diffusion in laminar flows [11], which is greatly accelerated by turbulence and is associated with large velocity gradients, especially in the small scales and high shear rates. For these reasons, turbulence is not suitable for mixing in biological situations where the high shear rates could damage bacterial cells and tear the granular sludge. In other situations such as highly viscous flows, the flow remains laminar. A novel and fundamental mixing process rooted in dynamical systems theory has been proposed [12] and termed chaotic advection. It was theorized that chaotic mixing would significantly increase the exposed surface area. Flow changes direction continuously under the chaotic mixing scheme, which is expected to greatly enhance organic removal by maximizing contact between biomass particles and substrates. Even though turbulent mixing is common in high Reynolds number flows where the large amount of energy drives particles (e.g., bacterial cells, flocs) in random directions and produces a homogeneous fluid, chaotic mixing can also occur at low Reynolds numbers when time-periodic oscillations with fixed amplitudes are imposed [13]. Chaotic advection causes simple non-turbulent flows to exhibit complicated particle trajectories that result in enhanced mixing, and has been applied to low Reynolds number plasma mixing without damaging bacterial cells, to enhancing heat transfer, and to creating long-chain polymers. Our group has applied this concept to a variety of wastewater and groundwater applications and scales [14-17]. Furthermore, parallel-competitive reaction systems have been simulated in a chaotic flow by solving the differential convection-diffusion-reaction and it was shown convincingly that besides the relative rates of reaction, the nature of the chaotic flow is a determining factor in the evolution of the aqueous concentrations [18]. The groundwork was presented in a previous study [19] by solving numerically the aforementioned differential equations. An excellent state-of-the-art review of the numerous applications and theoretical background of chaotic advection was presented this year [20].

Kinetic mixing models have been studied to elucidate the effect of mixing speed/intensity on mass dispersion, biogas production, substrate removal efficiency and mass viscosity. Idealized models demonstrating chaotic advection include the point vortex model, tendril-whorl flow, the pulsed source-sink system, and the three-vortex system. Until now, mixing in AD systems has been limited to centrally located impellers for the convenience of system configuration and operation [21]. There have been only a few studies of chaotic mixing (off center) on mass distribution and reaction [22] and no chaotic mixing has been applied in the field of AHP. In fact, mass distributions in AHP systems would vary with the mixing position. A flow visualization technique was developed by observing the fluorescent green dye in a vessel to elucidate the mixing characteristics [23]. The biofermentor can be divided into two sections – active and inactive volumes, with active volume being formed as a cavern around the mixer. It is expected that the size of the active volume would change with the mixing position in AHP biofermentors, leading to variable mass distribution, gas bubble dispersion, biogas production, substrate precipitation and liquid fermentation production. Previous studies had found that in the journalbearing system (a system similar to the AHP experimental design in this study) consisting of two eccentric cylinders, the annular region was filled with a viscous liquid and the cylinders were rotated alternately to produce chaotic advection [24]. Furthermore, the role of diffusion and transient velocities was studied in the dispersal of passive scalars produced in a low Reynolds number journal-bearing flow [25].

The breakthrough of this study was to elucidate the impact of chaotic mixing on the enhancement of biogas production and organic removal in an AHP biofermentor, with the objective to explore the relationships between the chaotic mixing theme and energy output of AHP process. There were four tasks in this study. First, a lab-scale AHP biofermentor was set up to use glucose, a typical carbon source for hydrogen production bacteria, and operated in different chaotic mixing conditions. Second, the important parameters in AHP such as gas production, liquid components and chemical oxygen demand (COD) removal efficiency were investigated under different mixing impeller positions. Third, a numerical finite element model was developed to explore the flow and particle motion in the AHP system and determine the relationship between mixing and energy output. Finally, the significance of applying chaotic mixing for anaerobic biofermentors was explored in the context of scientific and industrial perspectives.

#### 2. Materials and methods

# 2.1. Anaerobic hydrogen producing (AHP) biofermentor setup and operation

A BIOFLO 110 Fermentor (New Brunswick Scientific, NJ) (effective volume of 2 L, height: 25 cm, radius: 6.5 cm) was used as the AHP system. The vessel was sealed for air impermeability. An agitator with an impeller was installed for mixing. The radius of the impeller was 2 cm and the width of the impeller was 1.75 cm with 6 blades uniformly distributed around the circumference (Fig. 1). Before the tests, 20.625 g glucose was fully dissolved in 200 ml distilled water and added to the vessel as the carbon source, followed by filling with 2L nutrient solution containing 4.0 g NH<sub>4</sub>HCO<sub>3</sub>, 2.6 g NaH<sub>2</sub>PO<sub>4</sub>, 200 mg MgSO<sub>4</sub>·7H<sub>2</sub>O, 20 mg KCl, 20 mg Na<sub>2</sub>MoO<sub>4</sub>·2H<sub>2</sub>O, 20 mg CaCl<sub>2</sub>·2H<sub>2</sub>O, 15 mg MnSO<sub>4</sub>·7H<sub>2</sub>O and 5.6 mg FeCl<sub>2</sub>. The chemical oxygen demand (COD) of the mixed solution was 10 g/L. The vessel was then sparged with nitrogen gas (99.995% high purity, Airgas Co., CT) for 20 min to achieve the anaerobic environment. Organic soil (22g) collected from a greenhouse at the University of Connecticut was used as the inoculum for anaerobic bacteria to obtain the biomass particle concentration of 10 g/L in the AHP system.

Previous studies had found that the disturbance of shock loading was better absorbed by the digester at low speed mixing conditions (80 rpm) than at high speed mixing conditions (200 rpm) [26], since the bacteria in the anaerobic digestion process are more likely to live in a balanced micro-ecosystem [27], and the low speed mixing conditions helped to maintain this good process in digestors. Furthermore, the energy dissipated by the system at low mixing was only 1/16 as the high mixing condition [26]. In this study, the mixing rate of the impeller was set at 50 rpm (the lowest mixing setup for the BIOFLO biofermentor). The temperature of the AHP biofermentor was kept at 38 °C by using a heating pad wrapped around the vessel.

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