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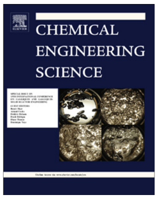

Mixing and liquid-to-gas mass transfer under digester operating conditions

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HIGHLIGHTS

- A dual-impeller stirred tank devoted to dark fermentation was investigated.
- Hydrodynamics, mixing and mass transfer were compared using nine mixer designs.
- Local flow field, vortex formation, k_La and straw suspension were also studied.
- Experimental data were successfully confronted to CFD simulations.
- Finally, the best compromise for mixer design in the digester was defined.

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ABSTRACT

This work deals with the analysis and the optimization of a dual-impeller design in terms of mixing, hydrodynamics, mass transfer properties and power input in a mechanically stirred digester devoted to biohydrogen production through acidogenic fermentation of lignocellulosic waste. Various mixer designs involving Rushton turbines, an Elephant Ear impeller and a marine propeller, were compared. Experimental data were successfully confronted to CFD-based simulations used to reveal the respective roles of impeller type, geometry and clearance. The results showed that the flow pattern was strongly influenced by the off-bottom and inter-impeller clearances, and by the size and type of the lower impeller. Straw suspension was enhanced by a small disk turbine with a low off-bottom clearance and a large inter-impeller clearance that promoted an axial flow circulation together with a small mixing time due to the interaction with the larger turbine used as the upper impeller. Conversely, k_La evolution was weakly dependent on impeller design, position, and rotation speed until a deep vortex formed on the free surface, showing that power input was too weak to enhance liquid-to-gas mass transfer. Finally, the design including an Elephant Ear turbine as the upper impeller and a smaller Rushton turbine as the lower impeller was selected as the best compromise between distributive and dispersive mixing, while the objective of a power input lower than 10 W/m^3 was achieved.

1. Introduction

The growth of global demand for energy, the depletion of fossil fuels, the volatility of energy prices and their respective effects on climate change are among the major global challenges that chemical engineers must face up because these strongly impact the economic, environmental and social sustainability of the process industries. Even though research efforts focused on “green” renewable energy have exploded in the last decade, the development of sustainable solutions is still limited in the field of biofuels for vehicles. Hydrogen often appears as an attractive solution for the future because it can be used directly as a zero-emission fuel in heaters and heat engines, or for producing electricity. Biohydrogen ($BioH_2$) derived from biomass is, therefore, an appealing option that can meet the standards for sustainability. Among the various pathways able to produce hydrogen from biomass, *dark fermentation*, also referred to as *acidogenic fermentation*, is an environment friendly alternative because this process can use agricultural, food and lignocellulosic waste as substrates, which corresponds to a 2nd generation biofuel with a high potential due to the wide availability of

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the resource. BioH_2 production through dark fermentation follows a biological pathway similar to conventional anaerobic digestion, except that methanogenesis is suppressed. Hence, the microbial consortium is less sensitive to shear in the absence of methanogenic *Archaea*, but mixing is generally poor, so as to maintain mechanical power input low in comparison to power production (Lemmer et al., 2013). Mixing must, however, ensure concentration homogenization, solid waste suspension and liquid-to-gas mass transfer for biogas recovery at the same time, as the presence of dead zones can induce local pH gradients and hydrogen supersaturation, so that less biologically-active zones and microenvironments with different microbial communities could arise (Bensmann et al., 2013; Zhang et al., 2013). In dark fermentation, the challenge is to make a trade-off between the need for higher biogas productivity with enhanced BioH_2 yield and selectivity, and the minimization of the capital and operating costs of the pretreatments and digestion steps (Nanqi et al., 2011).

Stirred tank technology is obviously more versatile to achieve good mixing conditions than hydraulic or gas-induced mixing (Trad et al., 2016). Even though anaerobic stirred bioreactors have been disregarded in the literature in comparison to their aerated counterpart, they share many of the same difficulties when the adequate mixing equipment must be selected:

- The culture medium is a complex multiphase and multi-species gas-liquid-solid system.
- Mixing must ensure the nutrient distribution to cells for healthy growth, and enhance the hydrolysis of particulate substrates; endothermic anaerobic processes also require good heat transfer.
- Liquid-to-gas mass transfer must also be enhanced for biogas desorption; in dark fermentation, dissolved hydrogen can reach high supersaturation levels that inhibit the biological processes (Zhang et al., 2013), while a small decrease of pH due to dissolved CO_2 can switch metabolic pathways (Rodríguez et al., 2006).
- Moderate shear and turbulence levels must be applied to prevent cell damage and limit power input, even though these favor heat and mass transfer, which is also a concern in conventional cultures (Garcia-Ochoa et al., 2013; Collignon et al., 2016).

Thus, the same parameters can be used in aerobic and anaerobic processes for impeller design, selection and scale-up: namely, the mixing time t_m, turbulent kinetic energy k, local dissipation rate ε, Kolmogorov length scale l_K, the average or maximum shear rate applied to the cells, the volumetric mass transfer coefficient k_La, the minimum agitation speed for suspension, and the power-per-volume ratio (Junker et al., 1998; Garcia-Ochoa et al., 2013). The same experimental techniques can be applied, such as particle image velocimetry (PIV) or planar laser-induced fluorescence (PLIF) (Aubin et al., 2004; Montante et al., 2007, 2008, 2013; Unadkat et al., 2011), and the same numerical tools are available, such as computational fluid dynamics (CFD) (Delafosse et al., 2009; Yang and Mao, 2014; Trad et al., 2015a; Collignon et al., 2016). Only the values of these parameters must be adapted to the conditions of acidogenic fermentation. With an average hydrogen productivity from starch between 1200 and 2500 L/m^3 day (Kapdan and Kargi, 2006), the maximum energy recovery from BioH_2 (energy density: 120 MJ/kg) lies between 150 and 310 W/m^3. Accounting for the 33–50% efficiency of fuel-to-electricity conversion, the resulting gross power generation (between 50 and 150 W/m^3) must also be decreased by the power consumption of auxiliary equipment. It results that in acidogenic fermentation, power draw for mixing must absolutely be lower than 10 W/m^3. This highlights that power consumption is a key issue, even though hydrogen recovery corresponds to a mass transfer rate lower by a factor between 10 and 60 in comparison to the oxygen transfer rate required by aerobic bioreactors. The drawback is that most of the abundant literature on mixers cannot be applied to digesters because aeration typically involves baffled tanks driven with power input higher than 10^2 W/m^3 and often above 10^3 W/m^3, particularly when high shear radial impellers, such as Rushton turbines, are used (Badino et al., 2001; Montante et al., 2007, 2008; Delafosse et al., 2009). Another drawback is that the literature focuses on baffled tanks, while anaerobic digesters are unbaffled (Montante et al., 2013; Ramírez-Gómez et al., 2015), especially when particulate substrates are processed because they promote the accumulation of solid deposits. Thus, a third major issue is the suspension of the lignocellulosic waste larger than 1 mm, but with a density close to water at low power input. The determination of the just-suspended agitation speed N_{js} of this solid is intrinsically a hard task, first because experimental values are strongly method-dependent (Tamburini et al., 2012). A rapid review of the literature shows that N_{js} strongly depends on the off-bottom clearance C_b/T (Armenante and Nagamine, 1998; Sharma and Shaikh, 2003) Conversely, the dependence on the tank diameter-to-height ratio D/T is weaker; high D/T values were ineffective, forming stagnant zones below 45° pitched blade turbines (Sharma and Shaikh, 2003). More recently, CFD also helped to better define the optimum suspension velocity, for example with the concept of sufficient suspension speed (Tamburini et al., 2012) or an analysis of the spatial distribution of ε (Collignon et al., 2016).

Finally, it emerges that in-depth investigations are necessary to improve our knowledge on mixer selection and design in anaerobic digesters. For example, multi-stage impellers have been barely discussed, despite their lower power-per-impeller consumption, their enhanced mass transfer property and their ability to suspend solids (Gogate et al., 2000; Kiełbus-Rapala and Karcz, 2009). Similarly, impellers typically developed for the gentle mixing of aerobic bioreactors could constitute an alternative. A design that has gained attention in the last decade is the Elephant-Ear turbine (EE). EE is an axial-radial impeller marketed as a *low shear* device (Zhu et al., 2009; Miro and Voll, 2009; Collignon et al., 2010; Bustamante et al., 2013; Trad et al., 2015a), even though this has been a subject of controversy (Simmons et al., 2007). Zhu et al. (2009) advocated that EE operated in the down-pumping mode (EED) improved solid suspension under unaerated conditions, while up-pumping EE (EEU) reduced power requirements under gassed conditions. Their results agreed with Bustamante et al. (2013) who measured k_La. However, Collignon et al. (2010) preferred EED for enhancing at the same time microcarrier suspension and gas-liquid mass transfer. To the best of our knowledge, only Buffo et al. (2016) investigated EE in dual-stage impellers by comparing seven combinations of EEU and EED with Rushton turbines (RT). These authors reported that when EED as the upper impeller was associated with EEU as the lower impeller, good mass transfer properties were maintained, while power draw was minimized and the shear level was 60% lower than with a RT-RT combination.

Therefore, the aim of this paper is to evaluate and compare, both experimentally and numerically, nine dual-impeller configurations in terms of mixing, hydrodynamics, mass transfer properties and power draw under conditions mimicking the acidogenic fermentation of wheat straw for BioH_2 production. EE operated in the down-pumping mode was used as the upper impeller, while a marine propeller and two Rushton turbines with different diameters were operated as the lower impeller; for each of these three designs, three impeller clearances were compared in a relation to of rotation speed using advanced experimental techniques, such as PIV and PLIF, but also flow visualization to investigate straw