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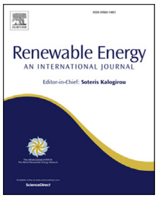

Multiscale mixing analysis and modeling of biohydrogen production by dark fermentation

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ABSTRACT

Hydrogen production by dark fermentation (DF) from wastewater, food waste, and agro-industrial waste combines the advantages to be renewable, sustainable and environmentally friendly. But this attractive process involves a three-phase gas-liquid-solid system highly sensitive to mixing conditions. However, mixing is usually disregarded in the conventional strategies for enhancing biohydrogen productivity, even though H_2 production can be doubled, *e.g.* versus of reactor design (0.6–1.5 mol H_2/mol hexose). The objective of this review paper is, therefore, to highlight the key effects of mixing on biohydrogen production among the abiotic parameters of DF. First, the pros and cons of the different modes of mixing in anaerobic digesters are described. Then, the influence of mixing on DF is discussed using recent data from the literature and theoretical analysis, focusing on the multiphase and multiscale aspects of DF. The methods and tools available to quantify experimentally the role of mixing both at the local and global scales are summarized. The 0-D to 3-D strategies able to implement mixing in fermentation modeling and scale-up procedures are examined. Finally, the perspectives in terms of process intensification and scale-up tools using mixing optimization are discussed with the issues that are still to be solved.

1. Introduction

In the last decade, anaerobic treatments of wastewater effluents and organic waste have regained interest because they present the advantage to combine pollution abatement, volume reduction and waste stabilization with the production of biogas, biofuels and biomass-derived platform molecules for the production of bio-based chemical products and materials [1,2]. Even though anaerobic digestion (AD) is mainly devoted to the production of methane-rich biogas, dark fermentation (DF), *i.e.* the fermentative conversion occurring when methanogenesis is prevented, constitutes an attractive alternative because it is able to produce biohydrogen as a biofuel, together with volatile fatty acids (VFAs) as platform molecules for the production of basic chemicals and polymers [2–6]. Second generation biohydrogen generation by DF from wastewater, food waste, crop residues and agro-industrial waste combines the advantages to be renewable, sustainable and environmentally friendly, as it can face with the issue of mobility of the future and reduce greenhouse gas emissions at the same time [7]. Hydrogen is a clean energy carrier with a high lower heating value (120 MJ/kg), which is higher than any other fossil fuel. This biological process is more attractive than conventional physical/chemical methods for H_2 production because it requires low investment and is well suited for decentralized energy production in regions where biomass or organic waste are available, thus avoiding the expenditure and energy cost of transport. DF process suffers, however, from several major drawbacks, among which a low hydrogen productivity and a low energy efficiency; additional costs are induced because DF must usually be carried out under pH control to avoid pH inhibition [8–10]. This impairs the economic feasibility of DF. Specific challenges have been reviewed recently [4,5,8,10], and process intensification is therefore compulsory, which requires the simultaneous optimization of the biotic and abiotic factors.

Consequently, recent research to enhance the performance of DF has, first, focused on the biotic parameters, such as substrate selection [4–10], substrate pretreatment [10–14], and the selection of more efficient microbial strains coupled to an inoculum

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enrichment strategy (micronutrients, metal ions such as iron …) [10,13–16]. In practice, mixed cultures have to be preferred for economic and technical reasons. Even though mixed cultures usually exhibit lower H_2 yields than pure cultures, a review of the recent literature shows that yield and productivities approach values of pure cultures, such as more than 2 mol H_2/mol glucose eqv. with more than 50% hydrogen in the biogas [17–20]. These values have been reached once the influence of operating conditions had been studied, in particular the effects of temperature, pH, feed mode (batch, fed-batch, continuous), HRT (Hydraulic Retention Time), SRT (Solid Retention Time) and more rarely the influence of hydrogen partial pressure [16–20].

In comparison to other factors, the influence of mixing appears to have been far less extensively studied. However, mixing conditions constitute a key abiotic factor. Actually, mixing is the result at the same time of the time-averaged velocity flow field, the local intensity of turbulence and the energy dissipation rate ε (W/kg) which represents the rate at which mechanical power is dissipated locally and converted in fine into heat. Mixing also determines the local shear stress that the flow applies to microorganisms. As a result, one can conclude that mixing results from the overall flow structure and all the hydrodynamic properties of the flow. However, mixing cannot be directly controlled, but stems from a combination of operating conditions, bioreactor type and design and fluid properties (Fig. 1). The dark fermentation of lignocellulosic waste is sensitive to mixing because it involves a complex three-phase system in which the liquid digestate, biogas and a solid phase composed particulate substrates and sometimes granular sludge are brought into contact. In DF, mixing must ensure at the same time:

- the homogenization of local concentrations, *e.g.* to avoid VFAs and pH inhibitions;
- gas-liquid mass transfer to enhance hydrogen desorption and avoid H_2 inhibition in the bioreactor;
- liquid-solid mass transfer when granular of biofilm sludge is involved.

Heat transfer can also be improved. Actually, the hydrodynamic conditions also play an essential role in the formation, the structure and the metabolism of the microbial community. Consequently, shear should be maintained at a level that does not disturb the biological processes, even though the microbial community involved in hydrogen production has always been reported to be less sensitive to shear than methanogenic bacteria [21]. As a result, the mixing strategy determines not only the choice of the bioreactor, the yield and the productivity, but also the economic viability of the process, as the net power production of the digester must account for the mechanical energy required for mixing (Fig. 1) [21].

Finally, the objective of this review paper is, therefore, to better highlight the role of mixing in DF devoted to biohydrogen production from waste on the basis of the last five year litterature data, to point out the recent experimental and modeling tools able to assess mixing in digesters, to summarize the perspectives in terms of process intensification and scale-up tools based on mixing optimization, but also to discuss the issues that still need to be solved, in particular the coupling between hydrodynamics and biokinetic models.

2. Mixing in anaerobic bioreactors for hydrogen production

The mixing strategy involves, first, the selection of the reactor design, as mixing can be achieved by various technologies. The influence of bioreactor design has been far less studied than biotic parameters, even though it constitutes a key issue of $BioH_2$ production by DF [17–20]. A key point is to avoid or control solid settling, stratification or even flotation of the substrate [22]. Depending on the digester technology, the solid substrate may be:

- suspended mechanically;
- suspended pneumatically, *i.e.* by the gas phase;
- fluidized by the liquid phase.

Actually, there are several ways to classify anaerobic bioreactors. Another classification distinguishes digesters with suspended and immobilized cells, respectively. Immobilized cell bioreactors involve the entrapment of granular or biofilm sludge, which maintains a high microbial density in the reactor.

Mechanical mixing is the most common technology, first at the laboratory scale, but also for methane production from agro-waste in Europe. Mechanically stirred bioreactors involve one or several impellers and propellers of various size and design. These are

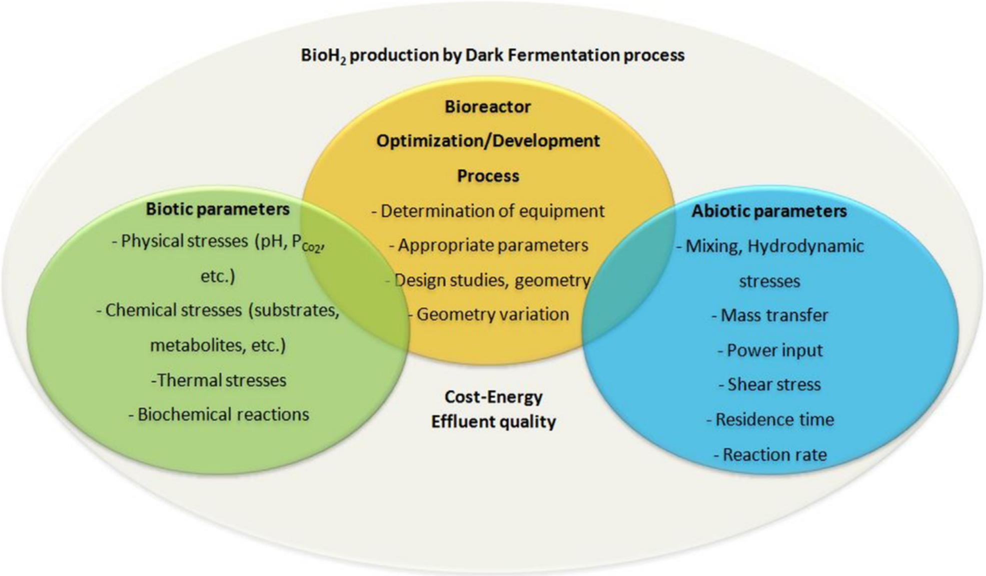

Fig. 1. Interactions between the design, operating and physicochemical and biological parameters in AD for $BioH_2$ production.