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Hydrodynamics simulation of a falling-film microstructured reactor and energetic analysis of the film stability

J.-M. Commenge[a,b,*], H. Semara[a,b], T. Roques-Carmes[a,b]

[a] Université de Lorraine, Laboratoire Réactions et Génie des Procédés, UMR 7274, Nancy F-54000, France
[b] CNRS, Laboratoire Réactions et Génie des Procédés, UMR 7274, Nancy F-54000, France

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ABSTRACT

Microstructured falling-film reactors have demonstrated their relevance for fast and exothermal gas-liquid reactions. Microstructuring enables to stabilize gravity-driven films and to operate with thin films at low flow rates. Unfortunately, the surface of the separating walls remains dry. For plasma-activated reactions, dry patches may create short-circuits and should be avoided: the liquid film should overflow from the channels and wet the whole surface. This film overflow is explored via experimental measurements and a numerical energetic analysis.

A set-up was designed to visualize the film flow with liquids that exhibit various contact angles, enabling to draw flow maps of the stability domain. These results have been compared to a set of CFD simulations for varying flow rates and contact angles: 24 geometries have been designed to consider various positions of the gas-liquid interface, and 78 3D simulations have been performed by combining these geometries and appropriate fluid properties. They enabled to draw numerical flow maps to be compared with experimental maps. This comparison yielded to a good agreement with particularly interesting trends confirmed by geometric considerations. By including energetic aspects related to the various surface energies involved, the frontiers of the numerical flow map confirmed the experimental measurements.

1. Introduction

Microreactors, and more generally microstructured reactors, have demonstrated their large potential to help chemical engineers face the current challenges related to economic, safety and environmental concerns. The specific features of these reactors make them a multifunctional tool in the process intensification toolbox [1]. Their compactness is valuable for mobile, embedded and delocalized applications. Their large surface-to-volume ratios greatly extend the reactors heat and mass transfer capacities and enable to explore new operating conditions and develop innovative chemical pathways [2]. The ability they provide to focus the required energy at the very place where the chemical system needs it is a chance to reduce the overall energy requirement of various physical and chemical unit operations [3–5].

Unfortunately, most of the published works related to microstructured reactors mainly focus on confined flow in microchannels for flow chemistry applications. Only a few of these scientific works keep in mind the opportunity these reactors offer to change and control the ranking of coupled phenomena [6,7]. One particular device took advantage of this possibility: the falling-film microreactor is among the few devices that deliberately take advantage of geometric microstructuring to change the competition between the various forces that govern the flow of a liquid film over a vertical surface. By forcing the liquid to flow through vertical open channels that are grooved on the surface of the plate, this structured falling-film flow selectively modifies the competition between the momentum, gravity and viscosity forces by introducing the impact of surface tension forces. By controlling this additional force through an appropriate structuring of the surface, this reactor enables to maintain stable thin liquid films and avoid their break-up into rivulets.

This possibility of operating very thin stable films led to various applications related to demanding gas-liquid reactions [8–14]. Since thin films also offer improved mass-transfer capacities, the falling-film microreactor has also been tested for physical absorption units [15], Monnier et al. [16–18], reactive absorption [19–25], and micro-distillation [26–30]. For these applications, various properties of these reactors have been characterized experimentally or numerically, such as their residence time distribution [31,32,25], their gas-liquid and gas-liquid-solid mass-transfer performance [33–35,13,36], as well as the flow distribution through the parallel channels [21,22,13]. Several of these studies also highlighted the limitations or drawbacks of this reactor, among which can be cited the partial vaporization of the flowing

* Corresponding author at: Université de Lorraine, Laboratoire Réactions et Génie des Procédés, UMR 7274, Nancy, F-54000, France.
E-mail address: jean-marc.commenge@univ-lorraine.fr (J.-M. Commenge).

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Nomenclature

d_c	Channel depth [m]
E_{tot}	Total specific energy of the liquid per channel and per unit length [J/m]
g	Gravitational acceleration [m/s^2]
L	Channel length [m]
P	Three-dimensional pressure field [Pa]
P_{GL}	Perimeter of the gas-liquid interface over the channel cross-section [m]
P_{SL}	Perimeter of the solid-liquid interface over the channel cross-section [m]
P_{GS}	Perimeter of the gas-solid interface over the channel cross-section [m]
Q	Liquid volume flow rate [m^3/s]
u	Local velocity [m/s]
$\vec{u}$	Three-dimensional velocity flow field [m/s]
w_c	Channel width [m]
w_s	Width of separating walls [m]

Greek letters

δ_L	Equivalent film thickness [m]
θ	Contact angle [°]
θ_{lim}	Limit contact angle [°]
μ	Dynamic viscosity [Pa s]
ρ	Density [kg/m^3]
σ_{GL}	Gas-liquid surface energy [J/m^2]Gas-liquid surface energy [J/m^2]
σ_{SL}	Solid-liquid surface energy [J/m^2]
σ_{GS}	Gas-solid surface energy [J/m^2]

liquid, a restricted range of attainable residence times, the complex upscaling by numbering-up and parallelization, etc. But only a few studies focused on the impact of surface microstructuring on film thickness and stability [37–39,16], and no guidelines are provided to design channel width and depth as a function of liquid properties and flow rates. More generally, the literature strongly lacks methodological guidelines for the selection and design of most-appropriate technologies that could fulfill the requirements of a given synthesis or separation step [40,1].

Such guidelines would have been very useful to design the falling-film microreactor required to operate the reaction considered as the final application envisioned by the present study. This water-treatment application includes a plasma-activated catalytic reaction that enables to eliminate persistent organic pollutants from the water [41,42], [43,44]. The plasma that is generated through a gas layer between an electrode and the catalyst-coated counter-electrode, over which the liquid flows as a film, enables to activate chemical gas species that enhance the catalytic degradation of the pollutants (Fig. 1). For this application, very thin films are favorable since they improve the mass transfer of the active species. Unfortunately, thin films are hardly stable on flat plates and generally break up into rivulets or generate dry patches. When the plasma is generated, these dry solid surfaces induce short-circuits and sparkles that strongly affect the material and catalyst.

To sum it up, microstructured falling-film reactors seem appropriate for this application to operate with thin films, but the film should also cover the separating walls. These objectives may seem contradictory, since the film stabilization is only made possible through the enhancement of surface tension forces that are controlled by the geometry of additional walls and the position of the gas-liquid-solid line, whereas the uniform wetting of the solid surface should annihilate these surface tension forces due to the necessary absence of this gas-liquid-solid line. Therefore, a compromise has to be identified to take advantage of the fact that the separating walls not only enable to control the position of the gas-liquid-solid line, but also play the role of guide to direct the falling liquid in one main direction.

In order to identify the channel geometry and operating conditions that will enable to control the balance between the various forces involved in the film hydrodynamics, a review of previous experimental and numerical works is informative. Various works deal with the experimental characterization of the film thickness in the channels as a function of the fluid properties and channel dimensions [10,38,37,33,39,45,46]. Among these works, [37] discussed the positions of the gas-liquid-solid line in rectangular channels as a function of the channel aspect ratio and contact angle, and proposed regime maps based on AFM micrographs and simulation. [39] experimentally investigated the flow patterns and film break-up in a single channel and correlated the minimum wetting flow rate (to avoid dry patches) to the fluid properties. [46] observed the various flow regimes of a liquid flowing over a plate with parallel channels: they could identify various wetting and break-up regimes and discussed the critical values of the break-up flow rate as a function of the liquid properties and dynamic advancing and receding contact angles.

Several works also report simulation results that should be distinguished with respect to the number of flowing phases considered and to the boundary conditions at the gas-liquid interface. If the hydrodynamic interactions and transfers between the gas and liquid phases are negligible, the simulation may only focus on the liquid phase with a no-shear boundary condition at the fixed gas-liquid interface [31]. If a mass transfer has to be considered between the gas and liquid phases, both domains must be included in the computational domain. The simulations may then differ according to the boundary conditions at the interface. If shear stress remains negligible, the gas-liquid interface can be geometrically fixed and a continuity condition can be written for the species mass transfer [20,18,30,25]. For example, concerning the hydrodynamic behavior in a single channel, [25] presented the various velocity distributions that can be expected in the liquid phase as a function of the flow rate and channel shape. If the hydrodynamic interactions between both phases are not negligible, more complex simulations of the two-phase flow are required including, for example, volume of fluid (VOF) models or level-set methods [47,23].

To explore the balance of the involved forces, an energetic analysis is required to potentially discriminate between similar flow configurations and quantify the most stable configuration. Unfortunately, only few recent works discuss the energetic analysis of a liquid flow over a

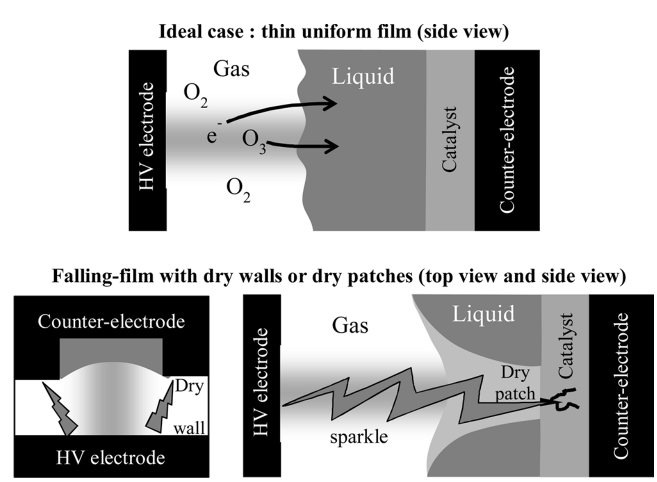

Fig. 1. Comparison of ideal flow conditions (top) and degraded conditions with dry walls or dry patches (bottom) while generating the plasma.