Milli-channel with metal foams under an applied gas–liquid periodic flow: External mass transfer performance and pressure drop

Jean-Noël Tourvieille, Régis Philippe, Claude de Bellefon *

Laboratoire de Génie des Procédés Catalytiques, UMR 5285 CNRS – CPE Lyon, Université de Lyon, 43 Boulevard du 11 novembre 1918, 69100 Villeurbanne, France

HIGHLIGHTS

- Mass transfer & pressure drop are measured in a foam reactor undergoing a G-L flow.
- Correlations are derived for pressure drop, G-L and L-S external mass transfers.
- Confinement brings better mass transfers with negligible impact on energy dissipation.

GRAPHICAL ABSTRACT
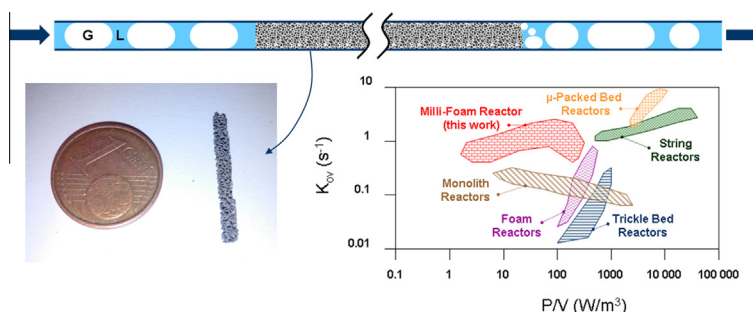
ABSTRACT

Overall external mass transfer and pressure drop performance are addressed for a square 2 mm width channel filled with metal foams as catalyst supports for gas–liquid–solid reactions under an applied pulsing regime. Overall mass transfer coefficients are determined for various foam morphologies and flow conditions by performing α-methylstyrene catalytic hydrogenation over a layer of wash-coated Pd/Al$_2$O$_3$. Values ranging from 0.25 to 1.9 s^{-1} are obtained. A theoretical model and correlations are derived to explain the observed results and agree well with the numerous experimental points. Required energy inputs are estimated by evaluating pressure drops. A predictive correlation is also proposed, as well as a comparison with other correlations from the literature. Finally, the performance of this reactor is compared with other macro- and milli-scale technologies. It has been concluded that foams in confined channels allow mass transfer rates to be increased by up to 50% in comparison with empty channels (monoliths) while maintaining very low energy inputs.

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1. Introduction

Multiphase reactions are a predominant class of reactions for industrial purposes. In the particular case of heterogeneously catalyzed reactions, several reactor technologies have been developed for Fischer Tropsch synthesis, hydrogenation, hydrodesulphurization, oxidation…etc, involving a solid active phase, either mobile or fixed. These multiphase catalytic reactors, considered now as conventional, have been the subject of extensive research because in many cases they present heat or mass transfer limitations [1].

In micro-technology, new reactors have been developed offering a wide range of applications [2]. Due to their high surface-to-volume ratio, more efficient mass and heat transfers are achievable leading to better process control and sometimes to exploration of new process windows. Moreover, the small reacting quantities involved, allied with small characteristic lengths, are suited to safe chemical production of very demanding reactions [3]. So, micro-reactors can provide really useful tools for catalyst screening and even small scale production.

Nomenclature

a_c	intrinsic catalyst activity (mol s^{-1} g$_{pd}^{-1}$)
a_{gl}	gas–liquid specific surface area (m^2 m$_{bed}^{-3}$)
a_{ls}	liquid–solid specific surface area (m^2 m$_{bed}^{-3}$)
Ca	capillary number = $\frac{\mu_L \cdot u_{TP}}{\sigma_L}$
C_{AMS}^o	initial α-methyl styrene concentration (mol m$_{liq}^{-3}$)
$C_{H_2}^{su}$	hydrogen concentration at the catalyst surface (mol m^{-3})
$C_{H_2}^*$	hydrogen concentration at thermodynamic equilibrium (mol m^{-3})
d_{strut}	strut diameter (m)
d_{cell}	cell foam cell diameter (m)
d_{reac}	width of the reactor channel (m)
D_h	hydraulic diameter (m)
$D_E^{H_2}$	effective hydrogen diffusivity in catalyst particle (m^2 s^{-1})
$D_M^{H_2,l}$	molecular diffusivity in the liquid phase (m^2 s^{-1})
E	energy dissipation (W/m^3)
f	friction factor
F_L, F_G	F-functions used in two-phase pressure drop estimation
G	total specific mass flux (kg m^{-2}-s^{-1})
H	reactor height (m)
k_{gl}	gas–liquid mass transfer coefficient (m s^{-1})
K	permeability of the porous material (m^2)
k_{ls}	liquid–solid mass transfer coefficient (m s^{-1})
K_{ov}	mean gas–liquid–solid mass transfer coefficient (s^{-1})
$\bar{r}$	apparent reaction rate (mol s^{-1} g$_{Pd}^{-1}$)
Re$_g$	gas Reynolds number = $\frac{\rho_g \cdot u_{g,s} \cdot d_{cell}}{\mu_g}$
Re$_l$	liquid Reynolds number = $\frac{\rho_l \cdot u_{l,s} \cdot d_{cell}}{\mu_l}$
Sc$_l$	liquid Schmidt number = $\frac{\mu_l}{\rho_l \cdot D_M^{H_2,l}}$
Sh$_{gl}$	gas–liquid Sherwood number = $\frac{k_{gl} \cdot d_{cell}}{D_M^{H_2,l}}$
Sh$_{ls}$	liquid–solid Sherwood number = $\frac{k_{ls} \cdot d_{cell}}{D_M^{H_2,l}}$
S_p	specific surface area (m^3 m$_{foam}^{-3}$)
T	temperature (K)
$u_{g,s}$	gas superficial velocity (m s^{-1})
$u_{l,s}$	liquid superficial velocity (m s^{-1})
u_{TP}	two phase flow mean velocity (in Taylor flow) (m s^{-1})
V	volume (m^3)
We	Weber Number = $\frac{\rho_l \cdot u_{l,s}^2 \cdot d_{cell}}{\sigma_L}$
x	gas mass quality

Greek letters

δ_{cat}	catalyst thickness (μm)
$\Delta P/H$	linear pressure drop (kPa m^{-1})
ε	porosity (m^3 m$_{packing}^{-3}$)
ε_{cat}	corrected porosity accounting for catalyst coating (m^3 m$_{packing}^{-3}$)
η	effectiveness factor (accounting for internal mass transfer resistances)
μ	dynamic viscosity (Pa s)
ρ	density (kg m^{-3})
σ	surface tension (N m)
φ	biphasic multiplier in pressure drop correlations
φ	Thiele modulus in Eq. (13)
X_{AMS}	α-methylstyrene conversion
X_{vv}	Martinelli parameter

While these reactors are easy to use for single phase or fluid–solid reactions, their use for gas–liquid–solid catalytic reactions has been the subject of extended questioning and recent works. Wall-coated microreactors ([4–6] present interesting results for heat and mass transfer with short diffusion lengths and good contact between phases but raise the question of catalyst rejuvenation which may be difficult to handle without changing the whole reactor. A good alternative is to fill a micro- or a milli-channel with small particles to obtain a mini packed bed [7–11]. Overall gas–liquid-solid mass transfer rates up to 15 s^{-1} have been measured and reported [7]. However, undesirable pressure drops are likely to be with such a dense packing for industrial applications.

To cope with this difficulty, de Loos et al. [12] proposed the use of removable micro pillared plates which can be coated with catalyst, inserted in a rectangular micro-channel and fed with a Taylor flow. Their study showed that under certain conditions, the Taylor flow regime is maintained along the whole reactor length. However, such pillared structures can be made only through high precision micro-machining which could result in a poor diffusion of this technology. Moreover these objects do not appear realistic for large production capacities.

Open-cell foams (OCF) may represent a very good tradeoff between the spaced pillars and the packed-particles approaches. They are easy to fabricate on a large scale, commercially available in many materials (metals, ceramic, …) which have already been investigated as packing in fixed-bed type applications of large production capacity and offer rather low pressure drop but benefit from short characteristic dimensions and thus potentially high transfer capabilities. With their high void fraction (from 0.60 to 0.97), good thermal performance and large external specific surface area (up to 10,000 m^2/m^3), open cell foams have demonstrated

their interest as heat exchangers, internal mixers and as catalyst supports [13].

For three phase systems, most studies investigated G-L hydrodynamics, pressure drop and mass transfer characteristics in macro-scale reactors (suitable for large production capacities) in different configurations [14–19]. However, no studies have been devoted to the use of solid foam as catalyst support in a gas–liquid–solid milli-reactor. At this scale, the gas–liquid flow is affected by the proximity of the walls which confine it. This confinement forces the gas–liquid flows to go through the packing rather than taking unconstrained paths (case of reactors where walls are far away from each other). In their works, Liu et al. [20] approach this concept for Fischer–Tropsch synthesis, but they operated their reactor in a gas–solid configuration, at a scale larger than millimetric (2 cm). Another team worked on monophasic flow, Hutter et al. [21] performed PIV and LIF measurements in a 7 mm inner diameter cylindrical reactor used as a static mixer. They concluded that mixing properties induced by foam are enhanced compared to an empty channel. This can be explained by their pseudo-random structure which can induce more perturbations than fully organized systems, thus leading to better mixing properties. Combining these interesting properties with a gas–liquid two-phase flow through the foam packing and constrained by the reactor walls may lead to enhanced mass and heat transfers. Moreover, such foam-filled millimetric channels present an unusually large ratio of wall area per reactor volume. This property could for example be useful in the design of multichannel compact heat exchanger reactors including foam internals.

This work reports the use of open cell metallic foams for the first time as low pressure drop supports for catalyst insertion in a milli-metric square-cross section channel, where a preformed gas–liquid