Chemical Engineering Journal 382 (2020) 122801

Contents lists available at ScienceDirect

Chemical Engineering Journal

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

Short communication

Packed foams for the intensification of catalytic processes: assessment of packing efficiency and pressure drop using a combined experimental and numerical approach

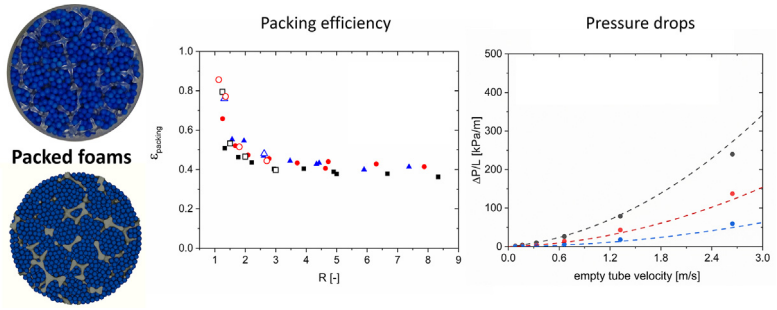
Matteo Ambrosetti[1], Mauro Bracconi[1], Matteo Maestri, Gianpiero Groppi, Enrico Tronconi*

Laboratory of Catalysis and Catalytic Processes, Dipartimento di Energia, Politecnico di Milano via La Masa 34, 20156 Milano, Italy

HIGHLIGHTS

- Experimental and numerical analysis of the packing efficiency in open-cell foams.
- Quantification and modeling of pressure drop in packed foams.
- Conductive packed foams enable process intensification.

GRAPHICAL ABSTRACT



ARTICLE INFO

Keywords:
Packed foams
Packing efficiency
Pressure drop
Process intensification

ABSTRACT

Thermally conductive packed foams have been proposed as an effective solution for the intensification of non-adiabatic catalytic processes in tubular reactors where high heat transfer rates and large catalyst inventories are necessary. Some of the open issues of this innovative solution for its scale-up to industrial process are the packing efficiency and the pressure drop. In this work, these aspects were addressed by performing both experimental activities on 3D printed foams and simulations on packed foam structures. The packing efficiency was studied by considering different spherical pellets and foam samples. The ratio between the foam window and the pellet diameter, R, was identified as the governing parameter: only pellets smaller than the window size can be packed inside the cavities of open cell foams. The packing efficiency increases with R, reaching the same asymptotic value of random packing in a tube at $R > 5$; for R less than 1.3 the porosity exceeds 50% and local channeling may be present. Due to commercial foam specifications, this limits the application of packed foams to processes where pellets smaller than 2 mm are employed. Pressure drop in packed foams was studied as well both by experimental tests and by numerical simulations. Despite the presence of the foam structure, pressure drops in packed foams are comparable or lower than the pressure drops in packed beds with the same pellet diameter due to the increase of the porosity inside the system. An Ergun like correlation corrected by the overall void fraction and the total wetted surface is able to describe the pressure drop in these systems with reasonable accuracy.

* Corresponding author.
E-mail address: enrico.tronconi@polimi.it (E. Tronconi).
[1] These authors contributed equally to the work.

https://doi.org/10.1016/j.cej.2019.122801
Received 2 July 2019; Received in revised form 6 September 2019; Accepted 10 September 2019
Available online 11 September 2019

Notation

d_c	foam cell diameter [m]
d_p	diameter of the pellets [m]
d_t	tube diameter [m]
d_{window}	foam window (pore) diameter [m]
ε_{foam}	void fraction of the foam [–]
$\varepsilon_{packing}$	void fraction of the packing [–]
ε_{PB}	void fraction of the packed bed [–]
ε_{tot}	total porosity of the system
L	length of the reactor [m]
μ	gas viscosity [Pa s]
ΔP	pressure drops [Pa]
ρ	gas density [kg/m^3]
ρ_{pellet}	pellet density [g/cm^3]
R	ratio between foam window diameter and pellet diameter $R = \frac{d_{window}}{d_p}$ [–]
Re_p	pellet Reynolds number $Re_p = \frac{\rho u d_p}{\mu}$ [–]
$S_{v,foam}$	foam specific surface area [m^{-1}]
$S_{v,packing}$	packing specific surface area [m^{-1}]
$S_{v,tot}$	total specific surface area [m^{-1}]
u	superficial velocity [m s^{-1}]
$V_{pellets}$	volume of loaded pellets [m^3]

1. Introduction

Process intensification is one of the most important research areas in modern chemical engineering, being a key enabler for the profitability of remote energy sources or for increasing the efficiency and effectiveness of existing processes [1].

Many catalytic applications are run in conventional multi-tubular reactors filled with randomly packed pellets, which enable high catalyst inventory. However, as a drawback, the performances and the operability windows of some strongly exo- or endo-thermic processes are limited by the poor heat transfer [2]. Different structured reactor technologies have been proposed to promote heat transfer in reactors for processes where temperature control is critical for both safety and productivity. In the last decades, studies of innovative reactor internals such as conductive honeycombs [3], fibers [4], stacked microchannels [5], open-cell foams [6] and 3D printed supports [7] were reported in the literature. However, conventional catalytic activation methodologies for structured reactors (i.e. washcoating [8]) does not allow catalyst loading in excess of 20% of the reactor volume, which adversely affects the application of these technologies in kinetically controlled processes.

Recently, a combination of packed beds and structured reactors was proposed, based on filling the open cavities or cells of conductive structured support with catalyst pellets to combine the high catalyst inventory of packed bed reactors with the increased heat transfer associated with the presence of a conductive structured internal. In particular, packed foams were investigated by pure heat transfer tests [9] showing that the characteristic heat transfer mechanisms of packed beds and foams work in parallel and this results in a synergetic effect on the overall heat transfer coefficient. This concept was successfully demonstrated in strongly exo-thermal and endo-thermal catalytic reactions like the Fischer-Tropsch synthesis [10] and the Methane Steam Reforming [11], resulting in a remarkable increase of the productivity with respect to conventional packed bed reactors in the same operative conditions, proving that the packed foam concept is a potential enabling technology for scaling down the reactor size in view of the exploitation of small methane resources.

Some of the key aspects for the application of packed foams, however, were not yet addressed. In this work, we investigate the packing density inside the structures in order to compare packed foams and packed beds in terms of catalyst inventory. Furthermore, pressure drop are crucial information for their potential use in industrial reactors to reduce the compression costs and to satisfy the constraints of many processes [12]. The tradeoff between the catalyst inventory, pressure drop and the increase of the heat transfer associated with this technology will determine the potential success of the envisioned solution. These aspects are herein addressed both experimentally and computationally by analyzing packing porosity and pressure drop of different combinations of foams and pellets. Discrete Element Method (DEM) approach [13,14] was applied in parallel to perform a virtual packing of foams structures [15], to cross-validate the experiments, highlight local packing efficiency and provide the numerical domain for CFD simulations aimed at gaining insight in the local transport properties of these systems.

2. Materials and methods

2.1. Evaluation of the packing density

2.1.1. Experimental tests

Different sets of foams and spherical particles were coupled for the evaluation of the packing density (Tables 1 and 2). Commercial spherical glass beads from Sigma Aldrich and γ-Al_2O_3 spherical pellets from Sasol were characterized by determining the particle density and the average particle size by granulometry. Virtual foam samples were generated by a virtual reconstruction methodology [15] and manufactured with a SLA 3D printer FormLabs 2.0 in Clear resin and characterized as described in [16]. The virtually-reconstructed foams exhibit a representative geometry in terms of topological and geometrical properties of real foams [15]. The aim of the packing tests was the determination of the packing density ($\varepsilon_{packing}$) in the foam cavities, which is defined as:

$$\varepsilon_{packing} = 1 - \frac{V_{pellets}}{V_{foam,empty}} = 1 - \frac{V_{pellets}}{\varepsilon_{foam} V_{total}} \quad (1)$$

The measured value of ε_{foam} (Table 1) was used for the calculation. Foam samples were fitted in a sample holder and then the particles were poured on the top surface of the foam sample. To mimic the industrial practice of packed beds loading [17], the sample was vibrated in order to increase the volumetric loading. As an internal verification, glass beads with nominal size d_p equal to 1 and 2 mm were packed in a graduated cylinder (d_t = 36 mm). We determined the void fraction of the bed pouring water up to the free surface of the cylinder and weighting. As expected, for large d_t/d_p the asymptotic voidage of 0.38 was recorded. Then, we used the packing test to measure the density of the particles, using the relation with the porosity.

Table 1
Geometrical properties of open-cell foams used for packing tests.

Foam sample	d_c [mm]	ε_{foam} [–]	$S_{v,foam}$ [m^{-1}]	d_{window} [mm]	Real ε_{foam} [–]
A	3.00	0.70	985	1.328	0.671
B	4.00	0.70	738	1.771	0.681
C	5.00	0.70	591	2.213	0.687
D	3.00	0.80	935	1.419	0.779
E	4.00	0.80	701	1.892	0.783
F	5.00	0.80	561	2.365	0.792
G	3.00	0.90	763	1.514	0.884
H	4.00	0.90	572	2.019	0.891
I	5.00	0.90	458	2.524	0.897

Table 2
Geometrical properties of pellets used for packing tests.

Pellet sample	*Material*	Nominal d_p [mm]	ρ_{pellet} [g/cm^3]	Measured d_p [mm]
j	Al_2O_3	0.6	1.14	0.64
k	Al_2O_3	0.8	1.11	0.92
l	Al_2O_3	1	1.23	1.04
m	Al_2O_3	1.2	1.08	1.18
n	Al_2O_3	1.6	1.08	1.70
o	Glass	0.5	2.45	0.56
p	Glass	1	2.45	1.13
q	Glass	2	2.11	1.83

2.1.2. Numerical simulations

The numerical random packing was obtained by means of Discrete Element Method (DEM) [13] simulations. Spherical monodisperse particles were injected from the top of the computational domain and fell due to the action of the gravity force. In DEM, a force balance is drawn for each particle considering the gravity and the interactions between the particles and the solid walls, i.e. the tubular container and the foam surface. The DEM simulation stops when the steady-state conditions of the particle within the foam are reached. This occurs when the velocity of each sphere approaches zero. In this work, the DEM algorithm reported in [14] was employed.

2.2. Evaluation of pressure drop

2.2.1. Experimental test

Pressure drop in packed beds and in packed foams were evaluated experimentally using a tubular reactor with two pressure ports [16]. To contain the bed within 8 cm length, two thin disks of FeCrAl foams with 60 PPI and two spacers were placed at the inlet and at the outlet of the system. In the case of packed foams, 4 cylinders of 3D printed foams were loaded and then the particles were poured and shaken to improve the packing regularity. The reactor was connected to ¼" gas line and a mass-flow controller (Brooks 5851) was used to regulate the flow rate to perform measurements for superficial velocities in the range of 0.1–2 m s^{-1} corresponding to Re_p in the range 2–300. Tests were performed with air at 25 °C and the outlet pressure of the system was atmospheric. Pressure drop was measured with a differential manometer connected to two pressure ports located at the inlet and at the outlet of the loading zone (Comark 9553 for $\Delta P > 10^4$ Pa, Testo 410 for ΔP in the range 10^2–10^4 Pa, Techmark 635 for ΔP in the range 10–100 Pa).

2.2.2. Numerical simulations

Computational Fluid Dynamics (CFD) was employed to calculate the pressure drop in packed foams. The simulations were carried out by means of simpleFoam, a steady state incompressible laminar and turbulent solver adopting the SIMPLE algorithm part of the OpenFOAM framework. In this work, air at ambient conditions (298 K and 1 atm) is employed as working fluid, modeled as an incompressible perfect gas under isothermal conditions. Several flowrates have been tested to span a significant range of operating conditions corresponding to superficial velocity between 0.06 and 2.6 m/s. corresponding to Re_p in the range 2–300.

The packed foam samples investigated in the experimental campaign consisted of a large number of foams cells with spherical particles packed in the void spaces. The size of the samples was usually large both in the stream-wise in the transversal direction making the direct reproduction of the entire system unfeasible due to the huge computational burden. To overcome such limitations, we considered periodic and symmetry boundary conditions along the stream-wise and transversal directions, respectively. Along these lines, it was possible to reduce the computational cost by investigating a reduced but representative portion of the entire foam. The envisioned approach had already been successfully employed for the simulation of pressure drop in open-cell foams [15,16] where additional details can be found. Mesh generation procedure and characteristics are reported in the Supplementary material along with details on the adopted procedure for the numerical solution and boundary conditions.

3. Results and discussion

3.1. Packing density

Two examples of packed foams obtained by means of experimental and numerical methods are shown in Fig. 1.

The packing porosities evaluated for different combinations of foams and particles are reported in Fig. 2 for real and virtual experiments. The packing porosity is plotted against the ratio R of the foam window (e.g. pore) to the pellet diameter, being the d_{window} evaluated from d_{cell} and ε_{foam} accounting for the geometrical model reported in [18]:

$$R = \frac{d_{window}}{d_p} \tag{2}$$

A remarkable agreement is observed between the two methodologies, being the deviations less than 1% in the range of main industrial

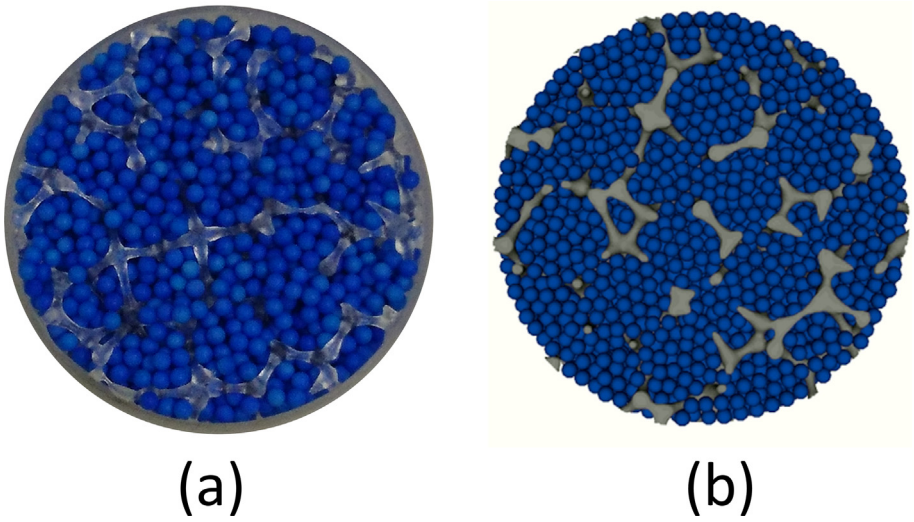


Fig. 1. Images of a real packed foam (a) and of virtually generated packed foam (b).

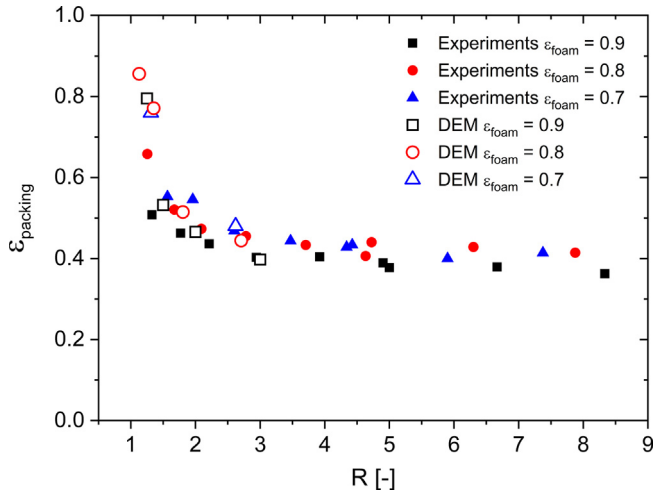


Fig. 2. Packing porosity vs window to pellet size ratio.

interest (R > 2). The trend in Fig. 2 recalls the relation between porosity and tube-to-particle-ratio in packed tubes. Two asymptotic behaviors are present. On one hand, an asymptotic porosity of 0.38 is reached for large values of R (> 5). On the other hand, the packing porosity has a steep increase when R approaches 1. A large region for R between 1.5 and 4 corresponds to a packing porosity in the range 0.5–0.4. Reduction of > 30% of the catalyst inventory is manifest for R less than 1.3. The experiments also evidenced a secondary effect of the porosity of the foams, the packing efficiency slightly increasing with ε_{foam}, likely due to the increase of the surface area of the foam on decreasing the structure porosity.

The numerical experiments provided a deeper insight into the local packing efficiency, being able to look at slices of the bed in different axial positions. When the porosity is higher than 0.5, local portions of the reactor have no packing, as shown in Fig. 3. This would likely result in gas bypass and, in the case of non-adiabatic applications, in local temperature inhomogeneity. Operations in that region are therefore not recommended.

3.2. Pressure drop

Different tests were performed for the assessment of pressure drop in packed foam systems. As an example, pressure drop in a packed bed and in two packed foams with the same pellets are shown for two different pellet sizes in Fig. 4. In the case of small pellets (Fig. 4 (a)), for a foam with an high void fraction (ε_{foam} = 0.9) the pressure drop is comparable with that of the corresponding packed bed, whereas for a foam with a low void fraction (ε_{foam} = 0.8) pressure drop is significantly higher, since the overall void fraction of the system is lower than in packed beds. In the case of bigger particles (Fig. 4(b)), packing densities ($\varepsilon_{packing}$) in the range of 0.48–0.52, corresponding to total porosities (ε_{tot}) in the range 0.42–0.43 were obtained in packed foams, which results in lower pressure drop by about 40% than in the corresponding packed bed. This opens interesting opportunities in the reactor design for processes that suffer from pressure drop limitations, allowing, for example, increased superficial velocity and/or tube length.

CFD simulations allowed to carry out a parametric analysis of the effect of the geometrical properties. Fig. 5(a) shows the effect of different pellet size packed in the same open-cell foams. On increasing the pellet diameter, the packing porosity increases resulting in lower resistance with respect to the fluid flow. Fig. 5(b) shows the effect of different foam porosities at constant pellet size. By decreasing the foam porosity, the packing efficiency is reduced resulting in higher packing void fraction. Hence, the pressure drop decreases despite the lower void fraction of the open-cell foam. We have found that an Ergun like correlation [19] (Eq. (3)) is able to describe the observed behavior when using the total porosity, ε_{tot} and the total specific surface area, $S_{v,tot}$ as the geometrical parameters. The total porosity of the system can be computed from the definition of porosity for both the foam and the packing (Eq. (4)). The total wetted surface area is the sum of the exposed surface of the foam and of the packing (Eq. (5)). For the evaluation of the specific surface area of the foam ($S_{v,foam}$), the geometrical model proposed in [18] was adopted. The evaluation of the surface area of the packing is computed according to Eq. (6), through the classical equations used for the surface area of a packed bed of spheres, corrected for the porosity of the foam.

$$\frac{\Delta P}{L} = 4.17\frac{(S_{v,tot})^2}{\varepsilon_{tot}^3}\mu u + 0.292\frac{(S_{v,tot})}{\varepsilon_{tot}^3}\rho u^2 \tag{3}$$

$$\varepsilon_{tot} = \frac{V_{void}}{V_{tot}} = \frac{V_{void}}{V_{foam,empty}}\frac{V_{foam,empty}}{V_{tot}} = \varepsilon_{Packing}\varepsilon_{foam} \tag{4}$$

$$S_{v,tot} = S_{v,foam} + S_{v,Packing} \tag{5}$$

$$S_{v,packing} = \frac{6(1-\varepsilon_{packing})}{d_p}\varepsilon_{foam} \tag{6}$$

Parity plots of the correlation estimates against experimental and CFD simulation results are shown respectively in Fig. 6. A satisfactory agreement is observed, being the mean average percentage error (MAPE) equal to 15%, within the typical deviations usually present in pressure drops measurements of packed bed reactors [20]. The correlation can be used for the estimation of pressure drop for Re_p in the range 2–300.

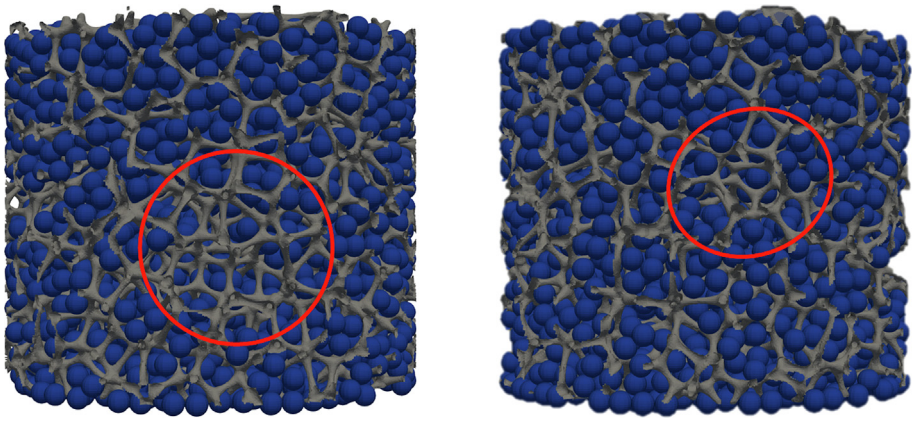


Fig. 3. Packing inhomogeneity for R = 1.3.

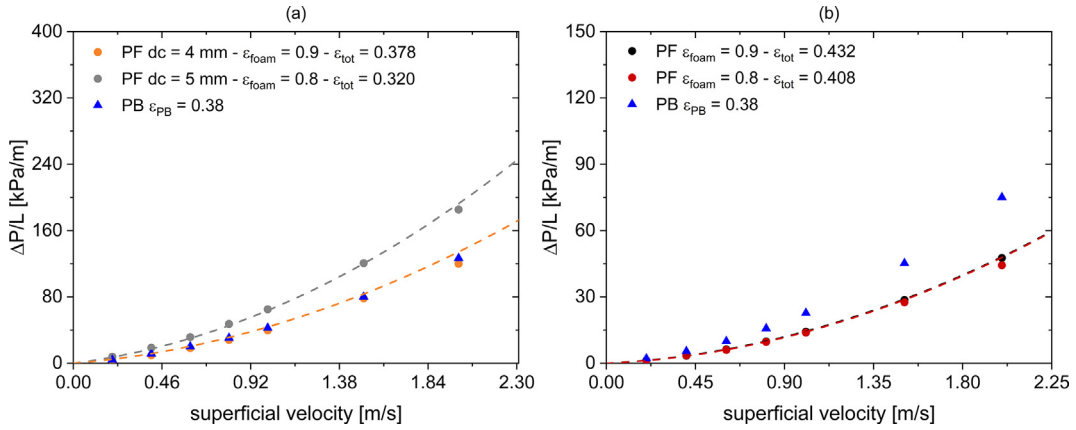


Fig. 4. Experimental pressure drop in packed beds and packed foams (symbols = experiments, dashed lines = correlation, Eqs. (3)–(6)), (a) 1 mm particles, (b) 1.6 mm particles and foam cell size 5 mm.

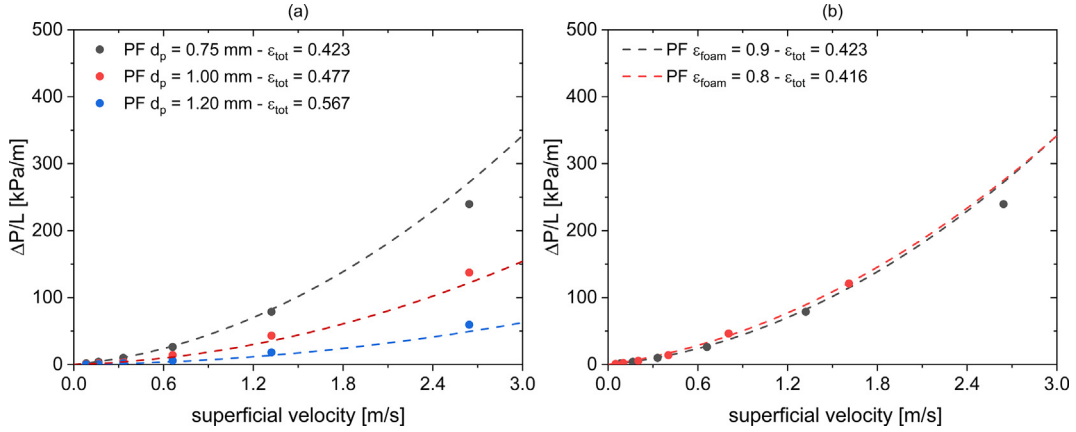


Fig. 5. CFD pressure drop in packed foams (symbols = experiments, dashed lines = correlation, Eqs. (3)–(6)), (a) effect of particle size (d_c = 3 mm, ε_{foam} = 0.9), (b) effect of foam porosity (d_c = 3 mm; d_p = 0.75 mm).

4. Conclusions

The aim of this work was to analyze in detail two aspects of the novel packed foam concept. Our analysis reveals that the packing efficiency is a function of the pore to pellet size ratio. For large values of the ratio, the packing efficiency is close to the asymptotic value in packed tubes, while there is a steep increase of the packing porosity when the ratio approaches one. Pressure drop in packed foams can be estimated by an Ergun-like correlation accounting for the total porosity and the total wetted area of the system (pellet and foam). These aspects can be now considered for the design of catalytic reactors based on this technology, accounting for the tradeoff between the reduction of the catalyst inventory, increase or decrease of the pressure drop – depending on the selected combination of pellet and foam sizes – and the heat transfer enhancement previously reported for this structured reactor configuration.

Acknowledgments

The research leading to these results has received funding from the European Research Council under the European Union's Horizon 2020 research and innovation program (Grant Agreement no. 694910/

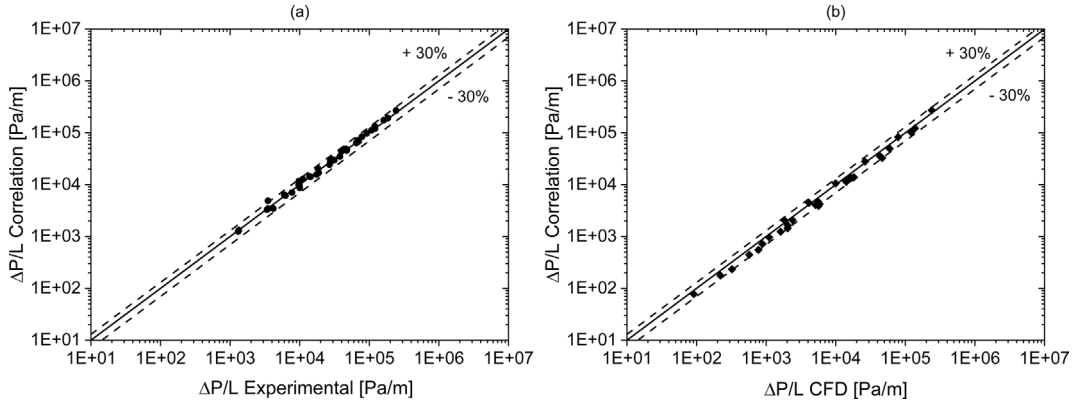


Fig. 6. Parity plot of experimental data vs proposed correlation (a), CFD data vs proposed correlation (b).

INTENT). Computational time from CINECA is gratefully acknowledged. The authors thank Sasol for providing the pellet samples.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.cej.2019.122801.

References

[1] A.I. Stankiewicz, J.A. Moulijn, Process intensification: Transforming chemical engineering, Chem. Eng. Prog. 96 (2000) 22–33.
[2] E. Tronconi, G. Groppi, C.G. Visconti, Structured catalysts for non-adiabatic applications, Curr. Opin. Chem. Eng. 5 (2014) 55–67, https://doi.org/10.1016/j.coche.2014.04.003.
[3] T. Boger, A.K. Heibel, Heat transfer in conductive monolith structures, Chem. Eng. Sci. 60 (2005) 1823–1835, https://doi.org/10.1016/j.ces.2004.11.031.
[4] S. Danaci, L. Protasova, J. Lefevere, L. Bedel, R. Guilet, P. Marty, Efficient CO_2 methanation over Ni/Al_2O_3 coated structured catalysts, Catal. Today. 273 (2016) 234–243, https://doi.org/10.1016/j.cattod.2016.04.019.
[5] R.J. Kee, C. Karakaya, H. Zhu, Process intensification in the catalytic conversion of natural gas to fuels and chemicals, Proc. Combust. Inst. 36 (2017) 51–76, https://doi.org/10.1016/j.proci.2016.06.014.
[6] I. Gräf, G. Ladenburger, B. Kraushaar-Czarnetzki, Heat transport in catalytic sponge packings in the presence of an exothermal reaction: characterization by 2D modeling of experiments, Chem. Eng. J. 287 (2016) 425–435, https://doi.org/10.1016/j.cej.2015.11.042.
[7] C. Busse, H. Freund, W. Schwieger, Intensification of heat transfer in catalytic reactors by additively manufactured periodic open cellular structures (POCS), Chem. Eng. Process. Process Intensif. 124 (2018) 199–214, https://doi.org/10.1016/j.cep.2018.01.023.
[8] D. Merino, O. Sanz, M. Montes, Effect of the thermal conductivity and catalyst layer thickness on the Fischer-Tropsch synthesis selectivity using structured catalysts, Chem. Eng. J. 327 (2017) 1033–1042, https://doi.org/10.1016/J.CEJ.2017.07.003.
[9] C.G. Visconti, G. Groppi, E. Tronconi, Highly conductive “packed foams”: a new concept for the intensification of strongly endo- and exo-thermic catalytic processes in compact tubular reactors, Catal. Today. 273 (2016) 178–186, https://doi.org/10.1016/j.cattod.2016.02.060.
[10] L. Fratalocchi, C.G. Visconti, G. Groppi, L. Lietti, E. Tronconi, Intensifying heat transfer in Fischer-Tropsch tubular reactors through the adoption of conductive packed foams, Chem. Eng. J. 349 (2018) 829–837, https://doi.org/10.1016/j.cej.2018.05.108.
[11] R. Balzarotti, A. Beretta, G. Groppi, E. Tronconi, Copper foams for the intensification of methane steam reforming, React. Chem. Eng. 4 (2019) 1387–1392, https://doi.org/10.1039/c9re00125e.
[12] G. Bellussi, M. Bohnet, J. Bus, K. Drautz, H. Greim, K.-P. Jackel, et al., Ullmann’s Encyclopedia of Industrial Chemistry, seventh ed., 2011.
[13] P.A. Cundall, O.D.L. Strack, Discussion: a discrete numerical model for granular assemblies, Géotechnique 30 (1980) 331–336, https://doi.org/10.1680/geot.1980.30.3.331.
[14] R. Uglietti, M. Bracconi, M. Maestri, Coupling CFD-DEM and microkinetic modeling of surface chemistry for the simulation of catalytic fluidized systems, React. Chem. Eng. 3 (2018) 527–539, https://doi.org/10.1039/c8re00050f.
[15] M. Bracconi, M. Ambrosetti, M. Maestri, G. Groppi, E. Tronconi, A systematic procedure for the virtual reconstruction of open-cell foams, Chem. Eng. J. 315 (2017) 608–620, https://doi.org/10.1016/j.cej.2017.01.069.
[16] M. Bracconi, M. Ambrosetti, O. Okafor, V. Sans, X. Zhang, X. Ou, C.P. Da Fonte, X. Fan, M. Maestri, G. Groppi, E. Tronconi, Investigation of pressure drop in 3D replicated open-cell foams: coupling CFD with experimental data on additively manufactured foams, Chem. Eng. J. (2018), https://doi.org/10.1016/j.cej.2018.10.060.
[17] S. Afandizadeh, E.A. Foumeny, Design of packed bed reactors: guides to catalyst shape, size, and loading selection, Appl. Therm. Eng. 21 (2001) 669–682.
[18] M. Ambrosetti, M. Bracconi, G. Groppi, E. Tronconi, Analytical geometrical model of open cell foams with detailed description of strut-node intersection, Chem.-Ing.-Tech. 89 (2017) 915–922, https://doi.org/10.1002/cite.201600173.
[19] S. Ergun, A.A. Orning, Fluid flow through randomly packed columns and fluidized beds, Ind. Eng. Chem. 41 (1949) 1179–1184, https://doi.org/10.1021/ie50474a011.
[20] E. Erdim, Ö. Akgiray, I. Demir, A revisit of pressure drop-flow rate correlations for packed beds of spheres, Powder Technol. 283 (2015) 488–504, https://doi.org/10.1016/j.powtec.2015.06.017.